m; four of these are  $|m| = \frac{1}{2}$  and the two remainders are  $|m| = \frac{3}{2}$ .

When the outgoing partial waves other than s wave are also contributing to the reaction, the fraction  $f_{1/2}$ <br>will decrease, because substates with  $|m| > \frac{1}{2}$  will also be populated independently of whether or not the neutrons undergo spin flip.

The relation between the fraction  $f_{1/2}$  or  $f_{3/2}$  and the spin-flip probability  $P_f$  will now be considered more quantitatively. Since only  $|m| \leq \frac{3}{2}$  can be populated when  $m_{\nu} = 0$ , the relation

$$
f_{1/2} + f_{3/2} = 1 \tag{A3}
$$

must be true. In the above discussion, it was noted that when  $P_f = 0$ , then  $f_{1/2} = 1$  and  $f_{3/2} = 0$ , and when  $P_f = 1$ , then  $f_{1/2} = \frac{1}{3}$  and  $f_{3/2} = \frac{2}{3}$ . Thus the expression for  $P_f$  is

$$
P_f = \frac{3}{2} f_{3/2} = \frac{3}{2} (1 - f_{1/2}). \tag{A4}
$$

The probability for no spin flip  $P_0$  is of course related to  $P_f$  through the normalization  $P_0+P_f=1$ .

PHYSICAL REVIEW VOLUME 173, NUMBER 4 20 SEPTEMBER 1968

# Total Internal-Conversion Coefficients for Low-Energy E2 Transitions in Ra<sup>224</sup>, Th<sup>228</sup>, U<sup>234</sup>, U<sup>236</sup>, and Pu<sup>240</sup><sup>†</sup>

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The total internal-conversion coefficients have been measured for  $E2$  transitions from first excited state to ground state in five heavy deformed nuclei,  $\alpha$ -particle gating was used to determine the population of the first excited levels from parent decays. Photon spectra, as observed by a well-type NaI(Tl) detector in coincidence with the gating  $\alpha$  particles, were analyzed to determine the ratio of  $\gamma$  transitions to total transitions. The  $\alpha$  gating spectra and photon spectra were analyzed on a computer, when necessary, using a nonlinear least-squares curve-fitting program. The resulting coefficients are  $19.6 \pm 1.4$  for the 84.4-keV transition in Ra<sup>224</sup>, 158 $\pm$ 7 for the 57.6-keV transition in Th<sup>228</sup>, 780 $\pm$ 55 for the 43.5-keV transition in U<sup>224</sup>  $607\pm29$  for the 45.28-keV transition in U<sup>236</sup>, and 845 $\pm$ 93 for the 42.88-keV transition in Pu<sup>240</sup>. The internalconversion coefficient for the transition in Pu<sup>240</sup> is somewhat lower than the theoretical predictions of Hager and Seltzer and of Sliv and Band, whereas the internal-conversion coefficient for the transition in U<sup>234</sup> is somewhat higher. The internal-conversion coefficients for the remaining transitions agree with the theoretical values within the experimental uncertainties.

#### L. INTRODUCTION

IN recent years many internal-conversion coefficient  $\blacksquare$  measurements have been made for pure  $E2$  transitions in the rare-earth deformed nuclei. Although large internal-conversion anomalies werc initially reported<sup>1-6</sup> for some of these transitions, the present trend is for the E-shell and total internal-conversion coefficients to agree with the theoretical calculations.  $6.7$ On the other hand, various  $L_1$ -subshell ratios<sup>8-11</sup> and

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- version Processes, edited by J. H. Hamilton (Academic Press Inc.,
- New York, 1966), p. 225.<br>
<sup>7</sup>R. S. Dingus, W. L. Talbert, Jr., and M. G. Stewart, Nucl.<br>
Phys. 83, 545 (1966).<br>
<sup>9</sup> J. H. Hamilton, Phys. Letters 20, 32 (1966).<br>
<sup>9</sup> P. Erman, G. T. Emery, and M. L. Perlman, Phys. Rev. 147
- 
- 858 (1966).<br>\_ <sup>10</sup> S. E. Karlsson, I. Andersson, Ö. Nilssen, G. Malmster
- 
- C. Nordling, and K. Siegbahn, Nucl. Phys. 89, 513 (1966).<br>
<sup>11</sup> W. Gelletly, J. S. Geiger, and R. L. Graham, Phys. Rev.<br>
157, 1043 (1967).

 $K$ - and  $L$ -shell particle parameters<sup>12</sup> have been reported which do not agree with theory. Since nuclear-structure effects are expected to be unimportant for these transitions, no apparent reason for these deviations exists within the framework of the present theory.<sup>13-15</sup>

In contrast to the E2 transitions in the rare-earth nuclei, the E2 transitions that occur in even-even nuclei in the heavy-element deformed region,  $A > 220$ , have not been extensively studied. These transitions arc characterized by low transition energies, normally less than 100 keV, and extremely large internal-conversion coefficients, typically greater than 100. Accurate  $L$ -subshell ratios for two of these  $E2$  transitions have been reported recently<sup>16</sup> but no total internal-conversion coefficients have been measured with uncertainties less than about  $9\%$ . Thus, an accurate comparison with theory has not been possible for the total internalconversion coefficients.

- <sup>12</sup> H. M. Nasir, Z. W. Grabowski, and R. M. Steffen, Phys. Rev. 162, 1118 (1967).<br>
<sup>18</sup> E. L. Church and J. Weneser, Ann. Rev. Nucl. Sci. 10, 193
- (1960).<br>
<sup>14</sup> C. P. Bhalla, Phys. Rev. **157**, 1136 (1967).<br>
<sup>15</sup> J. J. Matese, Bull. Am. Phys. Soc. **13**, 78 (1968).<br>
<sup>16</sup> J. H. Hamilton, B. van Nooijen, A. V. Ramayya, and W. H.
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- 
- Brantley, in *Internal Conversion Processes*, edited by J. H. Hamil-<br>ton (Academic Press Inc., New York, 1966), p. 541.

f Work was performed in the Ames Laboratory of the U. S. Atomic Energy Commission. Contribution No. 2273. 'F, K. McGowan and P. H. Stelson, Phys. Rev. 107, <sup>1674</sup>

<sup>(1957).&</sup>lt;br>
<sup>2</sup> B. N. Subba Rao, Nuovo Cimento 17, 189 (1960).<br>
<sup>2</sup> E. M. Bernstein, Phys. Rev. Letters 8, 100 (1962).<br>
<sup>4</sup> D. B. Fossan and B. Herskind, Phys. Letters 2, 155 (1962).<br>
<sup>5</sup> D. B. Fossan and B. Herskind, Nucl.



FIG. 1. Partial-decay scheme of Pu<sup>240</sup>.

The purpose of this investigation has been to measure accurately the total internal-conversion coefficients of the  $E2$  ground-state transitions in Ra<sup>224</sup>, Th<sup>228</sup>, U<sup>234</sup>, U<sup>236</sup>, and Pu<sup>240</sup>. All previous total internal-conversion coefficient measurements for transitions such as these, reviewed by Stelson in Ref. 17, were made using Coulomb-excitation and lifetime-determination techniques and show uncertainties as large as  $20\%$ . In this investigation, by using scintillation techniques and careful data analysis, uncertainties as low as  $5\%$  were obtained.

The partial-decay scheme of  $Pu^{240}$ , typical of these heavy deformed even-even nuclei, is shown in Fig. 1.'8 Since the  $\alpha$ -particle intensity decreases rapidly with decreasing  $\alpha$ -particle energy, the only excited state in  $U^{236}$  that is significantly populated is the 2<sup>+</sup> state at 45 keV. Because of the simplicity of the decay schemes of these deformed nuclei, accurate measurements of some total internal-conversion coefhcients of E2 ground-state transitions, typified by the  $45$ -keV transition in U<sup>236</sup>, are possible and are reported in this work.

### II. EXPERIMENTAL INVESTIGATION

#### A. General Procedure

The total internal-conversion coefficient was obtained from the ratio of the total number of observed transitions to the number of observed  $\gamma$  rays,  $N_0/N_{\gamma}$ . This ratio is related to the total internal-conversion coefficient  $\alpha$  by the relation

$$
\alpha + 1 = (N_0/N_\gamma)\epsilon_\gamma,\tag{1}
$$

where  $\epsilon_{\gamma}$  is the  $\gamma$ -ray detection probability. Photons from a transition of interest were detected in a NaI(Tl) scintillation spectrometer producing pulses which were accumulated in a multichannel analyzer gated by the detection of an  $\alpha$  particle directly populating the upper level of the transition. The total number of observed transitions,  $N_0$ , was equal to the number of gate pulses sent to the analyzer analog-to-digital converter (ADC) gate, and the number of  $\gamma$  rays detected,  $N_{\gamma}$ , was given by the number of counts contained in the  $\gamma$ -ray response curve in the photon pulse-height spectrum. The photon efficiency could be accurately calculated for a given source and detector configuration.

The techniques used in this experiment were an extension of the method developed by Dingus et al.<sup>7</sup> In particular,  $N_0$  was determined using a summing technique in which the gating pulses were not only sent to the analyzer ADC gate but also to a linear adding circuit. In this circuit the gating pulses acted as a pedestal to which the pulses from the photon detector were added before being sent to the ADC of the analyzer. The observed photon spectrum was shifted by the number of channels corresponding to the height of the pedestal pulse and a pedestal peak was produced in the channel corresponding to zero energy for the photon spectrum. (The pedestal peak was present because a  $\gamma$ ray or x ray was not detected every time the ADC gate was opened.) Thus the total number of observed transitions,  $N_0$ , was the number of times the gate was opened when the analyzer was able to accept and process a pulse and was given by the total number of counts in the photon spectrum, including the pedestal peak. This technique eliminated the dead-time correction necessary if  $N_0$  were the total number of gate pulses sent to the ADC gate since the gate could not be opened when the analyzer was busy processing a previous event.

The electronics associated with collecting the gated spectra are shown in the block diagram in Fig. 2. The



FIG. 2. Block diagram of equipment used for internalconversion coefficient measurements.

<sup>&</sup>lt;sup>17</sup> P. H. Stelson, in *Internal Conversion Processes*, edited by J. H. Hamilton (Academic Press Inc., New York, 1966), p. 213.<br><sup>18</sup> E. K. Hyde, I. Perlman, and G. T. Seaborg, *The Nuclear* 

Properties of the Heavy Elements (Prentice-Hall, Inc., Englewoo Cliffs, N. J., 1964), Vol. II.

pileup rejection circuit was designed specifically for the output of a double delay-line amplifier and is described in detail in Ref. 19. The linear amplifier of the multichannel analyzer was used for timing and pulse-shaping purposes.

The NaI(Tl) detector was a 2.75-in.-diam $\times$ 3.00-in.thick crystal manufactured by Harshaw Chemical Company. The crystal had a 1.75-in.-deep $\times$ 0.31-in.diam well which was lined with 0.015 in. of beryllium. The crystal-well surface was highly polished in order to reduce the response below the photopeak at low photon energies. '0 The advantages of this type of detector were that the  $\gamma$ -ray detection efficiency could be accurately calculated and that the iodine  $K$  x-ray escape peak was substantially reduced.

The sources, each with a source strength of approximately 0.3  $\mu$ Ci, were made on the Argonne National Laboratory isotope separator. The source backing was a thin disk of aluminized Mylar,  $0.200$  in. diam $\times 0.00025$ in. thick, on which the source material was deposited within a circle of radius 0.020 in. centered on the aluminum side of the Mylar. Since the isotope-separator ion beam was decelerated to an energy of 500 eV before striking the target, these sources were quite thin and no degradation in  $\alpha$  resolution due to the penetration of the ions into the Mylar backing was noticed. The sources were placed at the bottom of closed-end Mylar cylinders made from 0.005-in. Mylar. The diameters of the cylinders were such that they would just fit inside the NaI(Tl) detector well. The  $\alpha$  detector was positioned approximately 0.125 in. above the source and the assembly was lowered into the detector well so that the source was positioned at the bottom of the well. A complete description of the source and detector assembly is given in Ref. 19.

The silicon surface-barrier  $\alpha$  detectors were manufactured by R.C.A., Ltd. of Montreal, Canada. These detectors, each of dimension 0.090 in. $\times$ 0.090 in. and 50  $\mu$  thick (with an active area of 0.050 in. $\times$ 0.050 in.), were mounted on  $0.200$ -in.-diam $\times$ 0.040-in.-thick copper disks to which the detector signal leads were fastened. The signal leads supported the detector in the NaI(TI) detector well. The best resolution full width at half-maximum (FWHM) obtained with these detectors at room temperature was 17.5 keV, with the source and detector in an evacuated chamber. Unfortunately, the detector well could not be evacuated because of the fragile beryllium well lining; thus an atmosphere having a low electron density was desirable to minimize degradation of the  $\alpha$  resolution. The best practical environment was helium gas, in which a resolution of 23 keV was obtained for a source-to-detector distance of 0.125 in. ; a helium gauge pressure of  $1-2$  in. of water was continually maintained in the detector well during data accumulation.

## <sup>20</sup> J. S. Eldridge and P. Crowther, Nucleonics 22, 56 (1965).

#### B.  $\alpha$ -Spectra Analysis

The pulse-height spectrum from the detection of monoenergetic  $\alpha$  particles by these  $\alpha$  detectors was an asymmetrical peak with a low-energy tail extending below the full-energy peak. The presence of this tail in the single-channel analyzer window resulted in the detection of  $\alpha$  particles from the high-intensity  $\alpha$  group populating the  $0^+$  ground state of the daughter. Since no transitions were in coincidence with the ground state, a correction to the pedestal peak was required as a result of the tail of the ground-state  $\alpha$  group. In order to perform this correction, a detailed analysis of the  $\alpha$ pulse-height spectrum was necessary. In describing the analysis, the  $\alpha$  group to the ground state and the  $\alpha$  group to the 2<sup>+</sup> state will be referred to as the  $\alpha_0$  group and the  $\alpha_2$  group, respectively.

The entire photon spectrum from the NaI(T1) detector was used to gate the  $\alpha$  detector, thus eliminating the  $\alpha_0$  group from the pulse-height distribution, and the  $\alpha$ detector response to the  $\alpha_2$  group was obtained. This response was subtracted from the singles  $\alpha$  spectrum to determine the amount of tailing from the  $\alpha_0$  peak under the  $\alpha_2$  peak. The procedure required a nonlinear leastsquares computer analysis in which the response function was the sum of a Gaussian peak and three non-Gaussian peaks. Each non-Gaussian peak, described by nine parameters, was a very general function that could assume many diferent shapes with the proper choice of parameters. No attempt will be made to describe this program because it has been adequately described elsewhere.<sup>7</sup> Since this analysis was very similar for all the isotopes studied, a description will be given only for the decay of Pu<sup>240</sup> leading to levels in  $U^{236}$  $(Fig. 1)$ .

A gated  $\alpha$  spectrum from the decay of Pu<sup>240</sup> and the resulting computer fit are shown in Fig. 3. The largest peak, a pure Gaussian, represents the full-energy peak while the low-energy tail is seen to the left of the Gaussian. The small non-Gaussian peak under the tail shows that the function being used to fit the tail does not have quite enough structure to provide a good fit over this region of the spectrum. The origin of the small peak above the Gaussian is unclear. This peak was present even for the  $\alpha_0$  group, with which no conversion electrons are coincident. Furthermore, the height of this peak decreased about a factor of 10 when the  $\alpha$ spectrum was observed using an evacuated test chamber and the same source-to-detector distance. Hence, summing with conversion electrons cannot account entirely for the origin of this peak, although about  $10\%$ of the peak could result from such summing.

The corresponding singles  $\alpha$  spectrum from the Pu<sup>240</sup> decay is shown in Fig. 4. In the computer analysis the shape of the  $\alpha_2$  peak was held constant while its amplitude was allowed to vary. By allowing various parameters of the  $\alpha_0$  peak to vary also, a fit was obtained which yielded the correct absolute intensity of the  $\alpha_2$  group,

<sup>&#</sup>x27; C. L. Duke and W. L. Talbert, Jr., U. S. Atomic Energy



 $24.4\%$ .<sup>21</sup> Since the position of the single-channel analyzer window across the  $\alpha_2$  peak was known, the relative contribution of the  $\alpha_0$  low-energy tail in the gating channels could be accurately determined by this analysis. The single-channel analyzer window, channels 100–165 for the spectrum illustrated in Fig. 4, included a known fraction of the high-energy shoulder of the  $\alpha_2$ peak. Thus the fraction of the  $\alpha_2$  peak not within the single-channel analyzer window because of summing with conversion electrons could be estimated. For the worst cases ( $U^{236}$  and  $Pu^{240}$ ), the resulting corrections increased the values of the conversion coefficients by about  $0.5\%$ .

At the beginning of each data run, a gated  $\alpha$  spectrum was obtained. Singles  $\alpha$  spectra were subsequently taken at various times during the run. Computer analyses as described above were performed when necessary for these  $\alpha$  spectra; and the  $\alpha_2$ -to-total ratio, i.e., the



<sup>21</sup> L. N. Kondraev, G. I. Novikova, Iv. P. Sobolev, and L. L. Gol'din, Zh. Eksperim. i Teor. Fiz. 31, 771 (1956) [English transl.: Soviet Phys.—JETP 4, 645 (1957)].

number of gating counts contained in the  $\alpha_2$  peak to the total number of counts appearing within the gating channel limits, was determined.

## C. NaI(T1) Detector Efficiency

The efficiency  $\epsilon_{\gamma}(E)$  of the NaI(TI) crystal is defined as the probability that a  $\gamma$  ray of energy E, emitted from a source positioned along the axis of a cylindrical well-type NaI(T1) detector, will lose some energy in the crystal. The integral expression for  $\epsilon_{\gamma}(E)$  is

$$
\epsilon_{\gamma}(E) = \frac{1}{2} \int_{\theta_0}^{\pi} \exp[-\alpha_2(E)D_2(\theta) - \alpha_3(E)D_3(\theta)]
$$
  
 
$$
\times \{1 - \exp[-\alpha_1(E)D_1(\theta)]\}W(\theta) \sin\theta \,d\theta, \quad (2)
$$

where  $\alpha_1(E)$  is the total (minus coherent) attenuation coefficient of the crystal,  $\alpha_2(E)$  and  $\alpha_3(E)$  are the photoelectric absorption coefficients for the beryllium well lining and copper support disk, respectively, measured in the direction of polar angle  $\theta$  as shown in Fig. 5. In this figure, the actual silicon wafer is not shown since its effect on the absorption of  $\gamma$  rays was determined to be insignificant compared to the absorption by the copper disk. The factor  $W(\theta)$  is the probability that a  $\gamma$  ray will be emitted into solid angle  $d\Omega$  at polar angle  $\theta$ following  $\alpha$  emission along the direction  $\theta=0^{\circ}$ . To perform the integration, a computer program was used after modification to include  $W(\theta)$ . The original program has been compared to other efficiency calculations for a source positioned on top of the crystal and for a zero diameter well. The calculations agreed to within  $0.1\%$  for low-energy photons.<sup>7</sup>

For the

$$
0^+\rightarrow 2^+\rightarrow 0^+
$$

$$
\alpha \qquad \gamma
$$

cascades considered in this investigation,  $W(\theta)$  has the simple form<sup>22</sup>

$$
W(\theta) = 1 + A_2 P_2(\cos \theta) + A_4 P_4(\cos \theta). \tag{3}
$$

Unfortunately, the coefficients  $A_2$  and  $A_4$  have not been measured for all of the transitions studied in this investigation; however, because of the well-type geometry, the efficiency is a slowly varying function with respect to changes in  $A_2$  and  $A_4$ . Asseries of efficiency calculations at a given photon energy using sets of values for  $A_2$  and  $A_4$  from angular correlation measurements<sup>23</sup> of several

$$
0^+\rightarrow 2^+\rightarrow 0^+
$$

$$
\alpha \qquad \gamma
$$

cascades in even-even nuclei,  $A > 220$ , agreed to within 1%. The values of  $A_2$  and  $A_4$  were corrected by the



FIG. 5. Detector configuration used for efficiency calculation.

efficiency program for the finite size of both the  $\alpha$  detector and the source. The uncertainties associated with these efficiency calculations are discussed in detail in Ref. 19 and represent a statistical combination of uncertainties resulting from 6nite source and detector size (about  $1\%$ ), measurement of source-to-detector distance (about  $1\%$ ), and uncertainties in  $A_2$  and  $A_4$ (with resulting effect of about  $1\%$ ).

#### D. Gated Photon Spectrum Analysis

The analysis of the  $\gamma$ -ray spectrum in coincidence with the  $\alpha_2$  group was less involved than the  $\alpha$ -spectrum analysis. Only one  $\gamma$  ray, well separated from the L x-ray peaks in the pulse-height distribution, was gated into the spectrum. Although the photon spectrum was not complicated, several problems, normally unimportant, became critical in this investigation.

Since the internal-conversion coefficients for these transitions are very large, the  $\gamma$ -ray intensity was much less than the  $L$  x-ray intensity. For example, for the 45-keV transition in  $U^{236}$  the  $\gamma$ -ray count rate was only about  $0.4\%$  of the  $L$  x-ray count rate. Thus, primary pulse pileup (random summing of two pulses at the output of the linear amplifier) from the  $L$  x-ray pulses could severely distort the  $\gamma$ -ray photopeak. Assuming an L x-ray counting rate of 1000 counts/sec, and an amplifier output pulse width of 1  $\mu$ sec, primary pulse pileup would occur at a rate of  $0.1\%$  of the L x-ray count rate. By rejecting these distorted pulses with the pileup detector, the pileup fraction was reduced by a factor of 25 to  $0.004\%$ .<sup>19</sup> factor of 25 to  $0.004\%$ .<sup>19</sup>

Because of the low  $\gamma$ -ray counting rate, data were accumulated for as long as 10 days, depending on the isotope being studied, in order to obtain a statistically meaningful number of  $\gamma$ -ray events. Thus, electronic drifts could become sources of distortion of the photopeak. These drifts, minimized by careful temperature regulation, were measured by accumulating a reference

<sup>&</sup>lt;sup>22</sup> H. Frauenfelder and R. M. Steffen, in Alpha-, Beta-, and Gamma-Ray Spectroscopy, edited by K. Siegbahn (North-Hollan

Publishing Co., Amsterdam, 1965), Vol. 2, p. 997.<br><sup>23</sup> E. S. Murphy, U. S. Atomic Energy Commission Repor<br>No. ANL-6685, 1963 (unpublished).



 $\gamma$  spectrum every 8 h during data accumulation. In general, such drifts were small and resulted only in a slight broadening of the photopeak.

A typical gated photon spectrum, obtained by gating with the 5.123-MeV  $\alpha$  group feeding the 45-keV level in U<sup>236</sup>, is shown in Fig. 6. As can be seen, the  $\gamma$ -ray intensity is much smaller than the  $L$  x-ray intensity. The Gaussian peak shown superimposed on the  $\gamma$ -ray peak was obtained by fitting a Gaussian curve to the right side of the peak and extrapolating it to the left side. This method for determining the photopeak area should be very accurate because no events from the summing of two  $L$  x-ray pulses could occur above 44 keV; all  $L$ x-ray groups from  $U^{236}$  have energies less than 22 keV and higher-order summing can be ignored.

The ratio of counts in the photopeak to the total number of counts resulting from the detection of monochromatic photons (photopeak-to-total ratio) can be used to determine the number of detected  $\gamma$ -ray events  $N_{\gamma}$  from the number of counts in the photopeak. Using the data collected from  $W^{181}$ ,  $Dy^{159}$ ,  $Co^{57}$ , and Ce<sup>139</sup> sources, the photopeak-to-total ratio for this crystal with the same source geometry as used in the conversion coefficient measurements was determined to be  $0.925 \pm 0.016$  throughout the range 45-166 keV. Finally, as explained previously,  $N_0$  was the number of counts included in the photon spectrum plus pedestal peak. For the spectrum shown in Fig. 6,  $N_0/N_\gamma$  was  $1696 \pm 129.$ 

## E. Final Results

At least three gated photon spectra were accumulated for each transition and analyzed as discussed in the previous sections. From this analysis, an internalconversion coefficient for each spectrum was calculated from Eq.  $(1)$ ; the reported internal-conversion coeffi-

TABLE I. Quantities used to determine the internal-conversion coefficient of the  $45$ -keV transition in U<sup>236</sup>.

FIG. 6. Gated photon spectrum of  $U^{236}$ .

 $\alpha$ 





cient for the transition is the average of the internalconversion coefficients from the separate runs. The error assignments were obtained by standard statistical methods from individual uncertainties and are to be interpreted as standard deviations.

As a typical example, the quantities used to determine the internal-conversion coefficient of the 45-keV transition in U<sup>236</sup> are shown in Table I. All of the entries have been discussed previously with the exception of  $\langle \alpha_2/(\alpha_2+\alpha_0) \rangle_{\rm av}$ , which is an average of the various  $\alpha_2$ -to-total ratios for the gating channels obtained from the  $\alpha$  spectra for each data set. The efficiencies  $\epsilon_{\gamma}(45)$ differ because the distance between the source and the  $\alpha$  detector was changed for each set.

The final internal-conversion coefficients for the five transitions studied are shown in Table II. The analyses

TABLE II. Experimental total interval-conversion coefficients for E2 transitions in the heavy deformed nuclei.

<b>Nucleus</b>	$E2$ transition energy (keV)	Internal-conversion coefficient
$Ra^{224}$	84.4	$19.6 \pm 1.4$
Th <sup>228</sup>	57.6	$158 \pm 7$
T <sub>7234</sub>	43.50	$780 \pm 55$
T <sub>1236</sub>	45.28	$607 + 29$
$P_{11}^{240}$	42.88	$845 + 93$

of the U<sup>234</sup>, U<sup>236</sup>, and Pu<sup>240</sup> transitions were identical to the U<sup>236</sup> analysis previously described. The relative uncertainties for these three transitions increase as the values of the internal-conversion coefficients increase, a direct result of the larger uncertainty in  $N_0/N_\gamma$  as the  $\gamma$ -ray counting rate decreases.

The internal-conversion coefficient of the 59-keV transition in Th $228$  has a smaller uncertainty for two reasons. First, the  $\alpha$  groups from the decay of U<sup>232</sup> were well separated by the  $\alpha$  detector, and the 5.267-MeV  $\alpha_2$ group from  $U^{232}$  had a higher intensity  $(32\%)$  than the  $\alpha_2$  groups for the other isotopes. Thus a more accurate determination of  $\langle \alpha_2/(\alpha_2+\alpha_0) \rangle_{\rm av}$  was possible for this transition. Second, the internal-conversion coefficient for this transition was small enough so that very good counting statistics could be obtained for  $N_{\gamma}$ , thus lowering the uncertainty in  $N_0/N_\gamma$ .

The internal-conversion coefhcient of the 84.4-keV transition in Ra224 was complicated by the presence of the 5.684-MeV (94%) and the 5.447-MeV (5.5%)  $\alpha$ groups from the decay of  $Ra^{224}$  in equilibrium with the Th<sup>228</sup> parent decay. The low-energy tails from these  $\alpha$ groups in addition to the tail from the Th<sup>228</sup>  $\alpha_0$  group interfered with the gating and had to be considered in determining the  $\alpha_2$ -to-total ratio. In this case a computer fit was impossible because of the number of  $\alpha$ groups involved; however, since the major  $\alpha$  peaks were well separated in the  $\alpha$  pulse-height distribution, the  $\alpha_2$ -to-total ratio for the gating channels could be determined with an uncertainty of  $5\%$  by hand analysis. In addition to the usual correction to  $N_0$ , a small correction of less than  $0.3\%$  was necessary because of the detection of the 241-keV  $\gamma$  rays in coincidence with the 5.447-MeV  $\alpha$  group from the Ra<sup>224</sup> decay.

## III. COMPARISON WITH THEORY

The first accurate internal-conversion coefficient calculations were performed by Rose'4 and by Sliv and Band<sup>25</sup>; however, as experimental techniques were improved, the need for more extensive calculations became evident. At least four such calculations are in progress evident. At least four such calculations are in progress<br>or have been completed.<sup>14,26–28</sup> The results of the calculations by Hager and Seltzer using Hartree-Fock-Slater screening procedures to calculate the electron<br>wave functions have recently been published.<sup>28</sup> The wave functions have recently been published.<sup>28</sup> The primary advantages in using the tables of Hager and

Seltzer are that internal-conversion coefficients were calculated for every atomic number and for energies as low as 1 keV above threshold, eliminating the need for extensive interpolation procedures.

The L-subshell internal-conversion coefficients  $(\alpha_{L_{I}}, \alpha_{L_{II}}, \alpha_{L_{III}})$  were obtained from the tables of Hager and Seltzer by a logE-versus-log $\alpha$  computer interpolation procedure using Newton's method of divided differences. <sup>29</sup> In general, the resulting subshell internalconversion coefficients were independent of the number of points used in the interpolation procedure provided that at least three points bridging the transition energy were used. Internal-conversion coefficients from the calculations of Sliv and Band were not obtained directly from the tables,<sup>25</sup> but were obtained by Dingus rectly from the tables,<sup>25</sup> but were obtained by Dingu using a computer interpolation procedure<sup>30</sup> which utilized only the L-subshell internal-conversion coefficients directly calculated by Sliv and Band for selected atomic number and energy values. The results of similar interpolations<sup>30</sup> applied to the conversion coefficients calculated by Rose<sup>24</sup> showed a difference of less than  $1\%$  in the total L-shell conversion coefficient compared to the values obtained from the calculations of Sliv and Band. The theoretical total internal-conversion coefficients calculated by Rose are not compared with the experimental values because the calculations by Sliv and Band<sup>25</sup> and by Hager and Seltzer<sup>28</sup> were made with smaller energy spacing between calculated points and were therefore considered to be more suitable for interpolation purposes.

The theoretical total internal-conversion coefficient for a given transition was obtained from the  $L$ -shell internal-conversion coefficient  $\alpha_L = \alpha_{L1} + \alpha_{L11} + \alpha_{L11}$  for that transition and from the ratio  $\alpha_L/(1+\alpha)$ , available from  $\beta$ -ray spectrometer measurements.<sup>31,32</sup> For the from  $\beta$ -ray spectrometer measurements.<sup>31,32</sup> For the transition in Ra<sup>224</sup>, for which  $\alpha_L/(1+\alpha)$  has not been reported,  $0.73 \pm 0.03$ , the average value of the results of Ref. 31, was used.

A comparison of the experimental total internalconversion coefficients presently available for  $E2$  transitions in heavy deformed nuclei with the theoretical predictions is given in Table III. In the table,  $\alpha_{\rm expt}$ <sup>S</sup> are taken from Ref. 17, while  $\alpha_{\rm expt}$  are the results of the present investigation. The results of Hager and Seltzer<sup>28</sup> and of Sliv and Band<sup>25</sup> are given by  $\alpha_{\rm th}$ <sup>HS</sup> and  $\alpha_{\rm th}$ <sup>SB</sup>, respectively. The uncertainties associated with the theoretical values result only from the reported experimental uncertainties in  $\alpha_L/(1+\alpha)$ . Uncertainties resulting from the calculations of the theoretical values (which are of the order of  $2\%$ ) are not included in order to emphasize the effects of the uncertainties in the reported values of  $\alpha_L/(1+\alpha)$ .

<sup>&</sup>lt;sup>24</sup> M. E. Rose, *Internal Conversion Coefficients* (North-Holland Publishing Co., Amsterdam, 1958).

<sup>~~</sup> L. A. Sliv and I. M. Sand, Coefficients of Internal Conversion of Gamma-Radiation (USSR Academy of Sciences, Moscow-Leningrad, 1956), Parts I and II; also see L. A. Sliv and I. M. Band, in Alpha-, Beta-, and Gamma-Ray Spectroscopy, edited by K. Siegbahn (North-Holland Publishing Co. , Amsterdam, 1965),

Vol. 2.<br><sup>26</sup> H. C. Pauli, Purdue University Report No. COO-1420-137,<br>1967 (unpublished).

<sup>&</sup>lt;sup>22</sup><sup>T</sup>R. F. O'Connell and C. O. Carroll (private communication).<br><sup>28</sup> R. S. Hager and E. C. Seltzer, U. S. Atomic Energy Com-<br>mission Report No. CALT-63-60, 1967 (unpublished); also, Nucl. Data 4A, Nos. 1 and 2 (1968).

<sup>29</sup> F. S. Hildebrand, Introduction to Numerical Analysis (Mc-Graw-Hill Book Co., New York, 1956). '0 R. S. Dingus and Niels Rud, University of Colorado Report

No. COO-535-577, 1967 (unpublished).<br><sup>31</sup> D. Engelkemeir and J. Halley, Phys. Rev. 134, A24 (1964).<br><sup>32</sup> D. H. Rester, M. S. Moore, F. E. Durham, and C. M. Class<br>Nucl. Phys. **22**, 104 (1961).

**Nucleus** 

 $Ra^{224}$ 

 $Th<sup>228</sup>$ 

 $Th<sup>232</sup>$ 

T<sub>1234</sub>

T<sub>1236</sub>

 $\dot{\bf U}^{238}$ 

 $Pu<sup>240</sup>$ 

Transition energy<br>(keV)

84.4

57.6

49.8

43.50

45.28

44.70

42.88

332  $±13$ 

664

985  $± 9$ 

 $160 \pm 2$ 

 $730 \pm 9$ 

641  $\pm 19$ 

 $\pm 27$ 



692  $\pm$  8

612  $\pm 18$ 

634  $±26$ 

939  $±9$ 

<sup>a</sup> When both  $\alpha$ <sub>expt</sub> and  $\alpha$ <sub>expt</sub><sup>*s*</sup> are given for a particular transition,  $\alpha$ <sub>expt</sub> is used in the comparison.

 $158 \pm 7$ 

780 ±55

845  $\pm$ 93

607  $±29$ 

For the transitions in Ra<sup>224</sup>, Th<sup>228</sup>, and U<sup>236</sup>, the agreement with the calculations of Hager and Seltzer and of Sliv and Band is quite good in that there is no deviation outside of the experimental uncertainties for these E2 transitions. The internal-conversion coefficient for the transition in  $U^{234}$  is  $12\%$  higher than the Hager-Seltzer value and appears to be larger than the Sliv-Band value. For the transition in Pu<sup>240</sup>, the measured total internalconversion coefficient is about  $14\%$  lower than the Sliv-Band value and appears to be smaller than the value from the calculations of Hager and Seltzer.

 $260 + 30$ 

 $702 \pm 80$ 

 $575 + 120$ 

 $558 + 60$ 

 $868 + 78$ 

#### IV. CONCLUSIONS

The results of this investigation are in agreement, as shown in Table III, with the internal-conversion coefficient measurements from the Coulomb-excitation and lifetime results. For the 45-keV transition in U<sup>236</sup>, the uncertainty in the present result is substantially lower, enabling a more accurate comparison with the theoretical values.

Stelson<sup>17</sup> has suggested that the internal-conversion coefficients for these transitions tend to be less than the theoretical values. As can be seen, the internal-conversion coefficients for the transitions in Th<sup>232</sup>, U<sup>238</sup>, and Pu<sup>240</sup> are definitely lower than theory; the remaining internal-conversion coefficients are in close agreement with the theoretical values with the exception of the transition in U<sup>234</sup>, which has an internal-conversion coefficient higher than the theoretical value. The observed differences with theory may not be serious for the following reasons. First, the Coulomb-excitation measurements used in determining the internal-conversion coefficients reported in Table III were performed

by two groups, 33,34 one of which<sup>33</sup> measured the  $B(E2)$ values for the determination of the Th<sup>232</sup> and U<sup>238</sup> internal-conversion coefficients. Thus a systematic error present in the measurements by this group would affect only the Th<sup>232</sup> and U<sup>238</sup> internal-conversion coefficients. Second, if the theoretical internal-conversion coefficients are assigned uncertainties of 2 or  $3\%$ , the disagreements are not so large, particularly for the Pu<sup>240</sup> and U<sup>234</sup> internal-conversion coefficients which have relatively small experimental uncertainties in  $\alpha_L/(1+\alpha)$ .

 $1.01 + 0.05$ 

 $0.81 + 0.10$ 

 $1.13 + 0.08$ 

 $0.99 + 0.06$ 

 $0.88 + 0.10$ 

 $0.90 + 0.10$ 

In conclusion, for the transitions in Ra<sup>224</sup>, Th<sup>228</sup>, and U<sup>236</sup>, the present picture of the total internal-conversion coefficients of these  $2^+ \rightarrow 0^+$  transitions in the very heavy nuclei shows reasonably good agreement between experimental and theoretical values. The deviations from theory, if real, cannot be understood in terms of any known phenomena. Hopefully, as experimental techniques improve, those internal-conversion coefficients which appear somewhat different from theory can be remeasured to a greater accuracy, so that a better comparison can be made.

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 $0.99 + 0.05$ 

 $0.78 + 0.10$ 

 $1.07 + 0.08$ 

 $0.95 + 0.05$ 

 $0.84 + 0.11$ 

 $0.86 + 0.09$ 

<sup>&</sup>lt;sup>33</sup> B. Elbek, Determination of Nuclear Transition Probabilities by

Coulomb Excitation (Munksgaards, Copenhagen, 1963).<br><sup>34</sup> A. M. Friedman, J. R. Erskine, and T. H. Braid. Bull. Am. Phys. Soc. 10, 540 (1965).