

It can also be shown that the median of  $p(\tau)$ ,  $\tau_m$ , is approximately at the first minimum of the correlation function,  $\psi(\tau)$ ,<sup>9</sup> so that Eq. (7) determines the correlation function at best up to the neighborhood of its first minimum. Finally it is of interest to note that  $p(\tau)$  is independent of the variance of  $f(X)$ , which is entirely determined by its normalization. This point has been remarked on by Rosenzweig.<sup>7</sup>

### CONCLUSIONS

Besides the level repulsion effect, which is observed, the only other prediction of this work which is immediately amenable to test is that the distribution of level spacings is asymptotically a negative exponential. Examination of Fig. 7 of reference 1 shows that for  $\tau/\langle\tau\rangle$  exceeding about  $\frac{3}{2}$  the experimental data are fairly well fitted by an exponential curve. On the other hand, reference to Fig. 9 of the same article shows that

Wigner's distribution,

$$p(\tau) = \frac{\pi}{2} \frac{\tau}{\langle\tau\rangle^2} \exp\left[-\frac{\pi}{4} \left(\frac{\tau}{\langle\tau\rangle}\right)^2\right], \quad (8)$$

predicts too few large spacings, a fact also noticed by Rosenzweig.<sup>†</sup> However, Wigner's distribution is a good fit for  $\tau \lesssim \langle\tau\rangle \sim \tau_m$ , and in this range predicts a correlation function of the form

$$\frac{\psi(\tau)}{\psi(0)} = \cos\left[2\pi \operatorname{erf}\left(\frac{\sqrt{\pi}\tau}{2\langle\tau\rangle}\right) - \pi \frac{\tau}{\langle\tau\rangle}\right] \quad \tau \lesssim \langle\tau\rangle. \quad (9)$$

Plotted in Fig. 1 is  $\psi(\tau)/\psi(0)$  vs  $\tau/\langle\tau\rangle$ . The solid portion of the curve is derived from Eq. (9), the dotted portions of the curve are merely illustrations of possible behaviors which, however, become vanishingly small for  $\tau/\langle\tau\rangle \gtrsim \frac{3}{2}$ .

## Scintillation Studies of Some Neutron Deficient Isotopes of Lutecium\*

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Lu<sup>170</sup>, Lu<sup>172</sup>, Lu<sup>173</sup>, and Lu<sup>174</sup> activities were produced by bombarding Lu<sub>2</sub>O<sub>3</sub> with bremsstrahlung from the University of Illinois betatrons. Gamma rays of energy 0.083, 0.190, 0.245, and 2.04 Mev were associated with the decay of Lu<sup>170</sup>. A gamma-gamma coincidence experiment showed that the 2.04-Mev gamma ray was coincident with the three low-energy transitions. Gamma rays of energy 0.079, 0.113, 0.181, 0.203, 0.325, 0.370, 0.525, 0.820, 0.900, and 1.09 Mev were associated with the electron capture decay of 6.7-day Lu<sup>172</sup>, the isotope studied in the most detail. Levels of energy 0.0787, 0.2602, 0.3731, 0.5769, 0.9015, 1.082, and 1.99 Mev above the ground state have been assigned to Yb<sup>172</sup> by gamma-gamma coincidence measurements and energy considerations. Gamma rays of energy 0.022, 0.079, 0.113, 0.145, 0.176, 0.274, 0.335, 0.440, 0.550, and 0.640 Mev were assigned to transitions between levels of Yb<sup>173</sup> while gamma rays of energy 0.077, 0.084, 0.113, 0.176, 0.230, 0.275, 0.990, and 1.245 Mev were associated with the decay of Lu<sup>174</sup>. A summary of all gamma-gamma coincidence experiments involving Lu<sup>173</sup> and Lu<sup>174</sup> is included in this paper. The 0.084-Mev transition associated with the decay of Lu<sup>174</sup> was interpreted to be the first excited level of Hf<sup>174</sup>. A rough calculation of the *K*-conversion coefficient of this transition yielded  $\alpha_K \lesssim 2.5$ .

### I. INTRODUCTION

WILKINSON and Hicks<sup>1,2</sup> were the first experimenters to make a survey of neutron-deficient lutecium isotopes. In that work the nuclides were produced by bombarding thulium with alpha particles of various energies, and by bombarding ytterbium with 10-Mev protons. After chemical separation Wilkinson and Hicks identified half-lives of  $1.7 \pm 0.1$  days with Lu<sup>170</sup>,  $8.5 \pm 0.2$  days with Lu<sup>171</sup>,  $6.7 \pm 0.05$  days with Lu<sup>172</sup>,  $4.0 \pm 0.1$  hours with Lu<sup>172m</sup>,  $\sim 1.4$  years with Lu<sup>173</sup>, and  $165 \pm 5$  days with Lu<sup>174</sup>. On the

basis of absorption techniques these workers listed gamma rays at  $\sim 2.5$  Mev belonging to Lu<sup>170</sup>,  $\sim 1.2$  Mev belonging to Lu<sup>171</sup>,  $\sim 1.2$  Mev belonging to Lu<sup>172</sup>,  $\sim 0.22$  and  $\sim 0.8$  Mev belonging to Lu<sup>173</sup>, and  $\sim 1$  Mev belonging to Lu<sup>174</sup>. In addition, they reported a  $\beta^-$  group of end point 0.6 Mev in Lu<sup>174</sup>.

More recently Mihelich, Harmatz, and Handley<sup>3</sup> have made a survey of neutron-deficient rare earth isotopes, including lutecium isotopes. These workers observed the conversion electrons with permanent-magnet spectrographs, obtaining information about the low-energy transitions. From *K/L* and *L/M* conversion ratios Mihelich *et al.* were able to assign the multiplicity of certain of these. This enabled them to interpret certain transitions as being between rotational

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<sup>†</sup> National Science Foundation Predoctoral Fellow.

<sup>1</sup> G. Wilkinson and H. G. Hicks, University of California Radiation Laboratory Report UCRL-429, 1949 (unpublished).

<sup>2</sup> G. Wilkinson and H. G. Hicks, Phys. Rev. **81**, 540 (1951).

<sup>3</sup> Mihelich, Harmatz, and Handley, Phys. Rev. **108**, 989 (1957).

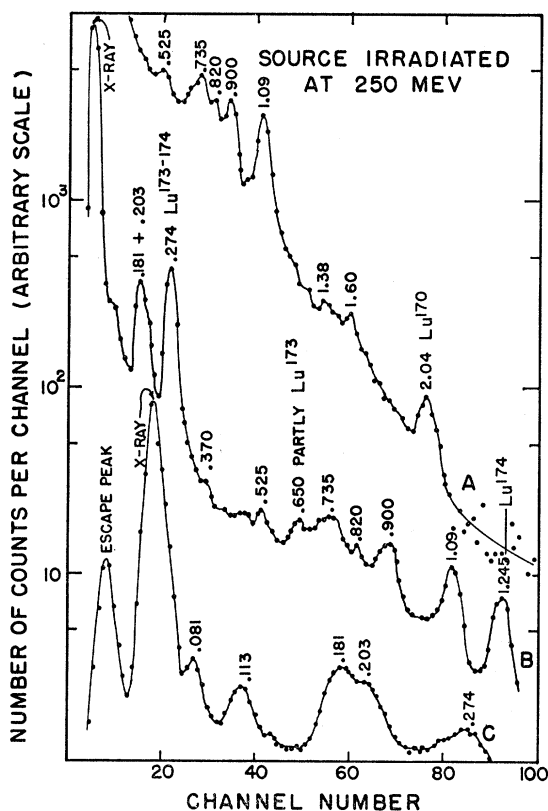


FIG. 1. Typical singles spectra of a source irradiated at 250 Mev at 3 different amplifier gains.

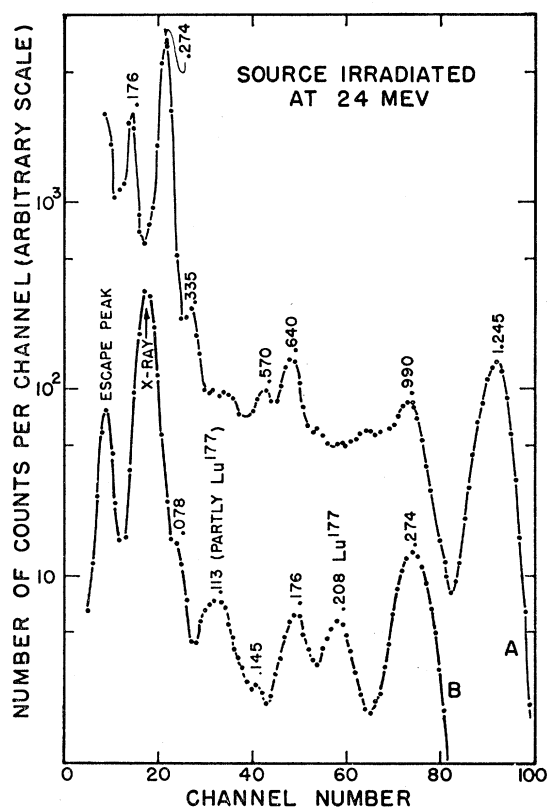


FIG. 2. Typical singles spectra of the source irradiated at 24 Mev at 2 different amplifier gains.

states. Such an interpretation was given to gamma rays of 0.0842 and 0.2778 Mev in  $\text{Yb}^{170}$ , 0.07865 and 0.2602 Mev in  $\text{Yb}^{172}$ , 0.0766 Mev in  $\text{Yb}^{174}$ , and possibly 0.0788 Mev in  $\text{Yb}^{173}$ . In addition, half-life measurements by Mihelich *et al.* were in agreement with those previously found by Wilkinson and Hicks.

Other investigators have studied certain aspects of the individual isotopes. An 0.0841-Mev transition has been observed in  $\text{Yb}^{170}$ , following the  $\beta^-$  decay of  $\text{Tm}^{170}$ .<sup>4,5</sup> This agrees with the result given by Mihelich *et al.* for the first excited state of  $\text{Yb}^{170}$ , following the electron capture decay of  $\text{Lu}^{170}$ . Nethaway *et al.*<sup>6</sup> investigated the  $\beta^-$  decay of  $\text{Tm}^{172}$ , observing gamma rays of 1.79, 1.44, 1.09, and 0.076 Mev associated with  $\text{Yb}^{172}$ . Two other less prominent gamma rays of 0.40 and 0.18 Mev were also mentioned by these workers. By means of Coulomb excitation, Heydenburg and Temmer<sup>7</sup> observed levels in Yb at 0.078 and 0.110 Mev. The 0.078-Mev peak was considered to be a composite of radiation which they assigned jointly to  $\text{Yb}^{170}$ ,  $\text{Yb}^{172}$ ,  $\text{Yb}^{173}$ ,  $\text{Yb}^{174}$ , and  $\text{Yb}^{176}$ . The 0.110-Mev transition was assigned to  $\text{Yb}^{171}$ .

<sup>4</sup> Pohn, Lewis, Talboy, and Jensen, *Phys. Rev.* **95**, 1523 (1953).

<sup>5</sup> Graham, Wolfson, and Bell, *Can. J. Phys.* **30**, 459 (1952).

<sup>6</sup> Nethaway, Michel, and Nervik, *Phys. Rev.* **103**, 147 (1956).

<sup>7</sup> G. M. Temmer and N. P. Heydenburg, *Phys. Rev.* **100**, 150 (1955).

In the present investigation,  $\text{Lu}_2\text{O}_3$  was bombarded with bremsstrahlung of peak energy 250 Mev from the University of Illinois 300-Mev betatron, in order to produce activities associated with  $\text{Lu}^{170}$  and  $\text{Lu}^{172}$  by  $(\gamma, 5n)$  and  $(\gamma, 3n)$  reactions, respectively. Comparison with previously known data gives no evidence that  $\text{Lu}^{171}$  activity was produced in observable amounts.  $\text{Lu}^{173}$  and  $\text{Lu}^{174}$  activities were produced by  $(\gamma, 2n)$  and  $(\gamma, n)$  reactions, respectively, using bremsstrahlung of peak energy, 24 Mev, from the University of Illinois 24-Mev betatron. Coincidence experiments involving all the observed isotopes were performed. However, the  $\text{Yb}^{172}$  gamma rays were studied in the most detail, and a proposed partial level scheme will be presented only for this isotope.

## II. APPARATUS

A total activity of the order of one microcurie was obtained from a 3.6-day irradiation (at 24 Mev) of a one-gram source of  $\text{Lu}_2\text{O}_3$ . Irradiations at 250 Mev not exceeding 7 hours were made so as not to obtain appreciable amounts of long-lived  $\text{Lu}^{173}$  and  $\text{Lu}^{174}$  activities. In addition, one 32-hour irradiation was made with a peak bremsstrahlung energy of 16 Mev so as to obtain mostly  $\text{Lu}^{174}$ , a  $(\gamma, n)$ -induced activity. The low specific activities of these sources precluded the use of a magnetic spectrometer.

All of the data obtained in the present work involved the employment of scintillation techniques in conjunction with a RIDL, Model 3300, 100-channel pulse-height analyzer and associated scintillation equipment. The apparatus and scintillation techniques used in the present study are described elsewhere.<sup>8</sup>

### III. DATA

#### (a) Energies and Lifetimes

Typical singles spectra of a source irradiated at 250 Mev and of the source irradiated at 24 Mev are shown in Figs. 1 and 2, respectively. In curve *A* of Fig. 1, the peak at 2.04 Mev is considered to be associated with the Lu<sup>170</sup> decay. The measured half-life of  $2.2 \pm 0.7$  days is in satisfactory agreement with the previously measured value of 1.7 days.<sup>2</sup> The peak at 1.09 Mev displayed a half-life of  $6.2 \pm 1.0$  days, and was attributed to the decay of Lu<sup>172</sup>. This agrees with the previously observed value of 6.7 days. Other gamma rays at 0.900,

0.820, 0.525, and 0.370 Mev in curves *A* and *B* of Fig. 1 were involved with the same decay scheme. In curve *C* of Fig. 1, all the prominent peaks are composite in nature. Yb<sup>170</sup>, Yb<sup>172</sup>, Yb<sup>173</sup>, and Yb<sup>174</sup> all have gamma rays of energy about 80 and 180 kev; hence all contribute to the 0.081-Mev and 0.181-Mev peaks in the figure. The peak at 0.113 Mev is due to the decays of Lu<sup>172</sup>, Lu<sup>173</sup>, Lu<sup>174</sup>, and Lu<sup>177</sup>. The last of these is formed via neutron capture by the naturally radioactive Lu<sup>176</sup> which is present to the extent of 2.3% in Lu<sub>2</sub>O<sub>3</sub>. The peak at 0.203 Mev arises because of Lu<sup>172</sup> and Lu<sup>177</sup>, while that at 0.274 Mev is attributed mainly to Lu<sup>173</sup>.

The peaks at 1.245 and 0.990 Mev in curve *A* of Fig. 2 were shown (by comparison with the activity of the source irradiated at 16 Mev) to be associated with the Lu<sup>174</sup> decay. All other peaks (the *K* x-ray excepted) in the curves of Fig. 2 are associated with the decay of Lu<sup>173</sup> unless otherwise noted. A considerable amount of the x-ray intensity in curve *B* originates in the decay

TABLE I. Summary of observed gamma rays.

	Obs. gamma ray energies (kev)	Relative intensity	Previously obs. gamma rays (kev)	References for previous data
Lu <sup>170</sup> ( $T_1 = 1.7$ days)	2040 ± 40		2000	1,2
	245 ± 5			
	190 ± 5		193.5	3
	83 ± 2		84.2	3,4,5
Lu <sup>172</sup> ( $T_1 = 6.7$ days)	1090 ± 10	52 ± 5	1200	1,2,6
	900 ± 5	26 ± 5		
	820 ± 7	9.3 ± 3		
	525 ± 10	19		
	370 ± 5	20 ± 6	373.1	3
	325		324.6	3
			270.5	3
	203 ± 5	19	203.8	3
	181 ± 5	29 ± 7	181.5	3,6
	113 ± 3	27	112.8	3
			90.6	3
79 ± 2	56 ± 10	78.7	3,6,7	
x-ray	1000 <sup>a</sup>			
Lu <sup>173</sup> ( $T_1 = 1.4$ yr)	640 ± 5	12 ± 3	800	1,2
	440			
	550 ± 10	4 ± 2		
	335 ± 10	3 ± 2		
	274 ± 3	150 ± 15	272.7	3
	176 ± 3	40 ± 10	171.5	3
	145 ± 5			
	113 ± 3	70 ± 15		
	79 ± 2	125 ± 20	100.9	3
	22 ± 2		78.8	3,7
x-ray	1000 <sup>a</sup>			
Lu <sup>174</sup> ( $T_1 = 165$ days)	1245 ± 5	27 ± 3	1000	1,2
	990 ± 10	4		
	275 ± 5	4		
	230 ± 10			
	176 ± 5	2		
	113 ± 3	13 ± 4		
	84 ± 2			
	77 ± 2	25 ± 7	76.6	3
x-ray	1000 <sup>a</sup>			

<sup>a</sup> The x-ray is arbitrarily assigned a relative intensity value of 1000 units.

<sup>8</sup> Henry, Dillman, Gove, and Becker, Phys. Rev. (to be published).

of  $\text{Lu}^{174}$ . This indicates that many of the  $K$ -capture transitions of  $\text{Lu}^{174}$  are to the ground state of  $\text{Yb}^{174}$ .

In Table I is found a summary of the observed gamma-ray energies (in keV for convenience) and their assignments to specific isotopes. Also included are relative intensity measurements, where determined, and a summary of previously known information.

### (b) Coincidence Measurements

In the case of  $\text{Lu}^{170}$  (data not shown), the 2.04-Mev gamma ray was found to be coincident with gamma rays at 0.190 and 0.083 Mev, interpreted<sup>3-5</sup> as transitions between rotational states of  $\text{Yb}^{170}$ . In addition, a gamma ray at 0.245 Mev was also shown to be in coincidence with the 2.04-Mev gamma ray.

$\text{Lu}^{172}$  is the isotope which was studied in the most detail. Hence the coincidence data are most extensive for this case. Figure 3 shows the radiation in coincidence with the 1.09-Mev gamma ray. Curve *B* indicates that the 1.09-Mev gamma ray is coincident with all the prominent low-energy transitions except the one at 0.274 Mev. The peak at 0.113 Mev is not relatively as intense in the coincidence curve as in the singles spectrum because a considerable amount of this peak

arises from 6.8-day  $\text{Lu}^{177}$ . Also, the peaks at 0.079, 0.181, and 0.203 Mev are partially due to coincidences with 1.09-Mev Compton events from the previously mentioned 2.04-Mev gamma ray. Curve *D* shows that a 1.09-Mev gamma ray is strongly coincident with 0.900 Mev and less strongly coincident with 0.820 Mev, 0.525 Mev, and 0.370 Mev. Coincidences with 0.900 Mev displayed a rather strong peak at 0.182 Mev as well as a peak at 1.09 Mev, as expected. No other peaks were observed which could not be explained as originating from 0.900-Mev Compton events of higher energy gamma rays.

The selected-sum coincidence method (described elsewhere<sup>8</sup>) was also employed for the study of coincidences among transitions of energy 0.525 Mev or lower. Figure 4 shows an example of this. There, curve *B* shows that 0.370 Mev and 0.525 Mev are in coincidence and, although these peaks are weak, they disappear completely when one moves off the sum-line energy. The two strong peaks in this curve are owing to the fact that the 0.181-Mev gamma ray is strongly coincident with 0.900 Mev; thus 0.181-Mev gamma rays plus Compton events of the 0.900-Mev gamma ray which add up to 0.900 Mev are also recorded. Curve *D*

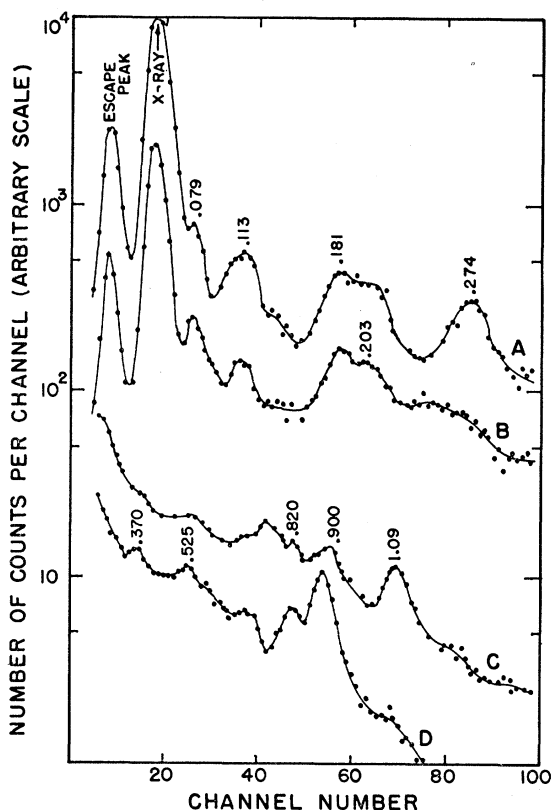


FIG. 3. Coincidence experiments with the  $\text{Lu}^{172}$  activity. *A* and *C*—singles spectra calibrations for curves *B* and *D*, respectively. *B* and *D*—coincidences with 1.09 Mev at 2 different amplifier gains.

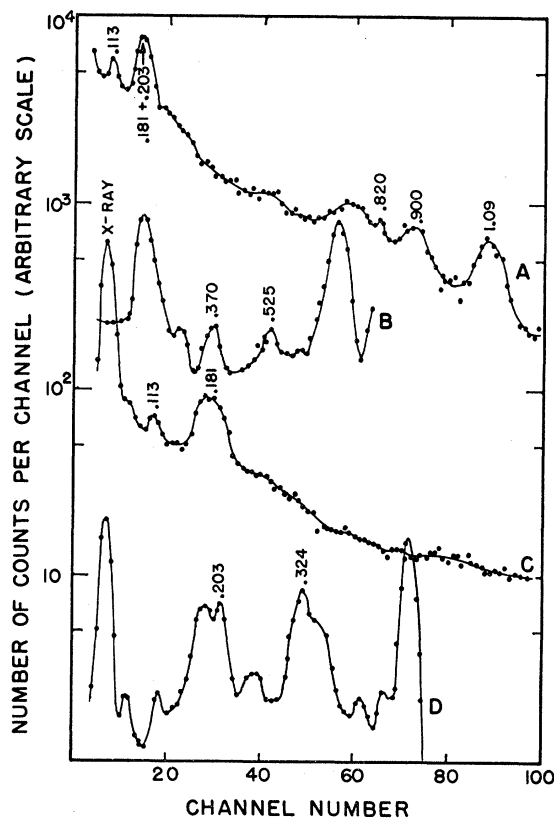


FIG. 4. Selected-sum coincidence experiments with the  $\text{Lu}^{172}$  activity. *A* and *C*—singles spectra calibrations for curves *B* and *D*, respectively. *B*—sum energy = 0.900 Mev. *D*—sum energy = 0.525 Mev.

TABLE II. Summary of coincidence data.

Lu <sup>170</sup>	83	190	245	2040						
2040	yes	yes	yes							
245										
190										
83										
Lu <sup>172</sup>	79	113	182	200	325	370	525	820	900	1090
1090	yes	yes	yes	yes		yes	yes	yes	yes	
900	no	no	yes			no	no	no		
820	yes	no	yes?	no?			no			
525		yes	yes			yes				
370		no	yes?							
325				yes						
200		yes?	yes?							
182	yes	yes								
113	yes									
79										
Lu <sup>173</sup>	22	79	113	145	176	274	440	550	640	
640		no	no	no	no	no	no	no		
550		yes	no	no	no	no	no	no		
440					yes	no				
274	no?	yes	yes	no?	yes	yes				
176	no	no	yes	yes	yes					
145		no	yes							
113	no	no?								
79	yes									
22										
Lu <sup>174</sup>	77	84	113	176	230	275	990	1245		
1245	yes	no	no	no?	no	no?	no			
990	no	no	no	yes	yes	yes				
275				yes?						
230										
176										
113		yes?								
84										
77										
β rays	no	yes	yes?							

of this same figure shows that 0.203 Mev is coincident with 0.324 Mev. These peaks arise partially from coincidences between 0.203 Mev and the high side (not completely off the peak) of 0.306 Mev. Coincident gamma rays of this energy exist in naturally radioactive Lu<sup>176</sup>. However, examination of the data reveals that the peaks are not entirely due to this cause. A summary of all coincidence data, including Lu<sup>172</sup> results, is tabulated in Table II.

Figure 5 illustrates two of the simple coincidence experiments performed with the Lu<sup>173</sup> activity of the source irradiated at 24 Mev. Curve *B* shows that 0.274 Mev is coincident with itself. The two gamma rays must be very close in energy since the resolution of this peak is about that expected for a single gamma ray. In curve *C* it is also to be seen that 0.176 Mev is coincident with itself. Other coincidence experiments indicate that the two energies may be about 0.172 and 0.176 Mev. The results of further Lu<sup>173</sup> coincidence experiments are tabulated in Table II.

Simple gamma-gamma coincidence experiments concerning the decay of Lu<sup>174</sup> were only possible where the single channel analyzer was set on either the 1.245-Mev or the 0.990-Mev gamma-ray peaks. Coincidences with 0.990 Mev showed strong peaks at 0.275, 0.230, and

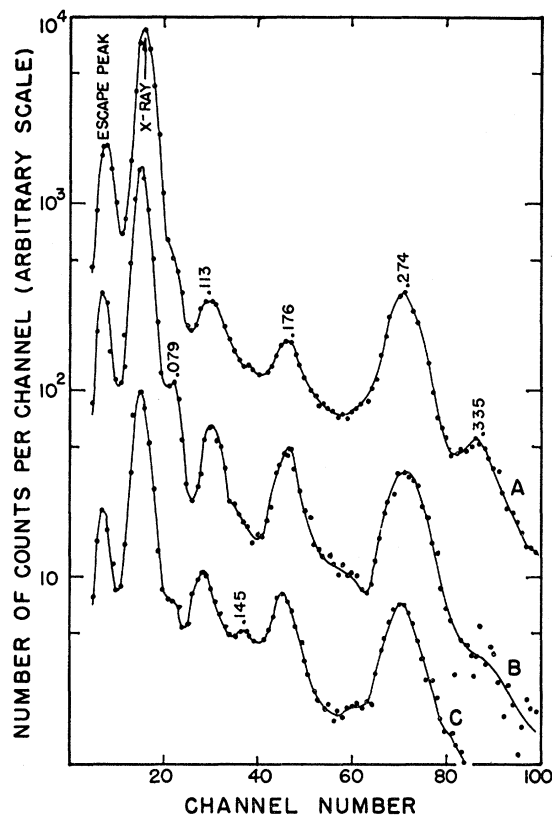


Fig. 5. Coincidence experiments with the Lu<sup>173</sup> activity. *A*—singles spectrum calibration for curves *B* and *C*. *B* and *C*—coincidences with 0.274 and 0.176 Mev, respectively.

0.176 Mev. Coincidences with 1.245 Mev revealed, in addition to the x-ray, a strong peak at 0.077 Mev.

Lu<sup>174</sup> was observed to decay both by electron capture and β<sup>-</sup> emission. A ½-mm thick by 1½-in. diameter stilbene detector was used to detect the beta particles. The end point of the highest energy beta group was observed to be approximately 1.2 Mev. This is the end point of the maximum-energy beta transition known to be associated with the natural radioactivity of Lu<sup>176</sup>. It was concluded that the maximum-energy beta transition associated with Lu<sup>174</sup> is equal to or less than about 1.2 Mev.

The gamma spectrum in coincidence with beta particles of energy above 0.25 Mev was also measured. Only beta particles above about 0.25 Mev were considered in order to eliminate conversion electrons of some of the strong low-energy transitions. The two highest energy peaks observed were at 0.203 Mev and 0.206 Mev. These are just the gamma-ray energies involved in the decay of Lu<sup>176</sup>. A peak at 0.084 Mev was interpreted as being the transition from the first excited rotational level in Hf<sup>174</sup> which had been populated through the β<sup>-</sup> decay of Lu<sup>174</sup>. Since the ratio of x-rays to 0.203-Mev and 0.306-Mev gamma rays of Lu<sup>176</sup> could be measured, and since the 0.084-Mev transition was the only other prominent peak, it was

possible to make a rough estimate of the  $K$ -conversion coefficient for the 0.084-Mev transition. After making all necessary relative absorption and relative efficiency corrections, the result obtained was  $\alpha_K \leq 2.5$ .

#### IV. DISCUSSION

##### (a) Proposed Level Scheme for Yb<sup>172</sup>

The proposed partial level scheme of Yb<sup>172</sup> is shown in Fig. 6. The numbers in parentheses are assigned relative intensities. The accurate energy values of the 0.9015-Mev, 0.8228-Mev, and 0.5284-Mev transitions were determined since they are crossovers of low-energy transitions whose energies have been previously accurately determined by Mihelich *et al.*<sup>3</sup> In agreement with Mihelich *et al.*, we interpret the 0.0787-Mev and 0.2602-Mev levels as the first and second excited rotational levels, respectively. The ratio of the energy of the second excited state to that of the first excited

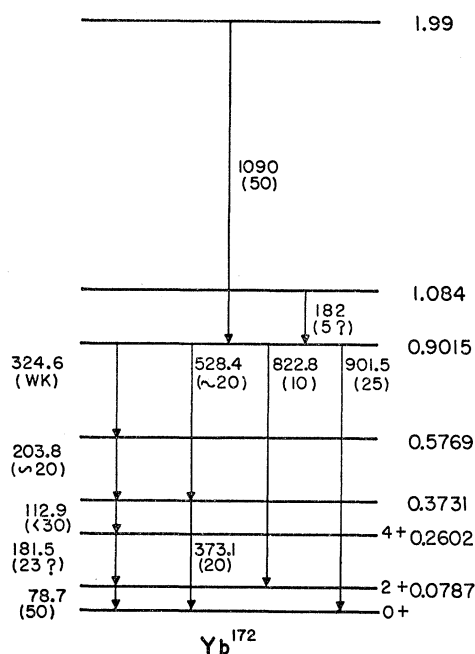


FIG. 6. Proposed partial level scheme of Yb<sup>172</sup>. All energies of transitions are in keV. The numbers (rounded off) in parentheses are the assigned relative intensities.

state is 3.31, which is in good agreement with the theoretically expected 3.33. The fact that the 0.9015-Mev gamma ray was in coincidence with a gamma ray at about 0.182 Mev necessitated the postulation of the level at 1.084 Mev. The apparent absence of transitions from the 0.373-Mev state to the 2+ rotational state is unexplained.

It will be noted that the 0.9015-Mev level decays to two members of the same rotational family, as well as to several other levels. In the limit of large deformations the transition probabilities to states of the same rotational family obey simple relations.<sup>9</sup> If the 0.9015-Mev level has a  $1\pm$  spin and parity, it can decay to either the 0+ or 2+ rotational levels with the same multipolarity transition. Employing the theory given by Bohr *et al.*,<sup>9</sup> we find that the ratio of the reduced transition probabilities is  $B(1\rightarrow 0+)/B(1\rightarrow 2+)=2.0$ , if  $K=1$  in the 0.9015-Mev state, where  $B$  is the reduced transition probability and  $K$  is the component of the total angular momentum on the nuclear symmetry axis. This agrees with the experimental ratio  $2.1\pm 0.3$ .

##### (b) Discussion of the Lu<sup>170</sup>, Lu<sup>173</sup>, and Lu<sup>174</sup> Data

Coincidence measurements confirmed the previously observed rotational states of Yb<sup>170</sup>, as well as the 0.0766-Mev first excited rotational level of Yb<sup>174</sup>. Attempts to fit the gamma rays of Yb<sup>173</sup> into a level scheme consistent with the coincidence data were not successful.

We have detected a previously unobserved first excited rotational level of  $84\pm 2$  keV in Hf<sup>174</sup>. For the first excited states of even-even nuclei in this region it has been observed that for a given neutron number the first excited states of isotones of higher  $Z$  are slightly higher.<sup>3</sup> This is borne out in this case by the fact that the first excited state of Yb<sup>172</sup> is 78.8 keV.

Finally, it was possible to make a rough estimate of the  $K$ -conversion coefficient for the transition from the first excited rotational level of Hf<sup>174</sup>. The result was  $\alpha_K \leq 2.5$ . (Sliv's value<sup>9</sup> is 1.2 for an  $E2$  transition.) One possible reason for our high upper limit was the assumption in the calculation that all observed x-rays were due to conversion of the 84-keV transition.

<sup>9</sup> L. A. Sliv and I. M. Band, Leningrad Physico-Technical Institute Report, 1956 [translation: Report 57ICCK1, issued by Physics Department, University of Illinois, Urbana, Illinois (unpublished)].