

The Evidence of the Once-Forbidden Spectra for the Law of β -Decay

H. M. MAHMOUD AND E. J. KONOPINSKI
Indiana University, Bloomington, Indiana

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A series of empirical arguments has developed which leave two alternatives for the law of β -interaction: an *STP* (scalar+tensor pseudoscalar) or a *VTP* ($V \equiv$ vector) combination. We offer a new argument, based on an interpretation of the reported "allowed" shapes of once-forbidden spectra, which favors the *STP* over the *VTP* form. This new development may be important because it is a possible last step in arriving at a unique law of β -decay (i.e., all three components of the *STP* form would become both admissible and necessary).

A critical stage in the new argument is the explanation of the "allowed" shapes. The theory may yield such shapes if the Coulomb potential energy at the nucleus is sufficiently larger than the kinetic energy with which electron and neutrino are emitted. That condition happens to be best fulfilled just for the

cases in which the "allowed" shapes are the most accurately observed. However, the dominance of the Coulomb effect cannot be a sufficient condition except with the *STP* or *VA* forms of β -interaction. Other forms make deviations from the allowed shape possible even when the Coulomb energy is supposed indefinitely large.

The last type of deviation has the same origin as the well-known Fierz interference in allowed spectra. The absence of the latter effect has been widely quoted but apparently never heretofore examined in detail. It is important in one of the series of arguments mentioned above. Hence, we investigate the empirical limits on Fierz-type deviations not only in once-forbidden but also in allowed spectra.

1. INTRODUCTION

THE Fermi theory of β -decay is not completely definite, even though it has dealt successfully with much that is known concerning β -processes. The definite part of the theory consists of criteria which are equally well satisfied by several specific formulations. The alternatives can be exhaustively enumerated in more ways than one. The customary way leads to a β -interaction expressed as an arbitrary linear combination of five "forms"; the scalar form, symbolized by *S*; the polar vector, *V*; the tensor, *T*; the axial vector, *A*; and the pseudoscalar, *P*. There has been a continuous effort to find and interpret experimental evidence that some definite combination of the five forms is the correct one.

The following points are currently regarded as having been established by the evidence:

I. The *T* or *A* forms must be part of the correct law of β -interaction. The main evidence is of two kinds. Only the *T*, or the *A*, form yields Gamow-Teller selection rules, which are needed for understanding the short lives of many β -transitions with a unit spin change. Only these two forms yield the "unique" spectrum shapes, found for n -times forbidden transitions in which the spin changes by $n+1$ units.

II. The *S* or the *V* form must also be included. The chief evidence for this is the short life found for β -transitions between nuclear states of zero spin, during the decay¹ of C^{10} and O^{14} . Forms other than the *S* or *V* lead to much smaller decay probabilities for these cases, relative to other transitions.

III. The *P* form is needed to explain the singular RaE spectrum, according to the analysis of Petschek and Marshak.² Actually, the *P* form is needed in combination with the *T* for this.

Each of the above arguments requires the inclusion of some component form without precluding the presence of other forms. The following argument seems to be the only generally recognized empirical one which tends to eliminate some of the possible combinations.

IV. It is usually regarded as evident that not both the *T* and *A* forms, nor both the *S* and *V* forms, can be parts of the β -coupling. If they are, then the spectra of allowed β -emission should be expected to deviate from the "allowed" (essentially statistical³) shape, through the operation of the so-called "Fierz interference." However, no unquestionable deviations have ever been found.⁴

Perhaps the weakest of the above arguments is **III**; points for and against it are put forward in Sec. 5. However, even if one accepts this one together with all the other arguments, they still do not lead to a unique law of β -interaction. They are equally consistent with either an *STP* or a *VTP* combination. We shall introduce here a possible method for deciding between those final two possibilities. Like **III**, and perhaps also **IV**, our new argument (**V**) needs further confirmation before it will become completely acceptable, but at present it points to the *STP* combination as the correct one. This development would be important because one will have finally arrived at a unique law of β -decay. The law would be unique in the sense that each of its component forms would be necessary, and also the only ones admissible. Formerly, it was not entirely clear that β -decay data alone would ever be able to lead to a unique law.

The argument **V** now offered is in one respect an extension of **IV**. A Fierz type of interference can be identified in the theory of once-forbidden spectra as

¹ Sherr, Muether, and White, *Phys. Rev.* **75**, 282 (1949); R. Sherr and J. Gerhart, *Phys. Rev.* **86**, 619 (1952).

² A. Petschek and R. Marshak, *Phys. Rev.* **85**, 698 (1952).

³ What is generally called the "allowed shape" (linear Kurie plot) is here called the "statistical shape" because we consider at least the possibility that allowed spectra deviate from linearity on Kurie plots.

⁴ C. S. Wu, *Revs. Modern Phys.* **22**, 386 (1950).

well as for allowed transitions. Whereas for the latter the Fierz interference arises only from SV and TA combinations, in the once-forbidden cases it also arises from SA , VT , and AP combinations. That such interference should not be admitted for once-forbidden spectra seems to be supported by evidence of the same kind as for allowed spectra: All the normal once-forbidden spectra are reported to have the statistical shape. (The one "abnormal" example, RaE, needs separate consideration, Sec. 5.)

An important preliminary to argument **V** is an explanation of how a once-forbidden spectrum may have a statistical shape even in the absence of Fierz interference. The various effects which can arise are differentiated in Sec. 2. The condition for a statistical shape then becomes expressible as: a large Coulomb energy, Ze^2/R , at nuclear radius R , as compared to the kinetic energy which the electron and neutrino finally obtain. This condition is best fulfilled just for the observed cases in which the statistical shape is best confirmed. Whether the condition is sufficiently well fulfilled for all cases reported to have the allowed shape is not easily settled because of the essential occurrence of nuclear matrix elements which can be only crudely estimated. Hence, in Sec. 2 we take the standpoint that the statistical shape is experimentally established and follow out its implications. The result is our argument **V**.

The importance of the experimental detectability of Fierz interference for both arguments **IV** and **V** leads us to make a critical examination of the data: in Sec. 3, the allowed transitions and in Sec. 4, the once-forbidden cases. The empirical limits put on the Fierz type of effect, in both kinds of spectra, are about equally narrow. However, because of the greater complication of the once-forbidden theory, the interpretation there is not as straightforward as it is for the allowed transitions. It becomes clear that measurements of once-forbidden spectra which show positive deviations from the statistical shape (low Z , high energy) are needed to check on our explanation of the cases of no deviation, before argument **V** can be regarded as established on the same footing as **IV**.

2. THE SIGNIFICANCE OF ONCE-FORBIDDEN SPECTRA WITH STATISTICAL SHAPES

We use notation in which the β -interaction energy density is

$$\sum_X G_X X + \text{Herm. conj.} \quad (1)$$

with $X=S, V, T, A$, or P . Each of these five symbols stands for one of the interaction forms of the list (6a-e) of Konopinski.⁵ Each G_X is a Fermi coupling constant, measuring the magnitude of interaction of form X which might be present in the correct law.

We present the theoretically expected spectra in terms of a "shape factor,"⁶ C_n , which multiplies the

⁵ E. J. Konopinski, *Revs. Modern Phys.* **15**, 209 (1943).

⁶ Called "correction factor" in the paper of E. J. Konopinski and G. E. Uhlenbeck, *Phys. Rev.* **60**, 308 (1941).

well-known statistical factor to give the spectrum characteristic of the degree of forbiddenness, $n(=0, 1, 2 \dots)$. For allowed transitions,⁷

$$C_0 = [G_S^2 + \kappa^2 G_V^2 \pm (2\kappa/W)G_S G_V] \left| \int \beta \right|^2 + [G_T^2 + \lambda^2 G_A^2 \pm (2\lambda/W)G_T G_A] \left| \int \beta \sigma \right|^2. \quad (2)$$

The upper sign refers to positron, the lower to negatron, emission. Of course, the intrinsic signs of $G_S G_V$ and $G_T G_A$, as well as their magnitudes, are to be treated as unknown. The real⁸ parameters κ and λ are defined by

$$\int 1 = -\kappa \int \beta \quad \text{and} \quad \int \sigma = -\lambda \int \beta \sigma, \quad (3)$$

which are nuclear matrix elements to be found in reference 5. W is the electron energy, inclusive of rest mass, in units of mc^2 .

The terms $\sim 1/W$ in (2) are the well-known Fierz interference terms. Their presence would cause allowed spectra to deviate systematically from a statistical shape. The apparent absence of such deviations is the basis of argument **IV** of the introduction. We proceed to show that interference effects of the same type can be identified in the theory of once-forbidden spectra.

The latter was worked out by Uhlenbeck and Konopinski⁶ for each of the five interaction forms separately. Recently, Smith⁹ and Pursey¹⁰ calculated the contributions from the interference between the various forms. The resulting shape factor C_1 , if written with the same generality as C_0 in (2), would require too formidable an expression for convenience. We therefore take several steps to simplify its exposition.

First, we restrict the name "once-forbidden" here to just the transitions having spin changes $\Delta I=0$ or ± 1 , together with parity change. The $\Delta I=\pm 2$ transitions are well understood, as indicated in argument **I** of the introduction, and need no discussion here.

Second, we accept argument **IV**,¹¹ so that we need consider as possible alternatives for the β -law only the four arbitrary combinations: STP , SAP , VTP , or VAP .

Third, we separate the shape factor into three parts:

$$C_1 = \dot{C}_1^{(0)} + C_1^{(1)} + C_1^{(2)}. \quad (4)$$

$C_1^{(0)}$ is to consist of the terms which contribute to $I_i=0 \rightarrow I_f=0$ transitions. Then the S and V forms do not contribute to $C_1^{(0)}$ at all, and only $C_1^{(0)}(TP)$ and $C_1^{(0)}(AP)$ will be needed [(12) and (13)]. $C_1^{(1)}$ is to consist of terms which contribute to $\Delta I=\pm 1$ but not to $\Delta I=\pm 2$ (or $0 \rightarrow 0$) transitions. The P interaction

⁷ M. Fierz, *Z. Physik* **104**, 553 (1937). The largest contribution of the P form is $G_P^2 \int \beta \gamma_6^2$; it requires a nuclear parity change and so is classed as once-forbidden, rather than allowed.

⁸ C. Longmire and A. Messiah, *Phys. Rev.* **83**, 464 (1951).

⁹ A. Smith, *Phys. Rev.* **82**, 955 (1951).

¹⁰ D. Pursey, *Phil. Mag.* **42**, 1193 (1951).

¹¹ Evidence for it is examined critically in Sec. 3.

TABLE I. Once-forbidden transitions.

| | ΔI^a | W_0 | $\alpha Z/R(W_0-1)$ | Ref. |
|-------------------|--------------|-------|---------------------|------|
| Pm ¹⁴⁷ | 5/2→7/2 | 1.415 | 49.5 | b |
| W ¹⁸⁵ | 3/2→5/2 | 1.84 | 27.5 | c |
| Pr ¹⁴⁸ | 5/2→7/2 | 2.82 | 11.8 | c |
| Rb ⁸⁶ | ? | 2.44 | 11.0 | d |
| Au ¹⁹⁸ | ? | 2.88 | 13.7 | c |
| Ba ¹³⁹ | ? | 5.5 | 4.6 | f |
| RaE | 0→0(?) | 3.29 | 11.5 | |

^a See reference 13.
^b See reference 14.
^c See reference 15.

^d See reference 16.
^e See reference 17.
^f See reference 18.

makes no contribution to $C_1^{(1)}$ since its only matrix element⁷ is $\int \beta \gamma_5$, which vanishes unless $\Delta I=0$. The $C_1^{(2)}$ is the complete shape factor for the $\Delta I=\pm 2$ transitions which we have dismissed; however, $C_1^{(2)}$ also contributes to $\Delta I=0, \pm 1$ transition, although not to $0\rightarrow 0, 1\leftrightarrow 0$ or $\frac{1}{2}\rightarrow\frac{1}{2}$ cases. As indicated in argument I, $C_1^{(2)}$ arises from T or A interactions only.

Fourth, we introduce certain real⁸ parameters, x_i, y_i, z_i , which help simplify the notation for the nuclear matrix elements which occur. The latter are defined in references 5 and 6. Here we define x_i by means of

$$\begin{aligned}
 \int \beta \mathbf{r} &= -ix_1(G_T/G_S) \int \beta \boldsymbol{\sigma} \times \mathbf{r}, \\
 \int \mathbf{r} &= -ix_2(G_A/G_V) \int \boldsymbol{\sigma} \times \mathbf{r}, \\
 \int \beta \mathbf{r} &= ix_3(G_A/G_S) \int \boldsymbol{\sigma} \times \mathbf{r}, \\
 \int \mathbf{r} &= ix_4(G_T/G_V) \int \beta \boldsymbol{\sigma} \times \mathbf{r}.
 \end{aligned}
 \tag{5}$$

Next, the y_i are defined by

$$\begin{aligned}
 \int \beta \boldsymbol{\alpha} &= y_1(\alpha|Z|/2R) \int \beta \boldsymbol{\sigma} \times \mathbf{r}, \\
 \int \boldsymbol{\alpha} &= y_2(\alpha|Z|/2R)(G_A/G_V) \int \boldsymbol{\sigma} \times \mathbf{r}, \\
 \int \boldsymbol{\alpha} &= -y_4(\alpha|Z|/2R)(G_T/G_V) \int \beta \boldsymbol{\sigma} \times \mathbf{r}.
 \end{aligned}
 \tag{6}$$

Here, Z is the nuclear charge and R the nuclear radius in units of \hbar/mc . Finally, the z_i are defined by

$$\begin{aligned}
 \int \gamma_5 &= iz_1(\alpha|Z|/2R) \int \boldsymbol{\sigma} \cdot \mathbf{r}, \\
 \int \beta \gamma_5 &= -iz_2(\alpha|Z|/2R)(G_A/G_P) \int \boldsymbol{\sigma} \cdot \mathbf{r}, \\
 \int \beta \gamma_5 &= iz_3(\alpha|Z|/2R)(G_T/G_P) \int \beta \boldsymbol{\sigma} \cdot \mathbf{r}.
 \end{aligned}
 \tag{7}$$

Attempts^{10,12} have been made to evaluate these matrix elements from simplified nuclear models. These estimates give x_i, y_i, z_i the order of magnitude unity when all $G_X/G_{X'}\rightarrow 1$ (except $z_{2,3}\rightarrow 0$, see below).

Fifth, we present the shape factor here only in the approximation $(\alpha Z)^2 \ll 1$. This is more than adequate for our purposes. We have actually used the precise expressions to be found in references 6, 9, and 10 for checking various of the smaller effects. The corresponding increase in accuracy is, however, largely illusory, because of uncertainties in nuclear matrix elements, in the nuclear radius, and in the finite nucleus and screening effects.

We are now ready to give $C_1^{(1)}$ for the $ST(P)$ and $VA(P)$ interactions. For both, one can use the single, positive-definite expression:

$$\begin{aligned}
 C_1^{(1)} &= G_A(T)^2 \left| \int (\beta) \boldsymbol{\sigma} \times \mathbf{r} \right|^2 \left\{ \left[\frac{\alpha Z}{2R} (1+x_i-y_i) \right. \right. \\
 &\quad \left. \left. + (1+x_i) \frac{p^2}{3W} - (1-x_i) \frac{q}{3} \right]^2 + \left(\frac{1+x_i}{3} \frac{p}{W} \right)^2 \right. \\
 &\quad \left. + \frac{(1-2x_i)^2}{18} p^2 + \frac{(1+2x_i)^2}{18} q^2 \right\}.
 \end{aligned}
 \tag{8}$$

The upper sign applies to the VA combination, the lower to ST . Further, one may change to the case of positron emission by changing the sign of y_i , $i=2$ and 1 , respectively, for VA and ST . p and q are electron and neutrino momenta, respectively, in units of mc . $q=W_0-W$ if W_0 is the spectrum end-point value of the electron energy W .

Expression (8) makes clear an important result: The main Coulomb effect on the electron, $\sim(\alpha Z/2R)^2$, is *energy-independent*. Dependence on energy here means on the kinetic energy with which the electron emerges, and naturally the Coulomb potential energy is independent of that.

As contrast, we immediately present $C_1^{(2)}$, which constitutes the entire shape factor for the $\Delta I=\pm 2$ transitions:

$$C_1^{(2)} = G_{T,A}^2 \Sigma |B_{ij}|^2 (p^2 + q^2) / 12. \tag{9}$$

B_{ij} is another nuclear matrix element defined in reference 6. This expression yields the distinctive shapes referred to in argument I and arises only from the T or A forms of interaction. Here, the large possible angular momentum change $|\Delta I|$ makes the role of orbital angular momenta important. As a consequence, energy-dependent centrifugal effects dominate, rather than such Coulomb effects as were described in the preceding paragraph. The energy-dependent terms of (8) are largely centrifugal effects like those of (9).

Not all the constant terms of (8) arise from the Coulomb effect on the electron. The matrix elements

¹² T. Ahrens and E. Feenberg, Phys. Rev. **86**, 64 (1952).

$\mathcal{F}(\beta)\alpha$ and $\mathcal{F}(\beta)\gamma_i$ of (6) and (7) contribute to the once-forbidden shape factors in the same way as do the $\mathcal{F}(\beta)$ and $\mathcal{F}(\beta)\sigma$ to the allowed shape factor (2), i.e., with constant terms. However, as the estimates of (6) and (7) mentioned above indicate, here also the Coulomb energy is a measure of the size of the contribution. This results in the combination of Coulomb terms actually shown in (8). Hence, the size of all the energy-independent contributions to the shape factor can be said to be measured by the Coulomb energy at the nuclear radius, $\alpha Z/R$.

The existence of the constant terms has particular significance in view of the fact that all the measured once-forbidden spectra are reported to have the statistical shape (except RaE, Sec. 5). Thus, the experiments appear to demand an energy-independent shape factor, C_1 .

One can now see that it is important for all the observed cases, that the Coulomb energy should dominate over the electron-neutrino kinetic energies: $\alpha Z/R \gg W_0 - 1$. The uncertain parameters x_i, y_i cannot take on values which would tend to cancel the Coulomb terms. The only alternative would be for the kinetic energy terms to preserve a precarious balance against each other. Such a balance should in principle vary from case to case and frequently fail, giving deviations such as have not so far been observed. It is fortunate for the theory that in all the cases for which the statistical shape is reported, the condition $\alpha Z/R \gg W_0 - 1$ is fulfilled. This is shown in Table I¹³⁻¹⁸ for the once-forbidden cases in which the spectra are best measured (i.e., unobscured by concurrent transitions).

Supporting evidence for our interpretation that it is the dominance by the Coulomb terms $\sim(\alpha Z/2R)^2$ which is responsible for the statistical shapes comes from the $\Delta I = \pm 2$ transitions. A notable fact about these distinctive transitions is their long comparative half-lives ($\langle \log ft \rangle_w \approx 9$) as against those for $\Delta I = \pm 1$ ($\langle \log ft \rangle_w \approx 7$). The main part of this difference must be due to the absence of the large Coulomb contribution $\sim(\alpha Z/2R)^2$ in (9). This implies that if the Coulomb term were suppressed in (8), e.g., by adverse x_i, y_i values, then the $\Delta I = \pm 1$ half-lives would become as long as the $\Delta I = \pm 2$ ones, contrary to what is actually observed.

Not only (8), but all the possible $\Delta I = 0, \pm 1$ shape factors contain the Coulomb terms $\sim(\alpha Z/2R)^2$. For the $SA(P)$ and $VT(P)$ combinations, we write $C_1^{(1)}$ keeping only these usually dominant terms:

$$C_1^{(1)}(SA) \approx G_A^2 \left| \int \sigma \times \mathbf{r} \right|^2 (\alpha Z/2R)^2 \times [1 + x_3^2 - 2x_3/W], \quad (10)$$

¹³ Mayer, Moszkowski, and Nordheim, Revs. Modern Phys. **23**, 315 (1951).

¹⁴ Langer, Motz, and Price, Phys. Rev. **77**, 798 (1950).

¹⁵ L. M. Langer (private communication); see also F. B. Shull, Phys. Rev. **74**, 917 (1948).

¹⁶ H. R. Muether and S. L. Ridgway, Phys. Rev. **80**, 750 (1950).

¹⁷ L. M. Langer and H. C. Price, Phys. Rev. **76**, 641 (1949).

¹⁸ L. R. Shepherd and S. M. Hill, Nature **162**, 566 (1948).

$$C_1^{(1)}(VT) \approx G_T^2 \left| \int \beta \sigma \times \mathbf{r} \right|^2 (\alpha Z/2R)^2 [(1 - y_1)^2 + (x_4 - y_4)^2 - 2(1 - y_1)(x_4 - y_4)/W]. \quad (11)$$

The striking fact about these expressions is that they predict deviations from the statistical shape even for large Coulomb energies. Yet we depend on the latter to account for the occurrence of statistical shapes among once-forbidden spectra.

The energy-dependent terms of (10) and (11) are $\sim 1/W$ and arise from the calculations in the same way as do the Fierz interference terms in the allowed shape factor C_0 of (2). Thus, an argument develops against the inclusion of SA and VT combinations in the β -law which is of the same kind as argument **IV** against SV and TA combinations.

There remains to be considered the shape factors $C_1^{(0)}$ which form a part of the total shape factor (4) for $\Delta I = 0$ transitions and constitute the entire shape factor for $0 \rightarrow 0$ transitions. For either the $(S)TP$ or $(V)TP$ forms of the β -law,

$$C_1^{(0)}(TP) = G_T^2 \left| \int \beta \sigma \cdot \mathbf{r} \right|^2 \left\{ \left[\frac{\alpha Z}{2R} (1 - z_3) + \frac{1}{3} \left(\frac{p^2}{W} - q \right) \right]^2 + \left(\frac{p}{3W} \right)^2 \right\}. \quad (12)$$

Clearly, this leads again to the statistical shape in the usual case of $\alpha Z/R \gg W_0 - 1$. However, the $(S)AP$ or $(V)AP$ forms yield

$$C_1^{(0)}(AP) \approx G_A^2 \left| \int \sigma \cdot \mathbf{r} \right|^2 (\alpha Z/2R)^2 [(1 - z_1)^2 + z_2^2 + 2z_2(1 - z_1)/W], \quad (13)$$

when $\alpha Z/R \gg W_0 - 1$. Here, the Fierz type of deviation occurs again.

The complete roster of possible once-forbidden shape factors, (8) to (13), has led to the following conclusions. The reported statistical shapes (energy-independent shape factors) seem best explainable, according to either an STP or VA form of β -law, as due to the normal dominance of the Coulomb energy. The VTP or SAP forms are probably incorrect because even for large Coulomb energies they, in general, predict deviations from the statistical shape, of a Fierz type.

Two considerations tend to keep this argument from being completely conclusive, at least for the present. These apply to some degree also to the widely accepted argument **IV**, which has a similar basis in allowed spectra. They are:

(a) The uncertain quantities x_i, y_i, z_i may have such values, in all the cases observed so far, that the Fierz interferences would be unobservable.

(b) Although the spectra are reported to have sta-

tistical shapes, the experiments might have been too inaccurate to detect Fierz deviations.

The adequacy of the experiments is discussed in detail in Sec. 3, for allowed spectra, and in Sec. 4, for once-forbidden spectra.

As regards point (a), one may point out at once that it is unlikely for the matrix elements, in all the cases observed, to take on just such values as would hide the effects of Fierz interference. In principle, the matrix elements may vary markedly, relative to each other, from case to case. Certainly, the objection (a) will become difficult to believe if the reports of statistical shapes of greater accuracy continue to accumulate.

For a less vague consideration of the objection (a), we may rely on the estimates of the nuclear matrix elements due to Pursey¹⁰ and to Ahrens and Feenberg.¹² One has

$$\kappa \approx 1 \quad \text{and} \quad \lambda \approx 1 \quad (14)$$

for the parameters in C_0 , (2). This results immediately from the nonrelativistic approximation for the Dirac β (≈ -1). That approximation is invalid if β does not commute with other operators occurring in the matrix element.

We quote the estimates for the ratios involved in (5), (6), and (7) in terms of the quantities ξ_i , η_i , ζ_i defined exactly like x_i , y_i , z_i except that the G 's are omitted, i.e.,

$$x_i \rightarrow \xi_i, \quad y_i \rightarrow \eta_i, \quad z_i \rightarrow \zeta_i \quad \text{as} \quad G_X/G_{X'} \rightarrow 1. \quad (15)$$

Further, we restrict the quotations for ξ_i , η_i to those applicable to the first three, better-defined cases in Table I:

$$\xi_1 \approx \xi_2 \approx \xi_3 \approx \xi_4 \approx 1, \quad (16)$$

$$\eta_1 \approx \eta_2 \approx \eta_4 \approx \Lambda, \quad (17)$$

where Λ is a quantity defined by Ahrens and Feenberg. These authors find $\Lambda \approx 1$ for such cases as ours. On the other hand, Pursey gives $\Lambda \approx 2$. The discrepancy stems from different assumptions as to the nuclear model. The estimates (16) are probably more reliable than the others since they follow from kinematical considerations whereas Λ requires more presumptions about the nuclear Hamiltonian.

Further, Ahrens and Feenberg give

$$\zeta_1 \approx 1, \quad (18)$$

while Feenberg and Primakoff¹² estimate

$$\zeta_2 \approx \zeta_3 \approx 0, \quad (19)$$

TABLE II. Allowed spectra.

| | Rad'n. | ΔI | W_0 | Ref. |
|------------------|-----------|------------|-------|------|
| N ¹³ | β^+ | 0 | 3.44 | a |
| S ³⁵ | β^- | 0 | 1.33 | b |
| Cu ⁶⁴ | β^- | 1 | 2.119 | c |
| Cu ⁶⁴ | β^+ | 1 | 2.285 | c |

^a See reference 19.

^b See reference 20.

^c See reference 21.

being of the order of m/M , the ratio of electron to nucleon mass. According to the last result, the P form will not contribute at all unless G_P has an unlikely high value.

If one accepts the estimates just reviewed, one sees that objection (a) does not hold for C_0 and for $C_1^{(1)}(SA)$. Thus, sufficiently accurate experiments should reveal the presence of Fierz interference if the SV , TA , or SA combinations occur in the correct law. The apparent absence of an observable Fierz deviation confirms argument IV and, further, also our new argument insofar as it is directed against the SA combination.

The consequence of the estimates for the VT combination (11) depends on whether $\Lambda \approx 1$ or $\Lambda \approx 2$. If $\Lambda \approx 1$, not only is the Fierz interference in the $C_1^{(1)}(VT)$ shape suppressed but *all* the Coulomb terms $\sim (\alpha Z/2R)^2$. Hence, a VT law would predict for $\Delta I = \pm 1$ transitions a distinctly nonstatistical shape very similar to the $\Delta I = \pm 2$ shapes; moreover, the half-lives of the $\Delta I = \pm 1$ and $\Delta I = \pm 2$ transitions would be expected to be about the same. Thus, Feenberg's evaluation ($\Lambda \approx 1$) would make the argument against the VT combination very strong.

On the other hand, Pursey's estimate of $\Lambda \approx 2$ would make the Fierz interference in the VT combination observable. If one accepts only the more reliable estimate $\xi_4 \approx 1$ for the first three cases of Table I, then (11) becomes

$$C_1^{(1)}(VT) \approx \left| \int \beta \sigma \times \mathbf{r} \right|^2 (\alpha Z/2R)^2 (1 - \Lambda)^2 \times [G_T^2 + G_V^2 - 2G_T G_V/W]. \quad (20)$$

Thus, if the constant terms here are appreciable at all ($\Lambda \neq 1$) then a Fierz deviation should be present unless $G_V G_T \rightarrow 0$.

Finally, accepting the estimates (19) as valid destroys our argument as directed against the AP combination (13). If the P form does not contribute at all, naturally its effects become unobservable and objection (a) holds. However, the RaE case, discussed in Sec. 5, may indicate that occasionally at least the estimate (19) fails.

On the basis of the estimates of the nuclear matrix elements, therefore, argument V can be successfully directed only against including VT and SA forms in the β -law. This, together with argument IV, leaves only the STP and VAP possibilities.

3. FIERZ INTERFERENCE IN ALLOWED SPECTRA

The absence of Fierz deviations in allowed spectra is frequently quoted as a basis for argument IV, but no systematic examination seems to have been presented so far. We have special interest in the details because the sensitivity of current measurements to the Fierz effect also determines the effectiveness of argument V, which depends on the absence of the Fierz type of deviation in once-forbidden spectra [see objection (b) in Sec. 2].

The most convenient way to derive information from spectrum shapes is to examine their (Fermi-)Kurie plots. The ordinates of these are, in theory,

$$K_n = (W_0 - W)C_n^{\frac{1}{2}}. \quad (21)$$

When the shape factor C_n is independent of the energy W , the spectrum has a statistical shape and the Kurie plot is linear $\sim (W_0 - W)$. Kurie plots of experimental points have an arbitrary ordinate scale, being proportional to the square root of the source intensity. This fact, together with the fact that the plot itself is used to determine W_0 , tends to make experimental Kurie plots insensitive to possible deviations from linearity. Nevertheless, the current accuracy of measurement does not seem to warrant more sensitive treatment.

If the Fierz type of effect should be present, then the Kurie plot should deviate from linearity, since it is now proportional to

$$K_n \sim (W_0 - W)(1 + 2\varphi/W)^{\frac{1}{2}}, \quad (22)$$

where φ is some number for each spectrum, given in theory by some one of the shape factors like (2), (10), or (11). The report that the spectra yield linear Kurie plots implies $\varphi = 0$, but some amount of deviation would have been undetected. Thus, we can only expect to set upper limits on $|\varphi|$ from the experiments. If these turn out to be substantially less than unity, their difference from zero will have been rendered unlikely. That conclusion has particular force when a small $|\varphi|$ implies some $|G_X/G_{X'}|$ is a small fraction. A theory with one Fermi constant much smaller than another strains credibility. The burden of proof is transferred to the point of view that the small component exists.

The observed spectra which have the best chance to reveal the presence of Fierz interference are those which are measured reliably down to a low electron energy. We therefore look at spectra which are not superposed with concurrent, lower energy, transitions. Table II seems to have the best measured examples.¹⁹⁻²¹

To provide an equal basis for judging the consistency with experiment of the theories with ($\varphi \neq 0$) and without ($\varphi = 0$) Fierz interference, we proceeded as follows. For each test, we chose a suitable (small) numerical value of φ . Then we compared the conventional experimental Kurie plot ($\varphi = 0$) with a plot of the same points divided by $(1 + 2\varphi/W)^{\frac{1}{2}}$. The second type we refer to as a "Fierz plot." The latter, rather than the conventional plot, should be linear if actually the $\varphi \neq 0$ is correct, as can be seen from (22). Of course, it turns out that both plots are nearly linear if $|\varphi|$ has been chosen small enough. We then located the straight lines which the experimental points of each plot ought to follow by using the points on the high energy side of the intensity maximum, the corresponding ones of the

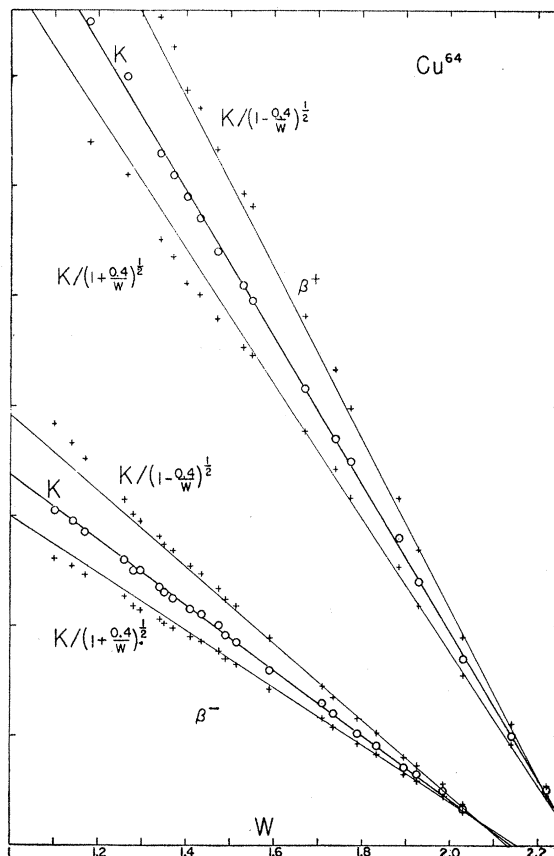


FIG. 1. Conventional Kurie (K) and Fierz ($K/(1+2\varphi/W)^{\frac{1}{2}}$) with $\varphi = \pm 1/5$ plots for the Cu^{64} spectra.

Kurie and of the Fierz plots. That part of every spectrum is always the most reliably measured. Finally we examined and evaluated the deviations from the constructed lines which are exhibited by the points not used in the constructions.

The conventional Kurie plots usually showed roughly random deviations from linearity. We took the root-mean-square of the percentage deviations, ϵ , as a measure of the experimental accuracy. Then we found in each case that values of φ surpassing certain limits would yield Fierz plots which deviated *systematically* from linearity. We regarded the limiting φ 's to have been reached when the systematic mean percentage deviation δ in the Fierz plot just exceeded the random deviation ϵ in the Kurie plot. This seemed quite a generous allowance for random error. Of course, we did not try infinite progressions of φ 's. From a suitable enough initial choice it can be shown possible to perform a type of linear extrapolation to the limiting φ : We find $\delta - \delta_0 \sim \varphi$ if δ_0 is the percentage deviation in the Kurie plot.

Cu^{64} (β^-)

Ten points with $W > 1.5$ were used to determine the straight lines for the Kurie plot and two Fierz plots with $\varphi = \pm 0.2$ in

¹⁹ W. Hornyak and T. Lauritsen, Phys. Rev. **77**, 160 (1950).

²⁰ R. Albert and C. S. Wu, Phys. Rev. **74**, 847 (1949); Langer, Motz, and Price, Phys. Rev. **77**, 798 (1950).

²¹ Langer, Moffat, and Price, Phys. Rev. **76**, 1725 (1949); C. Wu and R. Albert, Phys. Rev. **75**, 1107 (1949).

Fig. 1. For the Kurie plot the remaining twelve points were randomly distributed above and below the straight line with a mean relative deviation of $\delta_0 \approx 0.1$ percent; the root mean square of the relative deviations was $\epsilon = 0.9$ percent. As for the Fierz plots, the deviations were systematic, too low for $\varphi = +0.2$ and high for $\varphi = -0.2$, the mean value of their relative magnitudes amounting, respectively, to $\delta_+ = -2.0$ percent ($\varphi = +0.2$) and $\delta_- = +3.0$ percent ($\varphi = -0.2$). From these we concluded that the values of φ capable of giving Fierz deviations, undetectable within the accuracy of the measurements ($\epsilon = 0.9$ percent), were limited by $-0.06 < \varphi < 0.08$.

A disturbing factor in the course of the reasoning here is that the deviation in the $\varphi < 0$ Fierz plots resembles somewhat the effects of too thick a source being used in the measurement. Actually there is no serious danger that this is so; thick source effects tend to set in with distinctly greater abruptness than the theoretical deviations here. Furthermore, it is easy to put the whole question to rest by looking also at positron spectra, which are expected to have deviations of the opposite sign according to (2).

Cu⁶⁴ (β^+)

The procedure used for the electron spectrum of Cu⁶⁴ gave similar results when applied to the positron spectrum. Here seven points with $W > 1.6$ determined the straight lines, and eleven points with $W < 1.6$ were used to measure the deviations. With the notation used for the previous case we found $\delta_+ = -3.9$ percent, $\delta_- = 3.1$ percent, $\epsilon = 1.1$ percent, $\delta_0 = -0.7$ percent, with the consequent limit on unobserved Fierz deviations of $-0.1 < \varphi < 0.03$.

It now seems possible to say that $|\varphi| \lesssim 0.1$ for allowed transitions with $\Delta I = \pm 1$. In such cases, C_0 of (2) is simplified because $\int \beta = \int 1 = 0$. Then, theoretically,

$$\varphi = \pm \lambda G_T G_A / (G_T^2 + \lambda^2 G_A^2). \quad (23)$$

Hence, we can say $|\lambda G_A / G_T| \lesssim 0.1$ or $|G_T / \lambda G_A| \lesssim 0.1$. Considering that probably $\lambda \approx 1$, see (14), we have here the implication that one of the G 's is less than 0.1 of the other. This is about as closely as the present experiments can prove that not both G_T and G_A differ from zero. The significant limits are

$$G_A^2 / G_T^2 < 1 \text{ percent} \quad \text{or} \quad G_T^2 / G_A^2 < 1 \text{ percent.}$$

We hope to come to a similar conclusion about the pair G_S, G_V from the $\Delta I = 0$ experiments.

N¹³

The measurement of the deviations of thirteen points with $W < 2.5$ from the straight line drawn through ten points with $W > 2.5$ gave $\delta_+ = -3.6$ percent; $\delta_- = 4.3$ percent, $\epsilon = 1.2$ percent, $\delta_0 = +0.5$ percent, and consequently $-0.04 < \varphi < 0.09$.

S³⁵

Six points with $W > 1.16$ and nine points with $W < 1.16$ were used giving $\delta_+ = -2.2$ percent, $\delta_- = 1.7$ percent, $\epsilon = 1.2$ percent, $\delta_0 = +0.4$ percent, from which $-0.12 < \varphi < 0.12$.

The N¹³ experiment seems good enough to conclude that $|\varphi| \lesssim 0.1$ for $\Delta I = 0$ transitions, also. Theoretically,

see (2),

$$\varphi = \pm \kappa G_S G_V / \left[G_S^2 + \kappa^2 G_V^2 + (\lambda^2 G_{T(A)})^2 \left(\left| \int \beta \sigma \right|^2 / \left| \int \beta \right|^2 \right) \right] \quad (24)$$

depending on whether the near vanishing of (23) is taken to imply that G_T or G_A is to be neglected. For²³ N¹³, $|\int \beta \sigma|^2 / |\int \beta|^2 \approx 1/3$. The Fierz interference is expected to be less detectable when $\Delta I = 0$ than it was for $\Delta I = \pm 1$. Nevertheless, it is clear that $|G_S / G_V|$ or $|G_V / G_S|$ must be substantially less than unity and presumably zero.

4. FIERZ-TYPE INTERFERENCE IN ONCE-FORBIDDEN SPECTRA

We wish to compare argument V, against SA and VT combinations, with the argument IV, against SV and TA combinations, by showing that the once-forbidden spectra limit the amount of the Fierz-type deviation as much as do the allowed spectra.

We probably must forego directing our argument also against AP combinations. Not only does the estimate (19) of $\int \beta \gamma_5$ make it possible that P effects are unobservable anyway, but also there seem to exist no suitable measurements to implement the argument for this case. Many spectra have been observed which are expected to have¹³ $\Delta I = 0$ (Zr⁹⁷, Ag¹¹¹, Cd¹¹⁵, In^{115m}, La¹⁴¹, Dy¹⁶⁵, Er¹⁶⁹, Er¹⁷¹, Ce¹⁴⁴, Pr¹⁴⁴). However, all these are overlaid with concurrent transitions or other obscuring effects. The unobscured portion of each spectrum is reported to have the statistical shape, but the obscurations may have hidden considerable Fierz interference.

Table I lists all the once-forbidden spectra which seem to be well-measured down to low energies. Only for the first three cases does the shell model define the spin change fairly well, as $\Delta I = 1$. For these reasons we confined our examination to Pm¹⁴⁷, W¹⁸⁵, and Pr¹⁴³.

Pm¹⁴⁷

Figure 2 shows the conventional Kurie plots and also the Fierz plots for $\varphi = \pm 1/5$. For each, the points with $W > 1.2$ were used to locate the presumptive straight lines. The deviations of the points with $W < 1.2$ were random for the Kurie plot ($\delta_0 = 0.2$ percent), having a root-mean-square percentage deviation, $\epsilon = 0.9$ percent. The $\varphi = +1/5$ plot has a systematic mean percentage deviation, $\delta_+ = -1.9$ percent. This reduces to -0.9 percent if $\varphi = 0.1$. The $\varphi = -1/5$ plot has a systematic mean deviation $\delta_- = +4.5$ percent. That reduces to $+0.9$ percent if $\varphi = -0.03$. We conclude $-0.03 < \varphi < 0.1$.

W¹⁸⁵

The results here were much the same as for Pm¹⁴⁷. The points near the end point showed some scattering; hence, they were not

²³ E. P. Wigner, Phys. Rev. 56, 519 (1939).

relied on to determine the straight lines. It was necessary to use instead points more nearly at the middle of the spectrum. This naturally cut down the range in which the Fierz deviations could show themselves. In conclusion, it was then possible to say that $-0.09 < \varphi < 0.14$.

Pr¹⁴³

The accuracy here was found to be about the same as for W¹⁸⁵. The result was $-0.13 < \varphi < 0.09$.

Thus, with some indication of concurrence from W¹⁸⁵ and Pr¹⁴³, the most stringent limits on the amount of Fierz interference is imposed by the particularly carefully measured Pm¹⁴⁷ spectrum. From it we can conclude that $|\varphi| \lesssim 0.1$.

We interpret the result $|\varphi| \lesssim 0.1$ by supposing, in turn, that the Fierz interference arises from the *SA* combination as in (10), then from the *VT* combination, as in (11) or (20). According to (10) and (15),

$$|\varphi| = |x_3|/(1+x_3^2) = |\xi_3 G_S G_A|/(G_A^2 + \xi_3^2 G_S^2).$$

We can put $\xi_3 \approx 1$ according to (16) without introducing more uncertainty than was done for argument IV when $\kappa \approx \lambda \approx 1$ was supposed. Then $|\varphi| \lesssim 0.1$ implies with the present interpretation:

$$G_A^2/G_S^2 \lesssim 1 \text{ percent or } G_S^2/G_A^2 \lesssim 1 \text{ percent.}$$

When the *VT* combination is being tested,

$$|\varphi| = |(1-\eta_1)(\xi_4-\eta_4)G_V G_T|/[(1-\eta_1)^2 G_T^2 + (\xi_4-\eta_4)^2 G_V^2]$$

according to (11) and (15). When (16) is used,

$$|\varphi| \approx |G_V G_T|/[G_T^2 + G_V^2].$$

Hence, $|\varphi| \lesssim 0.1$ makes

$$G_V^2/G_T^2 \lesssim 1 \text{ percent or } G_T^2/G_V^2 \lesssim 1 \text{ percent.}$$

One must remember, however, that the terms under consideration here are multiplied with $(1-\eta_1)^2$, which may nearly vanish. It has already been pointed out, in Sec. 2, how this would only strengthen the case against the *VT* combination.

We have thus carried out argument V to the same kind of result as IV. Ratios of the type $G_X^2/G_{X'}^2$ were shown to be small enough for the presumption that they vanish to be likely. However, such a presumption is not necessary to complete argument V, as it was for argument IV. This is because we have an independent measure for the possible values of the ratios in question. One can show²⁴ that the comparative half-lives of H³ (or the neutron) and He⁶ can be consistent with each other only if

$$G_F^2/G_G^2 = 0.6 \text{ to } 2.4.$$

Here G_F may be G_S or G_V , whereas G_G is G_T or G_A . The

²⁴ E. J. Konopinski and L. M. Langer, Annual Reviews of Nuclear Science, 2 (1952).

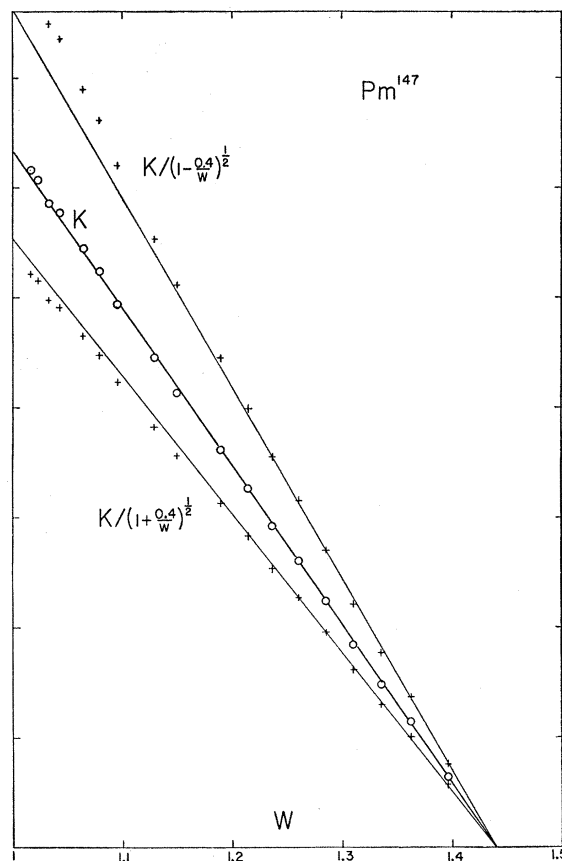


FIG. 2. Conventional Kurie (K) and Fierz plots ($\varphi = \pm 1/5$) for Pm¹⁴⁷.

subscripts *F* and *G* refer to “Fermi” and “Gamow-Teller” selection rules. One can see that the limits on the G ’s imposed by the absence of Fierz deviations make the *SA* and *VT* combinations inconsistent with the H³, He⁶ half-lives.

So far argument V may seem at least as secure as argument IV. However, we have ignored the small energy-sensitive terms, aside from the constant ones $\sim (\alpha Z/2R)^2$, which enter all the shape factors C_1 and are specifically shown in (8). We have been relying heavily on the reports of the statistical shape as an empirical fact from which the negligibility of the energy-sensitive terms follows. From the theoretical side, the negligibility of these terms depends on the precise values adopted for the nuclear radius, for the G ’s and for the relative sizes of the nuclear matrix element (the parameters ξ_i, η_i, ζ_i). Thus, the possible disturbing effect of the terms in question is subject to considerable uncertainty. It is fortunate that the best limits on φ have been derived from the Pm¹⁴⁷ case, which has a particularly low energy release W_0 and so should be the least disturbed.

The energy-sensitive terms might disturb the above conclusions in the following way. Contrary to those conclusions, the Fierz terms may actually be appreci-

able. Their presence might not be evident in deviations from the statistical shape because their effect is just cancelled by the energy-sensitive terms we have so far ignored. There is, of course, the immediate objection that it is unlikely for the statistical shape to depend on such accidental cancellations. We have nevertheless explored the possibility.

The efforts to save the *SA* and *VT* interactions, by supposing the constant shape factor to be a resultant of accidental cancellations of energy-sensitive terms, are easiest to describe for the *SA* case. This is because only the single uncertain parameter x_3 is involved. We did find that indeed $x_3 \approx +0.08$ yielded a more constant shape factor for Pm^{147} than did $x_3 = 0$ when a conventional value $R = 1.5A^{1/3}10^{-13}$ cm was used for the nuclear radius. However, $x_3 = \xi_3 G_S / G_A = 0.08$ makes $|\varphi| \lesssim 0.1$, within the limits which were regarded as an argument against the *SA* combination above. Moreover, if one wanted to save the Pr^{143} shape in the same way, the larger energy-dependent terms here make it necessary to adopt an x_3 approximately 3 times as large as for Pm^{147} . The similarity of the nuclear states involved in the two cases (see Table I) makes it implausible that ξ_3 could be responsible for such a threefold variation. It does not seem likely that this method of obtaining the observed statistical shapes should be taken seriously.

Similar efforts to save the *VT* interaction met with similar objections. The greater number of uncertain parameters which is here involved makes the description of the details too lengthy to be given here.

5 CONCLUDING DISCUSSION

The arguments **IV** and **V** by themselves leave only the alternatives *STP* and *VAP* with which to reproduce the observed statistical shapes of the once-forbidden spectra. These forms are supposed to do that by way of the large constant Coulomb terms in the shape factors (8). The energy-sensitive terms of (8) are supposed to be negligible, at least within the accuracy of the observations. The uncertain factors in all these terms make it impossible to prove that they indeed are so negligible; all that can be done at present is to show that it is possible that they are.

A typical situation arises when a detailed effort is made to fit $C_1^{(1)}(ST)$ of (8) to Pm^{147} . The only appreciable perturbation to the constant $(\alpha Z/2R)^2$ term is the addition to it of the terms

$$\frac{\alpha Z}{3R} \left\{ \frac{G_T + \xi_1 G_S}{G_T(1-\Lambda) + \xi_1 G_S} \frac{p^2}{W} + \frac{G_T - \xi_1 G_S}{G_T(1-\Lambda) + \xi_1 G_S} q \right\}.$$

If we take $G_T = G_S$, $\xi_1 = 1$, $\Lambda = 1$, and $R = 1.5A^{1/3} \times 10^{-13}$ cm, this causes $\sim +3$ percent mean deviation from the statistical shape. This deviation can be reversed if $\Lambda \rightarrow 2$ and $G_T > G_S$ instead. Such conclusions were little changed when we used the more precise expressions of references 6 and 9, and when we considered finite

nucleus and screening effects. One sees that there is little point in pursuing this study in view of the uncertainties involved. Probably such uncertainties will be best resolved when cases with $W_0 - 1 \rightarrow \alpha Z/2R$ are found and measured. Certainly, deviative effects are better evaluated from definite deviations than through their apparent absence, as in the cases now available.

There is one case of definite deviation from the statistical shape which may be a once-forbidden transition: the singular RaE spectrum. This was analyzed by Petschek and Marshak,² who concluded that the shape could be reproduced only by $C_1^{(0)}(TP)$ and so must be a $0 \rightarrow 0$ transition. Rather than formula (12) they used its more precise equivalent,²⁵ to be found in references 6 and 9. In order to make the application successful, it was necessary to assume that

$$z_3 = \zeta_3 G_P / G_T \approx +1.$$

The main objection against the Marshak-Petschek treatment is Ahrens, Feenberg, and Primakoff's estimate (19) that $\zeta_3 \rightarrow 0$ and hence $z_3 \rightarrow 1$ is unlikely. This objection does indeed make the RaE analysis; hence argument **III**, doubtful. On the other hand, the RaE analysis may perhaps be regarded as empirical evidence that at least for some nuclear states the estimate $\zeta_3 \rightarrow 0$ ($\equiv \int \beta \gamma_5 \rightarrow 0$) may fail. The fact that the RaE case is the only one of its kind, even though deliberate searches¹⁷ have been directed toward finding similar cases, perhaps adds plausibility to the occurrence of the failure as a singular instance.

There is an additional, qualitative piece of evidence which supports the Marshak-Petschek result. $z_3 \approx 1$ means that the deviation from a constant shape factor, in spite of $\alpha Z/R \gg (W_0 - 1)$ as seen in Table I, is achieved through the approximate cancellation of the large term $\sim (\alpha Z/2R)^2$ in (12). This has the consequence that the half-life of RaE is lengthened; a highly uncertain estimate is a lengthening by a factor of about 10^2 .⁸ Now, the study of comparative half-lives reveals that RaE does indeed have a life $\sim 10^{2.5}$ times as long as that of otherwise similar nuclei. RaE has $\log ft \approx 8$, and involves in its transition an 83rd proton and a 127th neutron. The Pb^{209} transition also converts a 127th neutron into an 83rd proton. It has $\log ft \approx 5.6$. Hg^{205} and RaE'' involve 81st protons and 125th neutrons, one below magic numbers in each case. These nuclei have $\log ft = 5.4$ and 5.5 , respectively.

The Marshak-Petschek result provides the argument **III**, which is the only one favoring the *STP* over the *VAP* combination. Without it, our new argument **V** leaves both the latter alternatives, having only eliminated the *SAP* and *VTP* forms.

Our results contradict the Critchfield-Wigner²⁶ hy-

²⁵ They also find that the corrections for finite nuclear size are important.

²⁶ C. Critchfield, Phys. Rev. **63**, 417 (1943).

pothesis ($S-A-P$) and also the Tolhoek-DeGroot²⁷ symmetry principle. The latter leads one to expect either an SAP form or a VT form for the β -interaction. This may mean that the neutron and proton are not

quite as equivalent during Fermi interaction as is presumed in the symmetry principle.

We wish to acknowledge the help of Professor A. C. G. Mitchell in making this work possible. This work was assisted by the joint program of the ONR and AEC.

²⁷ H. Tolhoek and W. DeGroot, *Phys. Rev.* **84**, 150 (1951).

Variations in the Relative Abundances of the Isotopes of Common Lead

C. B. COLLINS, R. M. FARQUHAR, AND R. D. RUSSELL
Department of Physics, University of Toronto, Toronto, Canada
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Mass spectrometer measurements of the relative isotopic abundances of samples of lead ores from Archean-type rocks showed larger variations than reported by Nier. The isotopic constitution of one lead was within the limits calculated by Holmes and by Bullard and Stanley. From the new measurements combined with existing data estimates of the time of formation of the earth's crust of 3.5 billion years and of a maximum time of formation of the elements of 5.5 billion years have been calculated. These values are in reasonable agreement with previous estimates.

NIER¹ reported the first, and still the only, thorough investigation of the variations of the isotopic constitution of ordinary and radiogenic leads. Several authors have since used Nier's data to calculate the time of solidification of the earth's crust (Holmes,² 3.35 billion years, Bullard and Stanley,³ 3.29 billion years) and a limit on the time of formation of the elements (Alpher and Herman,⁴ 5.3 billion years). All these estimates have been based upon extrapolations from the abundances of ordinary leads ranging in age from 25 to 1400 million years. More analyses of ordinary leads dated at 2000 million years or older have been needed to check these estimates. We have therefore analyzed a number of common lead samples, dating as many as possible from Nier's ages and from analyses of radiogenic leads in our own laboratory. Calculations of the age of the earth's crust and of a maximum age of the elements have been made, using the new measurements listed in Table I combined with Nier's data.¹

The experimental work was carried out with a 180° direction-focusing Nier-type mass spectrometer with a resolution of about 1/300 of an atomic mass unit. Analyses were made using lead tetramethyl as previously reported by the authors^{5,6} and by Dibeler and

Mohler.⁷ The isotopic lead abundances listed in Table I are believed to be accurate to within 1 percent on an absolute basis, although comparative measurements are good to 0.1 percent.

Since Pb²⁰⁴ is not believed to be of radiogenic origin, the total amount of this isotope is assumed to have remained constant from the time of formation of the elements to the present. The abundances of Pb²⁰⁶, Pb²⁰⁷ and Pb²⁰⁸ are therefore given with respect to Pb²⁰⁴.

A value of 5.5 ± 0.2 billion years for the maximum limit on the time of formation of the elements has been calculated from the data given in Table I combined with Nier's measurements of the isotopic constitution of common leads of known age. The method of calculation and the assumptions made were essentially the same as those of Alpher and Herman,⁴ and all values of constants were the same except the half-life of U²³⁵, which was taken as 7.07×10^8 years.⁸

Using Nier's data, Alpher and Herman reported a figure of 5.3 billion years. The average abundances of the lead isotopes at different times have been listed in Table II.

The calculated relative number of atoms of U, Th, and Pb in the earth's crust at the present time are U—1.00, Th—3.83, and Pb—7.35.

The isotopic abundances of those lead minerals (Table I) which could be assigned an age were combined with Nier's measurements¹ on other dated lead minerals and used to calculate a value for the age of the earth's crust after the method of Bullard and Stanley.³ This method assumes that the isotopic constitution of lead

¹ A. O. Nier, *J. Am. Chem. Soc.* **60**, 1571 (1938); *Phys. Rev.* **55**, 150 (1939); Nier, Thompson, and Murphey, *Phys. Rev.* **60**, 112 (1941).

² A. Holmes, *Nature* **157**, 680 (1946); **159**, 127 (1947); **163**, 453 (1949).

³ E. C. Bullard and J. P. Stanley, *Suomen Geodeetisen Laitoksen Julkaisuja, Veroffentlichungen des Finnischen Geodatischen Institutes*, No. 36, 33 (1949).

⁴ R. A. Alpher and R. C. Herman, *Phys. Rev.* **84**, 1111 (1951).

⁵ Collins, Freeman, and Wilson, *Phys. Rev.* **82**, 966 (1951); C. B. Collins and J. R. Freeman, *Trans. Roy. Soc. Canada* **45**, Sec. IV, 23 (1951).

⁶ Collins, Lang, Robinson, and Farquhar, *Geol. Assoc. Canada* (to be published).

⁷ V. H. Dibeler and F. L. Mohler, *J. Research Natl. Bur. Standards* **47**, 337 (1951).

⁸ Fleming, Ghiorso, and Cunningham, *Phys. Rev.* **82**, 967 (1951).