Investigation of Higher Order Effects in the Allowed Beta Decay of Mn^{56}†

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Directional-correlation measurements were performed on the allowed 2.86-MeV β–0.845-MeV γ transition in the decay of Mn^{56}. The results indicate that a small negative correlation ($A_2=-0.0030\pm0.0011$) exists. For comparison, the β-γ directional correlation was also measured in the allowed β decay of I^{131}. The distribution was found to be isotropic within the experimental error and the correlation coefficient was found to be $|A_2|<0.0005$ in this case. The deviation of the Mn^{56} β spectrum from allowed shape is discussed and it is shown that a shape factor of the form $C\approx1+0.04W-0.01/W$ would not be inconsistent with the observed anisotropy.

I. INTRODUCTION

THE 2.86-MeV β transition in the decay of Mn^{56} to the first excited 2^+ state in Fe^{56} has a comparative half-life which is large for an allowed β decay (see Fig. 1). The $\log ft$ value of this $3^+(\beta)2^+$ transition is 7.2,[1] which means that the transition rate is considerably slower than what is normally the case for an allowed β decay. Therefore, one would suspect that higher order terms, which usually can be neglected in allowed β transitions, may have an observable effect. If this is the case, the leptons can no longer be considered as being emitted as an s wave, since, because of a reduction of the ordinary allowed matrix elements $\int 1$ and $\int \boldsymbol{\sigma}$, contributions from p- and d-wave leptons may become important. A measurement of the directional correlation between the β ray and the cascade γ ray should then show a small deviation from isotropy. Similarly, the energy distribution of the emitted β particles may be expected to have a nonstatistical shape.

Deviations from statistical shape have been observed in several accurate shape-factor measurements on various isotopes.[2] The shape factor can be expressed in general as

$$C(W)=1+aW+b/W+cW^2, \qquad (1)$$

where the magnitudes of the energy-independent coefficients a, b, and c are usually very small compared to unity. Thus, in most allowed and several first-forbidden transitions, a measurement of the β spectrum can be fitted with an energy-independent shape factor. A definite deviation from allowed shape has been observed[3,4] in the high-energy β decay ($W_0\approx33m_0c^2$) of B^{12} and N^{12}. The measured shape factor is of the form $C(W)=1\pm0.003W$ with opposite signs for negatron and positron decay. The effect can be understood on the basis of the conserved-vector-current (CVC) theory.[5] However, a deviation of experimentally observed β spectra from statistical shape has also been observed for allowed transitions with low-energy endpoints ($W_0\approx2$–$5\ m_0c^2$). In general, the coefficient a is small and ranges from 0.004 to 0.04. A striking shape-factor anomaly has been observed by the Indiana group, who find that most of their well-measured β spectra can be fitted by an empirical shape factor of the form $C(W)=1+b/W$. The coefficient b is positive for both negatron and positron decay and of the order $0.2\leqslant b\leqslant0.4$.[6]

Since the same particle parameters which appear in the theoretical expression for the shape factor are found in the expression for the β-γ directional correlation, a measurement of β-γ directional correlations in allowed β decay may help one to understand the observed shape-factor effects. Several attempts have been made to measure higher order effects in the β-γ directional correlation involving allowed β transitions. In most cases, the experimental accuracy limits the anisotropy to less than 0.1% except for Na^{22}, Co^{56}, Mn^{56}, and Tb^{160}, where anisotropies up to a few percent have been

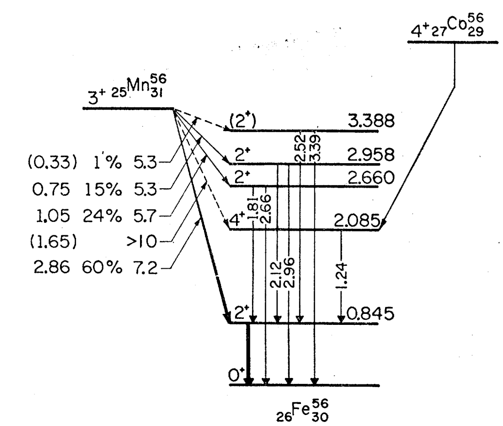

FIG. 1. Decay of Mn^{56}.

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[1] *Nuclear Data Sheets*, compiled by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences–National Research Council, Washington, D. C.), NRC 59-4-51.
[2] M. Morita, Progr. Theoret. Phys. (Kyoto) Suppl. **26** (1963).
[3] T. Mayer-Kuckuk and F. C. Michel, Phys. Rev. **127**, 545 (1962).
[4] Y. K. Lee, L. W. Mo, and C. S. Wu, Phys. Rev. Letters **10**, 253 (1963).
[5] M. Gell-Mann, Phys. Rev. **111**, 362 (1958).
[6] For a summary of shape-factor measurements, see for instance Table I in Ref. 2 and C. S. Wu, in *Alpha-, Beta-, and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland Publishing Company, Amsterdam, 1965), Chap. XXIVA.

reported.[7–9] There is, however, considerable disagreement between the different reports and it is not certain whether or not the effect exists. For these reasons, we engaged in a careful investigation of the β-γ directional correlation in the decay of Mn^{56}.

Contrary to an earlier measurement of Mn^{56}, where an isotropic distribution was reported,[10] a relatively large positive asymmetry of 2.2% was observed by Lobashov and Nazarenko.[8] It is therefore interesting to note that when Howe, Langer, Spejewski, and Wortman[11] performed a very accurate measurement of the β spectrum they found the distribution to be essentially statistical, and thus found no evidence for a contribution from higher order terms to the shape factor. Nevertheless, the data presented by Howe *et al.* were such that these authors were also able to fit the spectrum with a shape factor of the form $C(W)=1+0.3/W$. If this deviation from statistical shape is caused by higher order matrix elements, the β-γ directional correlation should show a small anisotropy. But the magnitude of this effect reported in Ref. 8 is rather surprising.

II. EXPERIMENTAL PROCEDURE AND RESULTS

Mn^{56} was prepared by irradiating metallic manganese with thermal neutrons for an average of 10 min. Sources were then prepared by one of two methods: (1) vacuum evaporation of the radioactive metal onto 2 mg/cm^2-thick nickel foils; (2) dissolving the radioactive metal in HCl and drying a drop of the solution on aluminized Mylar films for an average source strength in both cases of 0.25 mCi. The source diameter was 1 cm or less and the source thickness was 0.2 mg/cm^2 or less.

The measurements were performed with an automated scintillation spectrometer described earlier.[12] The γ detector consisted of a 3-in.×3-in. NaI(Tl) Harshaw integral line scintillator at a distance of 10 cm from the source with a surrounding lead shield and sufficient low-Z absorbing material in front of the crystal to stop 3-MeV β rays. The source was placed in an aluminum vacuum chamber (lined with Lucite to minimize scattering of the β rays) 7 cm from the face of a 0.6-in.-thick Pilot B plastic scintillator optically coupled to a photomultiplier. The chamber was provided with an aluminum shield thick enough to stop the β particles and which could be lowered between the source and the β scintillator to determine the γ-γ coincidence background. The electronics consisted of a Hamner transistorized modular fast-coincidence system with two β and one γ energy channels. Three coincidence modules were used simultaneously with resolving times (2τ) of 18, 28, and 40 nsec. This allowed the measurements to be taken at two beta energy ranges and at different true-to-chance ratios for a check on the accuracy of the chance correction.

Measurements were taken with the γ detector at 90°, twice at 180°, and at 270° (with respect to the β counter direction) and in the reverse order for 5 min at each point for an average of 28 points per run. Altogether, 80 runs were taken with the γ counter starting at any one of the three angular positions. The number of coincidences, γ singles, and β singles were recorded and transcribed onto IBM cards for each data point. The chance count was computed for each data point from the product of the β and γ singles with the resolving time (determined by the two-source method). The chance corrected coincidence counts were corrected for decay and source asymmetry (which averaged to be less than 0.1%) by dividing by the corresponding γ singles count. The asymmetry was calculated for each pair of data points as

$$A_{\beta\gamma}=[N_{\beta\gamma}(180°)/N_{\beta\gamma}(90° \text{ or } 270°)]-1\,,$$

where $N_{\beta\gamma}(\theta)$ is the coincidence rate at the angle θ. The over-all asymmetry was obtained from the individual asymmetries in two ways: First, an average asymmetry was calculated for each run separately and these were then averaged for all runs; second, all runs were combined and the average was taken. Both methods gave the same result. The errors quoted are the probable errors computed from the standard deviation.

The measurements on the 2.86-MeV β transition in Mn^{56} (see Fig. 1) were taken at average energies of 1.3 and 2.3 MeV with energy ranges of 300 and 700 keV, respectively. Approximately 10^6 true coincidences were accumulated at each of the γ counter positions for each energy range. The lower energy range was chosen to have a γ-γ correlation background large enough to allow a good measurement of its effect.

In order to measure the asymmetry of this background, an absorber was placed in front of the β detector and the measurements were repeated for several runs. To measure the extent to which the γ-γ background contributed to the measured β-γ correlation, the coincidence rate was measured several times alternately with the shield in place and with the shield removed, other experimental conditions remaining the same. This measurement was repeated for different positions of the γ detector and at the beginning of each β-γ correlation run. With the discriminator in the β channel set to accept pulses corresponding to the lower energy range ($1.10\leq E_\beta\leq 1.40$ MeV), the γ-γ coincidence background was 5% with an asymmetry $A_{\gamma\gamma}$

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[7] Z. W. Grabowski, R. S. Raghavan, and R. M. Steffen, Phys. Rev. **139**, B24 (1965).
[8] V. M. Lobashov and V. A. Nazarenko, Zh. Eksperim. i Teor. Fiz. **42**, 370 (1962) [English transl.: Soviet Phys.—JETP **15**, 257 (1962)].
[9] B. G. Pettersson, J. H. Hamilton, and J. E. Thun, Nucl. Phys. **22**, 131 (1961).
[10] M. Walter, O. Huber, and W. Zunti, Helv. Phys. Acta **23**, 697 (1950).
[11] D. A. Howe, L. M. Langer, E. H. Spejewski, and D. E. Wortman, Phys. Rev. **128**, 2748 (1962).
[12] H. J. Fischbeck and R. W. Newsome, Jr., Phys. Rev. **129**, 2231 (1963).

TABLE I. Directional-correlation results.

Energy range E(MeV)	Asymmetry[a] $A_{\beta\gamma}\times10^2$	Gamma-gamma asymmetry[a] $A_{\gamma\gamma}$	$N_{\gamma\gamma}/(N_{\gamma\gamma}+N_{\beta\gamma})$	Correlation coefficient[b] $A_2\times10^2$
		Mn^{56}		
1.10 –1.40	$+1.64\pm0.13$	$+0.329\pm0.006$	0.0522 ± 0.0011	-0.10 ± 0.15[c]
2.16 –2.86	-0.33 ± 0.18	$+0.1$	0.0033	-0.30 ± 0.11
2.16 –2.86	-0.45 ± 0.10			
		I^{131}		
0.400–0.500	$+0.01\pm0.03$	no interference		<0.05
0.500–0.606	$+0.02\pm0.07$			

[a] As measured, without corrections.
[b] Corrected for solid angle.
[c] Corrected for gamma-gamma background according to Eq. (2).

$=+0.329\pm0.008$; at the higher energy setting ($2.16\leqslant E_\beta\leqslant 2.86$ MeV), where only the weak 2.52-MeV γ transition contributes, the γ-γ coincidence background was 0.3% and $A_{\gamma\gamma}\approx+0.1$. The accuracies of the measured γ-γ asymmetries were limited by the low counting rates, particularly in the higher energy range. However, the measured β-γ asymmetry was positive for the lower energy range with the strong γ-γ interference and negative for the higher energy range (see Table I, column 2). Any correction of a negative β-γ asymmetry for a positive γ-γ background asymmetry tends to make the true β-γ asymmetry more negative. From the correction formula

$$A_{\beta\gamma}(\text{true})=[1/(1-s)][A_{\beta\gamma}(\text{measured})-sA_{\gamma\gamma}], \quad (2)$$

with

$$s=N_{\gamma\gamma}/(N_{\beta\gamma}+N_{\gamma\gamma}),$$

where $N_{\gamma\gamma}$ is the coincidence rate with the absorber in front of the β counter, we see that for $A_{\gamma\gamma}(\bar{E}=2.3\text{ MeV})\approx0.1$, $s\approx0.003$, and an assumed $A_{\beta\gamma}$(true) of zero, i.e., an isotropic β distribution, the measured asymmetry should be positive: $A_{\beta\gamma}(\text{measured})\approx+0.3\times10^{-3}$. The resulting γ-γ background correction, $sA_{\gamma\gamma}\approx+0.3\times10^{-3}$, is within the experimental uncertainty of the measured β-γ asymmetry at the upper energy and is therefore neglected. Similarly, interference from internal bremsstrahlung was considered negligible for the energy regions investigated.[13]

In order to be certain that the asymmetry measured was not a systematic error, a measurement on the allowed 0.606-MeV β–0.364-MeV γ cascade in I^{131} (see Fig. 2) was performed after 40 runs had been taken on Mn^{56}. Again, two β energies were selected: $400\leqslant E_\beta\leqslant500$ and $500\leqslant E_\beta\leqslant600$ keV. All other experimental conditions were the same as for Mn^{56}. A total of 10^7 true coincidences were accumulated in this measurement which gave the vanishingly small asymmetries of $+0.0001\pm0.0003$ and $+0.0002\pm0.0007$ for the two energy ranges, respectively. The measurement on Mn^{65} was then continued for the upper energy range for an additional 5×10^5 coincidences per angle. The result of this second set of measurements is in agreement with that of the first.

We conclude that the small negative correlation observed at the higher energy range in Mn^{56} is caused by a nonisotropic β distribution and that the effect is not due to a γ-γ correlation background or to misalignment of the apparatus. From the measured asymmetry, which was corrected for solid-angle effects ($Q_2=0.861$) and for γ-γ background at the lower energy, the directional correlation coefficient A_2 was calculated (see Table I). The value given at the lower energy serves mainly as a check of consistancy, since a small β-γ anisotropy would be completely masked by the strong γ-γ interference. Nevertheless, it is reassuring that the value obtained is not in disagreement with the result obtained at the upper energy.

III. DISCUSSION

The directional correlation coefficient $A_2(W)$ defined in

$$N(\theta,W)=1+A_2(W)P_2(\cos\theta) \quad (3)$$

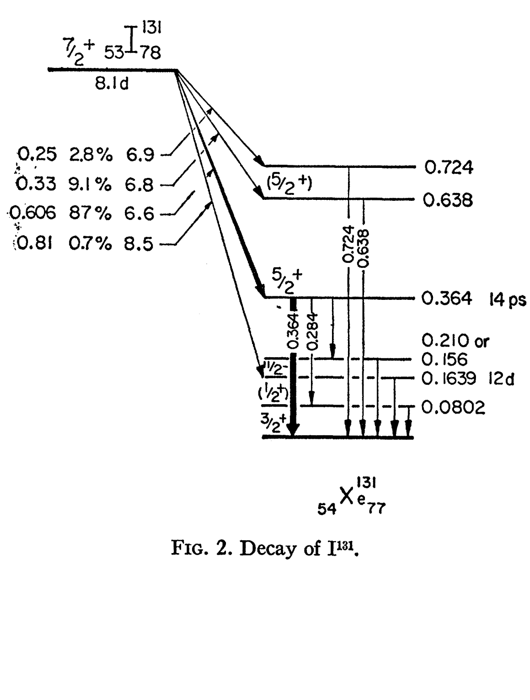

FIG. 2. Decay of I^{131}.

[13] P. Bolgiano, L. Madansky, and R. Rasetti, Phys. Rev. **89**, 679 (1953).

for a $3^+(\beta)2^+(\gamma)0^+$ transition is given by

$$A_2=(\tfrac{1}{6})^{1/2}b_{11}^{(2)}/7C, \tag{4}$$

where $C=-(\frac{1}{3})^{1/2}b_{11}^{(0)}$ is the spectrum shape factor.[14] The particle parameters $b_{11}^{(2)}$ and $b_{11}^{(0)}$ can be expressed [see Eqs. (2) and (3) of Ref. 14] in terms of the ratios of the interfering second-forbidden matrix elements to the Gamow-Teller matrix element:

$$x=\int i\gamma_5\mathbf{r}\Big/\int\boldsymbol{\sigma},\qquad u=\frac{C_V}{C_A}\int\boldsymbol{\alpha}\times\mathbf{r}\Big/\int\boldsymbol{\sigma},$$
$$y=\int(\boldsymbol{\sigma}\cdot\mathbf{r})\mathbf{r}\Big/\int\boldsymbol{\sigma},\qquad v=\int\boldsymbol{\sigma}r^2\Big/\int\boldsymbol{\sigma}, \tag{5}$$

and one obtains

$$A_2=\frac{1}{7}\frac{L_{12}[\frac{1}{3}q(v+y)+2x-u]+N_{21}(3y-v)}{L_0[1-\frac{2}{3}q(\frac{1}{2}qv+x-u)]+N_0[\frac{2}{3}q(2y-v)+2(x+u)]}, \tag{6}$$

where q is the neutrino momentum and L_0, N_0, L_{12}, and N_{21} are combinations of electron-wave-function components.[14] Terms involving second-rank tensors have been neglected. In principle, one has four unknown parameters x, y, u, and v in the above expression. However, certain relationships between the matrix elements allow a considerable simplification. Assuming uniform nuclear matter and the CVC theory, one obtains in the nonrelativistic limit[14]

$$x\approx\frac{1}{2M}-\frac{3}{20}\Lambda\alpha ZR\eta,\qquad u\approx\frac{C_V}{C_A}\frac{4.7}{M},$$
$$y\approx\tfrac{3}{5}R^2\eta,\qquad v\approx\tfrac{3}{5}R^2. \tag{7}$$

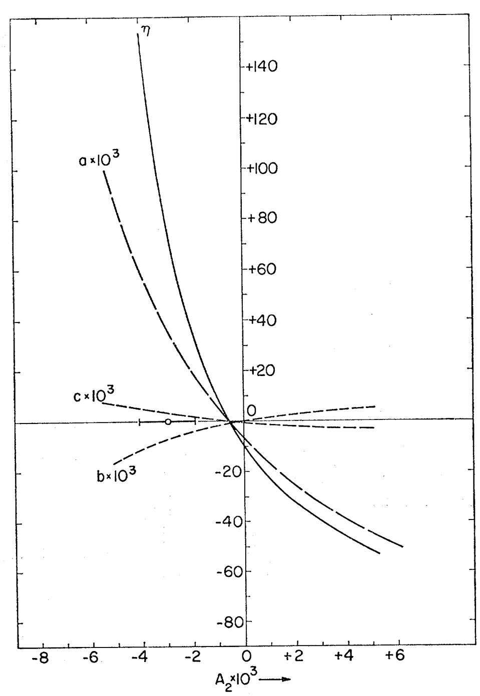

FIG. 3. Plot of the nuclear parameter η and the spectrum shape coefficients a, b, and c versus the directional correlation coefficient A_2.

[14] M. Morita, Phys. Rev. **113**, 1584 (1959).

The parameter Λ is of the order of unity and given by $\Lambda\approx1+(W_0-2.5)A^{1/3}Z^{-1}$. The only free parameter left is the shell-model-dependent factor η, which may be considerable larger than one. The nuclear mass M is 1836, the ratio of the coupling constants is taken as $C_A/C_V=-1.2$, and the nuclear radius as

$$R=3.1\times10^{-3}A^{1/3}(\hbar/m_0c).$$

Since the inclusion of finite nuclear size in the electron wave functions makes it impossible to express L_0, N_0, N_{21}, and L_{12} as explicit functions of the β-ray energy, the approximations given by Morita[14] including terms up to $(\alpha Z)^2$ have been used. The usual Konopinski-Uhlenbeck approximation cannot be used in this case since it gives expressions for A_2 and C which differ by several percent from the more nearly exact expressions which take into account finite de Broglie wavelength effects. By making use of Eq. (7), Eq. (6) can be solved for the parameter η in terms of the angular correlation coefficient and the β-ray energy. The numerical result is shown in Fig. 3, where a plot of η versus A_2 is given for a β energy of $5.5(m_0c^2)$. By grouping the terms in the shape factor

$$\begin{aligned}C(W)&=L_0[1-\tfrac{2}{3}q(\tfrac{1}{2}qv+x-u)]\\&\qquad+N_0[\tfrac{2}{3}q(2y-v)+2(x+u)]\\&=k(1+aW+b/W+cW^2)\end{aligned} \tag{8}$$

according to their dependence on the β energy, the coefficients a, b, and c can be obtained as functions of η and, therefore, as functions of A_2. These parameters are also shown in Fig. 3. As can be seen from this figure, a positive coefficient $b\approx0.3$ in the shape factor cannot be reconciled with the very small negative asymmetry observed. But the order of magnitude of the a coefficient which has been observed in low-energy allowed transitions is in agreement with a small negative anisotropy. It seems, therefore, unlikely that a b/W term with $b\approx0.3$ in the shape factor in an allowed β decay could be caused by higher order effects unless this phenomenon is accompanied by a very large anisotropy in the β-γ angular correlation. As can be seen from Fig. 3, a shape factor consistent with an angular correlation coefficient $A_2 \approx -0.003$ should be of the form $C(W) \approx 1+0.04W-0.01/W$. This energy dependence may be too small to be experimentally observed in the limited energy range available for an accurate measurement.

It is, of course, important to recognize that the above conclusions are based on the assumption that the decay of Mn^{56} as shown in Fig. 1 is essentially correct. Numerous experiments on the γ spectrum of Fe^{56} (see Ref. 1) have established the spins and parities of the observed levels. It is, however, puzzling why the β transition to the 2.085-MeV level in Fe^{56} from the Mn^{56} ground state is so strongly hindered ($\log ft > 10$).[15,16] According to the accepted decay scheme this should be an allowed transition. If, on the other hand, the ground state of Mn^{56} were 2^+, this transition would be second forbidden, while all other β transitions to Fe^{56} could still be characterized as allowed. However, a measurement of the Mn^{56} ground-state spin by Childs, Goodman, and Kiefer[17] can be reconciled only with a value of $I=3$.

ACKNOWLEDGMENTS

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[15] J. K. Bienlein and H. Dinter, Nucl. Phys. **55**, 113 (1964).
[16] H. J. Fischbeck, W. M. Greenberg, and E. B. Arnold, Bull. Am. Phys. Soc. **10**, 93 (1965).
[17] W. J. Childs, L. S. Goodman, and L. J. Kiefer, Phys. Rev. Letters **1**, 296 (1958); Phys. Rev. **122**, 891 (1961).

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Fission Yields of ^{156}Eu, ^{157}Eu, ^{158}Eu, and ^{159}Eu†*

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The cumulative yields of ^{156}Eu, ^{157}Eu, ^{158}Eu, and ^{159}Eu from thermal-neutron fission of ^{235}U and $\approx$25-MeV ^{3}He fission of ^{238}U were determined. The thermal yields are: ^{156}Eu, $0.0125\pm0.0010\%$; ^{157}Eu, $0.0060\pm0.0007\%$; ^{158}Eu, $0.0031\pm0.0006\%$; ^{159}Eu, $0.0011\pm0.0003\%$. The ^{3}He yields are: ^{156}Eu, $0.27\pm0.07\%$; ^{157}Eu, $0.13\pm0.02\%$; ^{158}Eu, $0.089\pm0.015\%$; ^{159}Eu, $0.057\pm0.010\%$.

I. INTRODUCTION

THE thermal-fission yields of the europium nuclides are of interest because they lie near the upper limit of fission product production in both mass number and atomic number, and help to define the high side of the fission mass-yield curve.

Winsberg[1] has determined the yields of ^{157}Eu and ^{158}Eu from the thermal-neutron fission of ^{235}U. Recently, Bunney and Scadden[2] have measured the yields of ^{157}Eu and ^{159}Gd from thermal-neutron fission of ^{233}U and ^{235}U.

In the present study, the cumulative yields of 15.15-h ^{157}Eu,[3] 45.7-min ^{158}Eu,[3] and 17.9-min ^{159}Eu[3] from thermal-neutron fission of ^{235}U and $\approx$25-MeV ^{3}He fission of ^{238}U were determined radiochemically. The europium fraction was isolated chemically, and the activities were related to that of ^{99}Mo. The yields of ^{158}Eu and ^{159}Eu relative to ^{157}Eu for $\approx$28-MeV ^{4}He fission of ^{238}U were also determined.

II. EXPERIMENTAL

A. Irradiations

Thermal-neutron irradiations of 93% ^{235}U (as metal foil or uranyl nitrate) were performed in the thermal-column rabbit port of the Los Alamos Omega West Reactor at 5 MW or in the rabbit port of the Los Alamos Water Boiler at 25 kW. The cadmium ratios for thin indium foils are $\approx$25 and $\approx$3, respectively. The target material was placed in a quartz tube inside either an aluminum rabbit (Omega West) or a nylon rabbit (Water Boiler) for irradiation.

The 25-MeV ^{3}He and 28.0- to 28.7-MeV ^{4}He irradiations of 3-mil, 99.9% ^{238}U foils were performed in the external center beam tube of the Los Alamos cyclotron. The foils were covered with 0.5-mil aluminum foil or 0.4-mil molybdenum foil during the irradiations. In the ^{3}He irradiation, the average energy at which fission occurred in the essentially infinitely thick uranium foil was estimated to be $\approx$24 MeV.

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* This paper includes a portion of the dissertation presented to the University of New Mexico by W. R. Daniels in partial fulfillment of the requirements for the degree of Doctor of Philosophy, 1964.
[1] L. Winsberg, in *Radiochemical Studies: The Fission Products*, edited by C. D. Coryell and N. Sugarman (McGraw-Hill Book Company, Inc., New York, 1951), Natl. Nucl. Energy Ser., Div. IV, Vol. 9, pp. 1292, 1302.
[2] L. R. Bunney and E. M. Scadden, J. Inorg. Nucl. Chem. **27**, 273 (1965).
[3] W. R. Daniels and D. C. Hoffman (to be published).