

FIG. 4. Comparison of experimental and theoretical values of the circular polarization of the resonant scattered gamma rays. The ordinate represents the averaged angular distribution,  $\bar{W}$ , according to Eq. (5) and the abscissa is the relative contribution of Gamow-Teller interaction. The curve gives the theoretical values assuming a positive Gamow-Teller to Fermi matrix-element ratio (upper branch) and a negative ratio (lower branch).

Equation (5) gives the following explicit answer:

$$\bar{W} = 1 - 0.536[0.0873(x^2 + 5.91x)/(1+x^2)].$$

If we plot  $\bar{W}$  as a function of  $x^2/(1+x^2)$ , the fractional

contribution of Gamow-Teller interaction, we obtain the curve reproduced in Fig. 4. The upper branch corresponds to positive values of  $x$  and the lower branch to negative values of  $x$ . The experimental point is also shown in this figure. We conclude from the experiment that  $x$  is negative and, therefore,  $M_{GT}/M_F$  is negative, although we are aware of the relatively large experimental uncertainty.

Metzger<sup>13</sup> puts an upper limit of 4% to the fluorescent scattering contribution of the 280-kev line (see Fig. 1) with respect to the 265-kev line. For the 121 kev-280 kev cascade one finds  $\bar{W} = 1 - 0.554A'$ .  $A'$  differs from  $A$  only by the  $F_1$ -coefficient of the second  $\gamma$  ray the ratio  $F_1(1,1,\frac{3}{2},\frac{5}{2})/F_1(1,1,\frac{3}{2},\frac{3}{2})$  being 2.3. A 4% contribution of the 280-kev  $\gamma$  line would therefore result in an increase of the absolute value of the anisotropy curve of Fig. 4 of 10%, not affecting our conclusion.

#### ACKNOWLEDGMENTS

We wish to thank Dr. H. Weidenmüller and Dr. L. Krüger for many helpful discussions.

### Radioactive Decay of $\text{Tm}^{166}$

R. G. WILSON AND M. L. POOL

*Department of Physics and Astronomy, The Ohio State University, Columbus, Ohio*

(Received January 25, 1960)

Erbium oxide enriched to 72.9% in the 166 mass number was irradiated with 6-Mev protons. An activity decaying by electron capture and positron emission with a half-life of  $7.69 \pm 0.05$  hours was produced by a  $(p,n)$  reaction and its assignment to  $\text{Tm}^{166}$  confirmed. The observed activity consists of the  $K$  x ray of erbium, gamma rays with energies of 81, 184, 289, 405, 460, 598, 674, 694, 707, 759, 782, 788, 878, 1052, 1179, 1276, 1351, 1589, 1874, and 2058 kev, annihilation radiation, and particle radiation with an end-point energy of  $2090 \pm 40$  kev. Gamma-gamma coincidence measurements and consideration of the energies and relative numbers of the observed radiations have led to the assignment or confirmation of energy levels at 81 (2+), 265 (4+), 554 (6+), 788 (2+), 863 (3+), 959 (4+), 1248 (2), 1317 (5), 1462 (0+), 1547 (3+), 1701 (4+), 1894 (5+), 2139 (3), and 2168 (0) kev in  $\text{Er}^{166}$ . The 2139-kev level is highly populated by electron capture and the positron transitions occur to the 265 (4+)-kev level. The positions of the observed radiations and the branching ratios of electron capture are shown in a proposed energy level scheme.

#### INTRODUCTION

AN activity decaying 99+% by electron capture and <1% by positron emission with a half-life of 7.7 hours has been assigned to  $\text{Tm}^{166}$ .<sup>1</sup> Gamma radiation of approximately 1.7 Mev was detected in this activity and the positron end-point energy was found to be 2.1 Mev. Conversion electron measurements following the proton irradiation of natural ytterbium oxide have led to the assignment of transitions with energies of 80.7, 154.6, 184.7, 194.8, and 215.4-kev and to the postulation of energy levels of 81 (2+) and 265 (4+)

kev in  $\text{Er}^{166}$ .<sup>2</sup> One group of workers has reported gamma rays in this activity with energies of 80, 180, 670, 800, and possibly 1320 kev.<sup>3</sup> Another group has reported gamma rays with energies of 80, 180, 690, and 780 kev in this activity and have postulated an energy level of 780 kev (2+) in addition to the 265 (4+)- and 81 (2+)-kev levels.<sup>4</sup> The assignment of an 80.6-kev level resulted

<sup>2</sup> J. W. Mihelich, B. Harmatz, and T. H. Handley, *Phys. Rev.* **108**, 989 (1957).

<sup>3</sup> W. E. Nervi and G. T. Seaborg, *Phys. Rev.* **97**, 1092 (1955).

<sup>4</sup> G. M. Gorodinskii, A. N. Murin, V. N. Pokrovskii, B. K. Preobrazhenskii, and N. E. Titov, *Doklady Akad. Nauk (S.S.S.R.)* **112**, 405 (1957) [translation: *Soviet Phys.-Doklady* **2**, 39 (1957)].

<sup>1</sup> G. Wilkinson and H. G. Hicks, *Phys. Rev.* **75**, 1370 (1949).

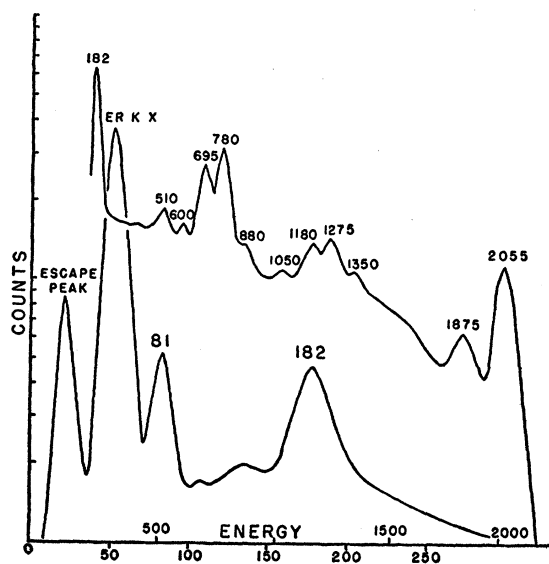


FIG. 1. Gamma-ray spectrum of 7.7-hour  $\text{Tm}^{166}$  measured with a  $3 \times 3$  inch crystal on a 100-channel scintillation spectrometer.

from Coulomb excitation experiments.<sup>5</sup> The energies and relative numbers of conversion electrons from 22 transitions in the activity of  $\text{Tm}^{166}$  have been reported.<sup>6</sup>

#### EXPERIMENTAL RESULTS

Erbium oxide enriched to 72.9% in the 166 mass number was irradiated with 6-Mev protons. The composition of the remaining portion is as follows in percent:  $<0.1 \text{ Er}^{162}$ ,  $0.1 \text{ Er}^{164}$ ,  $17.7 \text{ Er}^{167}$ ,  $8.5 \text{ Er}^{168}$ , and  $1.5 \text{ Er}^{170}$ . The atomic number of the resulting activity was determined by the identification of the erbium  $K$  x ray, which was compared with the known  $K$  x rays of terbium, dysprosium, thulium, ytterbium, lutetium, and tantalum emitted from radioactive  $\text{Dy}^{159}$ ,  $\text{Tb}^{160}$ ,  $\text{Yb}^{169}$ ,  $\text{Tm}^{170}$ ,  $\text{Hf}^{175}$ , and  $\text{W}^{181}$ , respectively. Ion-exchange separation was deemed unnecessary.

The mass number of the activity was determined by identifying and comparing the relative amounts of this activity and the well-known activities of  $\text{Tm}^{170}$ ,  $\text{Tm}^{168}$ , and  $\text{Tm}^{167}$  in similar proton irradiations of enriched  $\text{Er}^{166}$  and  $\text{Er}^{168}$ .

The half-life of the activity produced by the proton irradiation of enriched  $\text{Er}^{166}$  is  $7.69 \pm 0.05$  hours as measured by following the decay of the gamma-ray spectrum for over five half-lives with a 100-channel scintillation spectrometer. The original assignment of the 7.7-hour activity of  $\text{Tm}^{166}$  is therefore confirmed.

Figure 1 shows the observed gamma-ray spectrum of  $\text{Tm}^{166}$  which includes gamma rays with energies of  $81 \pm 1$ ,

TABLE I. Relative numbers of gamma rays,  $N_\gamma$ , conversion electrons,  $N_{ce}$ , and corresponding transitions,  $N_t$ , in the activity of  $\text{Tm}^{166}$  for gamma-ray energies,  $E_\gamma$ , expressed in kev.

$E_\gamma$	Ref.	$N_\gamma$	$N_{ce}$	$N_t$
K x ray		100		
80.7	2, 6	9	16000	86
154.1	2, 6		50	
184.4	2, 6	20	2200	31
193.2	2, 6		240	
214	2, 6		140	
289		a		$<1$
299	6		17	$<1$
347	6		22	$<1$
405	6	a	77	$<1$
460	6	a	89	$\sim 1$
595	6			
598	6	1.5	33	1.5
674	6		26	1.6
694	6	6	45	2.7
707	6		27	1.7
759	6		27	2.2
763	6	9	46	3.8
782	6		37	3.0
788	6			1.0
878		1.0		
1052		$\sim 1$		$\sim 1$
1179	6	4.0	27	4.0
1276	6	5.7	32	5.7
1351		1.1		1.1
1589		a		$<1$
1874		17		17
2058	6	39	15	30
2087	6		6	9

a Observed in coincidence spectra only.

$182 \pm 2$ ,  $600 \pm 5$ ,  $695 \pm 5$ ,  $780 \pm 5$ ,  $880 \pm 8$ ,  $1050 \pm 10$ ,  $1180 \pm 8$ ,  $1275 \pm 8$ ,  $1350 \pm 12$ ,  $1875 \pm 15$ , and  $2055 \pm 12$  kev in addition to the erbium  $K$  x ray. Gamma-gamma coincidence measurements have shown that in addition to the gamma rays mentioned above, there exist weaker gamma rays with energies of 285, 405, 460, and 1590 kev, and that the 695- and 780-kev peaks are each composed of more than one gamma ray. No gamma ray with an energy greater than 2200 kev and an intensity greater than 1% of that of the 2055-kev gamma ray exists. A peak in the gamma-ray spectrum at 510 kev was shown to be annihilation radiation by its strong 180 deg self-coincidence. Weak particle radiation with an end-point energy of  $2090 \pm 40$  kev was observed by the use of a Geiger tube with aluminum absorbers.

All of the reported radiations emitted from  $\text{Tm}^{166}$  are listed in Table I with the conversion electron references. The figures in the third column of Table I are the relative numbers of counts under the spectral distributions of the observed gamma rays after correction for crystal efficiency. The figures in the fourth column are the relative numbers of conversion electrons as reported in reference 6. The relative numbers of the 81- and 182-kev gamma rays were corrected for internal conversion<sup>7</sup> using the data in Table II. The numbers in the last column of Table I are the relative numbers of the transitions. In cases where several gamma rays were observed

<sup>5</sup> E. L. Chupp, J. W. M. DuMond, F. J. Gordon, R. C. Jopson, and H. Mark, *Phys. Rev.* **112**, 518 (1958).

<sup>6</sup> A. A. Bashilov, Ia. Gromov, G. M. Gorodinskii, B. G. Dzhelepov, et al., *Proceedings of the Second United Nations Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1958), U. S. Atomic Energy Commission micro-card A/CONF/15 P2477.

<sup>7</sup> M. E. Rose, *Internal Conversion Coefficients* (North-Holland Publishing Company, Amsterdam, 1958).

TABLE II. Internal conversion data for erbium. The  $\alpha$ 's are from reference 7.  $N_T/N_\gamma$  and  $N_T/N_K$  are the ratios of the total number of transitions to the number of gamma rays and  $K$ -converted transitions, respectively, for gamma-ray energies,  $E_\gamma$ , expressed in kev.

$E_\gamma$	$\alpha_K$	$\alpha_{L1}$	$\alpha_{L2}$	$\alpha_{L3}$	$\alpha_M$	$N_T/N_\gamma$	$N_T/N_K$
81	1.60	0.61	1.90	1.90	1.84	8.4	5.2
182	0.21	0.021	0.042	0.035	0.042	1.35	6.4

in one peak, the numbers in the third column were divided using the data in the fourth column. All of the numbers in the last column have been adjusted to read in percentages of disintegrations. The first number in

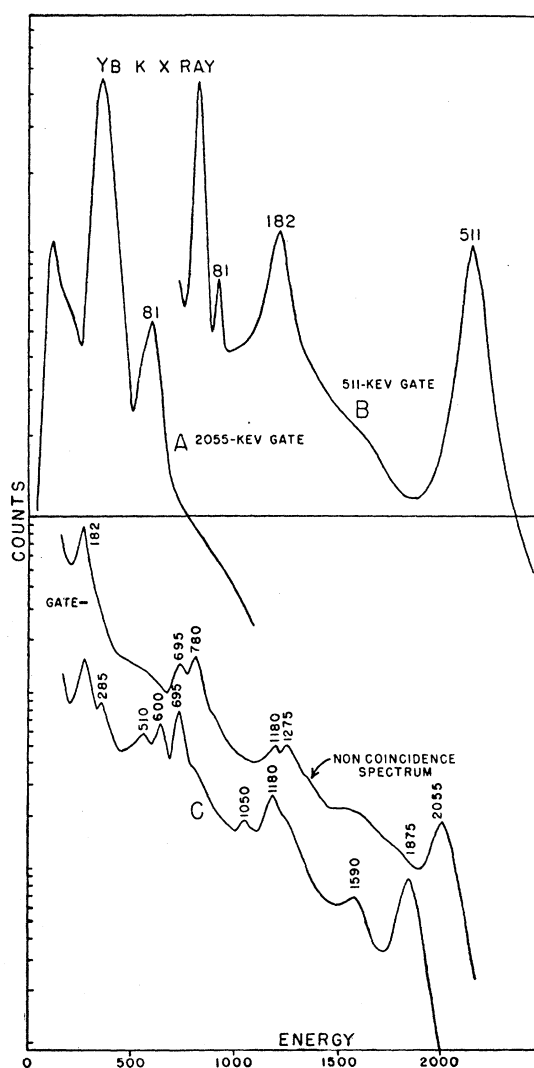


FIG. 2. Typical gamma-gamma coincidence spectra obtained for the activity of  $\text{Tm}^{166}$  with a coincidence circuit of resolving time  $2\tau = 1.5 \times 10^{-6}$  sec. Gamma energies are in kev. Individual spectra are coincidences gated by 2055-kev events for curve A, by 511-kev events for curve B, and by 182-kev events for curve C. Curve C shows the 1590- and 285-kev gamma rays which can be seen only in the coincidence spectrum.

the fifth column of Table I, 107, is the total number of  $K$  x ray producing events of which only 100 are observed because the  $K$ -fluorescence yield in erbium is 0.932.<sup>8</sup>

Table III is a tabulation of the gamma-gamma coincidence information obtained for the activity of  $\text{Tm}^{166}$  with a coincidence circuit of resolving time  $2\tau = 1.5 \times 10^{-6}$  sec and two  $1\frac{3}{4} \times 2$  inch crystals used at either 90 or 180 deg. Three typical coincidence spectra are shown in Fig. 2. Curve A shows that only the  $K$  x ray and the 81-kev gamma ray are in coincidence with the 2055-kev gamma ray and that there are a sufficient number of  $K$  x rays compared to the number of 81-kev gamma rays to allow a very high percentage of  $K$  x rays to precede the 2055-81-kev cascade. Curve B shows that the  $K$  x ray and the 81- and 182-kev gamma rays are in coincidence with the 180 deg self-coincident annihilation radiation. This curve shows that there is approximately a one to one relationship between the number of 81- and 182-kev transitions in coincidence with the annihilation radiation after correction for internal conversion. Curve C shows that the  $K$  x ray and gamma rays with energies of 81, 285, (510), 600, 695, 1050, 1180, and 1875 kev are in coincidence with the 182-kev gamma ray.

#### DISCUSSION

Figure 3 shows an energy level scheme for the decay of  $\text{Tm}^{166}$  which is consistent with the experimental evidence of this investigation. It accounts for 27 out of the 28 observed radiations listed in Table I.

The assignment of a 265-kev (4+) level is now confirmed because (1) strong coincidences are observed between the 81- and 182-kev gamma rays, (2) there exists no 265-kev crossover transition, (3) strong transitions occur from higher levels to this and the 81-kev (2+) levels as shown by the gamma-gamma coincidence measurements, and (4) as noted by others, this level is predicted for the second ground-state rotational level. The possibility of a 550-kev (6+) rotational level is suggested by the existence of a weak 285-kev gamma ray seen only in the coincidence spectrum gated by 182-kev events.

The existence of a 2139-kev level in  $\text{Er}^{166}$  which is highly populated by electron capture transitions from  $\text{Tm}^{166}$  is clearly shown by the strong coincidences observed between the 2055-kev gamma ray and only the 81-kev gamma ray and between the 182- and 1875-kev gamma rays. Further evidence of the 2139-kev level is seen by the coincidences observed between transitions from this level to lower excited states and transitions from these states to levels in the ground-state band.

Because a 2087-kev transition is reported in reference 6 in addition to a 2058-kev transition, it is assumed that the 2055-kev peak in the gamma-ray spectrum seen in Fig. 2 is composed of both of these transitions. The

<sup>8</sup> A. H. Wapstra, J. G. Hijgh, and R. Van Lieshout, *Nuclear Spectroscopy Tables* (North-Holland Publishing Company, Amsterdam, 1959).

TABLE III. Gamma-gamma coincidence information for the activity of  $\text{Tm}^{166}$ . Designations are gamma-ray energies in kev.

	K x ray	81	184	215	285	405	460	511	600	675 695 705	765 780 785	880	1050	1175	1275	1350	1590	1875	2055
K x ray	yes	yes	yes					yes	yes	yes	yes	yes	yes	yes	yes	yes		yes	yes
81	yes	no	yes					yes	yes	yes	w	yes	yes	yes	yes		yes	yes	yes
182	yes	yes		yes	yes			yes	yes	yes	no	no	yes	yes	no	no	yes	yes	no
511	yes	yes	yes		no			yes	no	no	no	no	no	no	no	no	no	no	no
695	yes	yes	yes	yes	no		w	no		yes	w	no	yes	no	yes	no	no	no	no
780	yes	yes	no		no	yes	yes	no		yes	no	no	no	no	yes	yes	no	no	no
1175	yes	yes	yes					no		yes	no	yes	no	no	no	no	no	no	no
1275	yes	yes	yes					no		no	yes	no	no	no	no	no	no	no	no
1350	yes	yes	yes					no		yes	yes	no	no	no	no	no	no	no	no
2055	yes	yes	no	no	no	no	no	no	no	no	no	no	no	no	no	no	no	no	no

coincidence information then shows that both of these transitions occur to the 81-kev level. Therefore a level at 2168 kev is tentatively assigned with a possible spin of 0 because no other transitions from this level are observed.

A 788-kev level with a spin of 1 or 2 is implied because (1) strong 707- and 788-kev transitions exist in the activity of  $\text{Tm}^{166}$ , (2) the 695-kev peak is strongly in coincidence with the 81-kev gamma ray, while the 780-kev peak is only weakly in coincidence with the 81-kev gamma ray, and (3) a 1350-kev gamma ray is in coincidence with both the 695- and 780-kev peaks. Reason (2) implies that the 780-kev peak is composed of at least two gamma rays, one of which is stronger and not in coincidence with the 81-kev gamma ray and one which is weaker and in coincidence with the 81-kev transition.

A level with a spin of 3 or 4 is implied at 863 kev because (1) a 782-kev gamma ray is in coincidence with the 81-kev but not with the 182-kev, (2) the 598-kev gamma ray is in coincidence with the 182-kev, and (3) the 1276-kev gamma ray is in coincidence with the 782-kev.

A 959-kev level of spin 3 or 4 is implied because the 878-kev gamma ray is in coincidence with only the 81- and 1179-kev gamma rays and the 695-kev peak is in coincidence with the 182- and 1179-kev gamma rays. This situation also requires that the 695-kev peak be composed of more than one gamma ray. The weak 405-kev gamma ray observed in coincidence with the 182-kev gamma ray may then occur between this level and the 6+ level of the ground-state band. The 788-, 863-, and 959-kev levels seem to comprise a rotational band with  $K=2$  and hence have spins of 2+, 3+, and 4+, respectively. If  $K$  were greater than 2, transitions within the rotational band ( $\Delta K=0$ ) would be more highly favored than transitions to the ground-state band ( $\Delta K \geq 3$ ). If  $K$  were 1, the 598-kev transitions would be replaced by an 863-kev transition and the observed strong coincidences between the 182- and 598-kev gamma rays would not occur. That these three levels do comprise a rotational band is favored by the ratio of the energy differences between them. This ratio is  $171/75 = 2.3$  which is in the range of values predicted by the formula  $E \propto [I(I+1) - I_0(I_0+1)]$  where  $I_0$  is the spin

of the ground state of the band and  $I$  is the spin of excited levels.

The strong transitions from the 2139-kev level to the 2+ and 4+ levels of the ground-state rotational band and the weaker transitions to the three levels of the  $K=2$  rotational band favor the assignment of spin 3 for the 2139-kev level. A spin of 4 is made less favorable by the spin assignment made for the ground state of  $\text{Tm}^{166}$ .

$^{69}\text{Tm}^{166}$  is an odd-odd nucleus in the region of elliptically deformed odd-odd nuclei. The collective nuclear model predicts a doublet of states for such a nucleus, one of which is the ground state. The measured ground-state spins of  $^{69}\text{Tm}^{169}$  and  $^{66}\text{Dy}^{163}$  are  $\frac{1}{2}+$  and  $\frac{5}{2}-$ ,

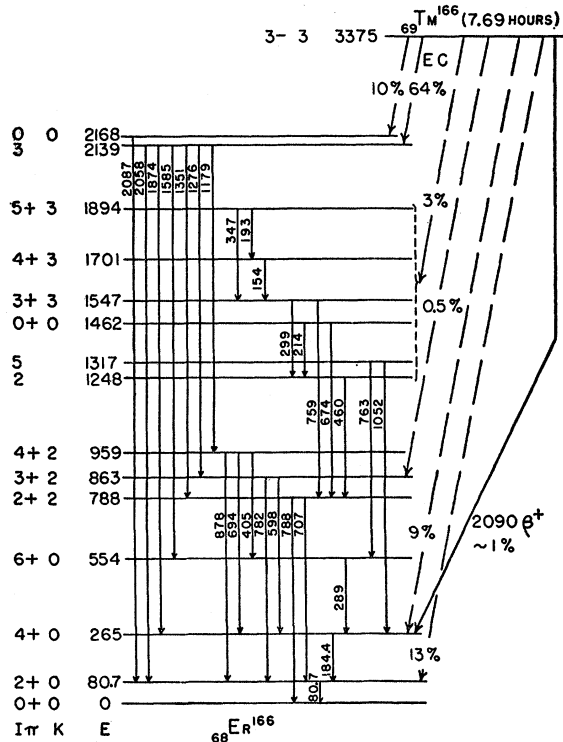


FIG. 3. Proposed energy level scheme for the decay of  $\text{Tm}^{166}$ . Energy level designations are in kev.

respectively. Spins of 2- and 3- are therefore predicted for  ${}_{69}\text{Tm}_{97}^{166}$ . The choice of 3- for the ground state of  $\text{Tm}^{166}$  is consistent with the population of the two levels with spin 4+, one of which is populated directly by electron capture decay, and of the 6+ (0+) ground-state rotational level.

The value of  $K$  for the 2139-keV level is not easily chosen. The high population of this level by electron capture from the now assigned 3- ground state of  $\text{Tm}^{166}$  implies the possibility of a single-particle state with  $K=3$ . However, then the strong transitions to the members of the ground-state band for which the change in  $K$  is 3 seem inconsistent. The possibility of  $K=0$  is consistent with transitions to the ground-state band but seems to be inconsistent with the high population of the 2139-keV level by electron capture.

A level at 1317 keV of possible spin 5 is suggested by the 1050-keV gamma ray, which is in coincidence with the 182-keV gamma ray and by the existence of a 763-keV transitions reported in reference 6. Levels at 1248 (2), 1462 (0), and 1547 (3) keV are suggested by the gamma-gamma coincidence information and the existence of transitions with energies of 214, 299, 347, 460, 674, and 759 keV reported in reference 6. The 460-, 674-, and 759-keV gamma rays are observed in the coincidence spectra gated by the gamma peaks which include the 707- and 788-keV gamma rays.

The transitions with energies of 154, 193, and 347 are not observed in the gamma-ray spectrum but are shown in Fig. 3 as comprising a rotational band with  $K=3$ . A rotational band is chosen because the 347-keV line can serve as a crossover transition and the energy ratio is then just that predicted and observed for rotational bands.  $K=3$  is chosen because transitions from the excited levels of the band occur to the lower members of the band rather than to other states.

The strong coincidences observed between the annihilation radiation and only the 81- and 182-keV gamma rays led to the conclusion that the positron decay of  $\text{Tm}^{166}$  populates the 265-keV (4+) level of  $\text{Er}^{166}$ . The relative numbers of the 81- and 182-keV gamma rays in the coincidence spectrum gated by 511-keV events

imply that there is approximately a one to one relationship between the 81- and 182-keV transitions following the positron decay. Positron decay transitions to the ground state of  $\text{Er}^{166}$  are precluded by the large  $\Delta I$  and  $\Delta K$ . The population of the  $265 \pm 3$ -keV level by the  $2090 \pm 40$ -keV positron places the ground state of  $\text{Tm}^{166}$   $3375 \pm 45$  keV above the ground state of  $\text{Er}^{166}$ .

The relative amounts of electron capture decay shown in Fig. 3 were obtained by subtracting from the 107  $K$  x rays in the fifth column of Table I,  $K$  x rays resulting from  $K$  conversion and one  $K$  x ray for every electron capture transition required to balance the difference between the number of transitions from a given level and transition into the same level. As seen in curve *A* of Fig. 2, a large number of  $K$  x rays exists in the coincidence spectrum gated by 2055-keV events. Enough  $K$  x rays are present to account for  $K$  conversion of the 81-keV transitions required to produce the number of 81-keV gamma rays observed and to allow one  $K$  x ray to precede every 2055-keV gamma ray which gated the observed 81-keV gamma rays. Therefore the 2139-keV level of  $\text{Er}^{166}$  must be populated largely by  $K$  capture.  $L$  capture to all of the levels of  $\text{Er}^{166}$  is assumed negligible with respect to  $K$  capture.

At the end of the section on  $\text{Tm}^{166}$  in reference 4, a rough calculation of the branching ratios of electron capture to the 80- and 264-keV levels of  $\text{Er}^{166}$  was made. The inconsistency mentioned there of these ratios with the predicted spin for  $\text{Tm}^{166}$  is now removed because this investigation shows that the 80- and 265-keV levels are largely populated from higher levels in  $\text{Er}^{166}$ .

#### ACKNOWLEDGMENTS

One of us (R. G. W.) is grateful to the National Science Foundation for the grant of a fellowship which enabled the completion of this research. Appreciation is expressed to R. P. Sullivan of the Department of Physics and Astronomy for assistance in the electronic phases of this research and to the Office of Naval Research for support in obtaining the enriched isotopes.