Electron Capture of Yb^{169} to Levels in Tm^{169}

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The decay of Yb^{169} by electron capture has been studied with permanent magnet spectrographs and scintillation counter techniques. Multipolarities of all transitions (including mixing) have been deduced. A level scheme is proposed. Two metastable levels of 4×10^{-7} and 4.5×10^{-8} second have been found, in addition to the well-known 6.4×10^{-7} sec level. These metastable levels are all depopulated by $D+Q$ mixtures in which the dipole radiation is greatly retarded (by factors of 10^6 to 10^8) and the quadrupole radiation is retarded by factors of 10^2 to 10^3 as compared to the single-particle estimates. For certain transitions the corrections to the $M1$ internal conversion coefficients (necessary because of finite nuclear size) have been obtained for the K, L_I and L_{II} shells. The screening correction for the K-LL Auger lines has been obtained for this nucleus ($Z=69$).

I. INTRODUCTION

It has been long known[1] that Yb (30.6-day)[2] decays via electron capture to levels in Tm, which are depopulated by a complex γ-ray spectrum. In addition, a metastable level of $\sim$0.7 μsec has been observed by several investigators.[1] However, no consistent decay scheme had been proposed because of the lack of sufficiently good experimental data and lack of knowledge of the behavior of level spacings and transition probabilities for nuclei in this region of Z and A. The recent striking successes of the Bohr-Mottelson[3,4] model, and the extensions to this model have made a reinvestigation of this complex level scheme quite worthwhile. The Coulomb excitation experiments[5] on Tm have indicated that the second excited state is at $\sim$130 kev, and by implication suggested that the first excited level is at $\sim$10 kev for this nucleus whose ground state spin is 1/2.[6] Johannson[7] has published a scintillation spectrometer study of the decay of Yb and has established that one may, indeed, construct a level scheme consistent with the theoretical premise that there exists here a rotational band ($K=1/2$) with spins 1/2, 3/2, 5/2, 7/2, and perhaps 9/2. In addition, there are levels not belonging to the $K=1/2$ band, and γ-ray transitions from these levels to levels in the rotational band are apparently greatly impeded. More recently, Cork *et al.*[2] have published the results of an investigation with high-resolution electron spectrographs. These results are in general agreement with those of Johannson[7] and Hatch *et al.*[8]

We have studied the radiations succeeding the electron capture of Yb with 180° permanent magnet photographic recording spectrographs and two scintillation spectrometers which could be operated with a variable time delay between them. We have obtained good values for the transition energies, in excellent agreement with those of Cork *et al.* and Hatch *et al.* We have resolved the L-conversion lines of most transitions, and have measured conversion line intensities on our photographic plates. We are able in all cases to assign multipole orders and intensities of the various transitions. These multipole assignments are also in agreement with those of Hatch *et al.*[8] We have confirmed the coincidence data of Johannson and have extended such measurements. Electron capture branching ratios have been obtained, which should give some information as to the character of the Yb ground state. In addition, we have established the existence of two additional metastable levels of half-lives 4.5×10^{-8} sec and 4×10^{-7} sec. We also have been able to determine empircally the corrections to $M1$ conversion coefficients, required due to the finite size of the nucleus.[9] We have calculated the correction required for the energies of K-LL Auger lines, this correction[10] being needed because the screening is changed when there exists a vacancy in one of the L subshells.

II. CONVERSION ELECTRON DATA

The spectrographs used were similar to those previously described by one of us,[11] and had fields of 82 and 180 gauss. The spectrographs had been carefully calibrated energy-wise against lines of known energy.[12] Electron intensities were obtained photometrically, and as a check, the relative intensities of conversion lines from a source of Ta^{182} were compared with those ob-

[1] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).
[2] Cork, Brice, Martin, Schmid, and Helmer, Phys. Rev. **101**, 1042 (1956).
[3] A. Bohr, Kgl. Danske Videnskab. Selskab, Mat-fys. Medd. **26**, No. 14 (1952).
[4] A. Bohr and B. R. Mottelson, Kgl. Danske Videnskab. Selskab, Mat-fys. Medd. **27**, No. 16 (1953).
[5] N. P. Heydenburg and G. M. Temmer, Phys. Rev. **100**, 150 (1955) and private communication from T. Huus to them.
[6] B. R. Mottelson and S. G. Nilsson, Phys. Rev. **99**, 1615 (1955).
[7] Sven A. E. Johannson, Phys. Rev. **100**, 835 (1955).
[8] Hatch, Marmier, Boehm, and DuMond, Bull. Am. Phys. Soc. Sec. II, **1**, 170 (1956).
[9] A. H. Wapstra and G. J. Nijgh, Nuclear Phys. **1**, 245 (1956).
[10] I. Bergstrom and R. D. Hill, Arkiv Fysik **8**, 21 (1954).
[11] Gillon, Gopalakrishnan, De-Shalit, and Mihelich, Phys. Rev. **93**, 124 (1954).
[12] Muller, Hoyt, Klein, and DuMond, Phys. Rev. **88**, 775 (1952), and Hatch, Boehm, and DuMond (private communication, 1955) to Nuclear Sci. data cards, No. 56-1-85.

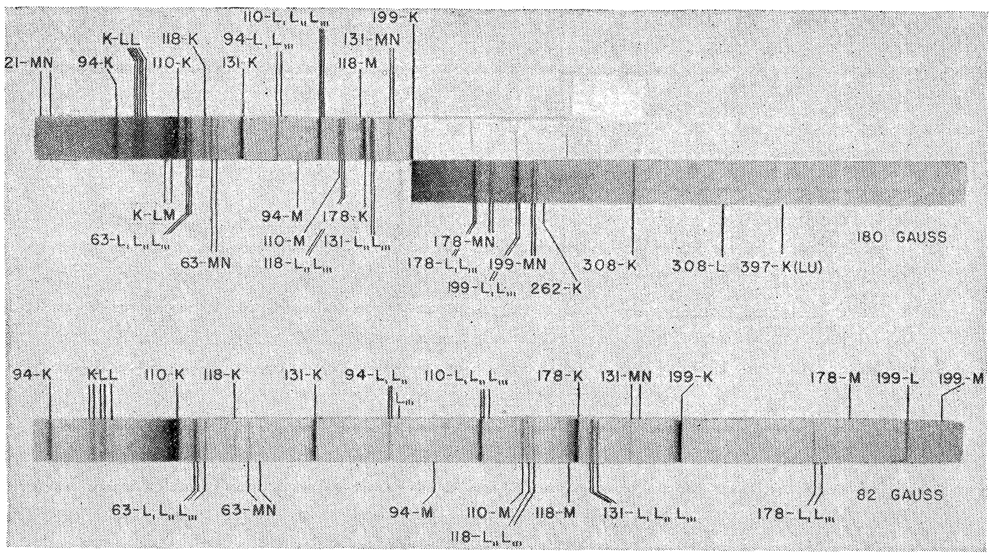

FIG. 1. Reproduction of internal conversion electron spectra of Yb^{169} as obtained with photographic recording permanent magnet spectrographs.

tained by Murray *et al.*[13] with a counter spectrometer. The agreement was good within 10% for electron lines of energy greater than 50 kev.

The sources were prepared as follows[14]: Yb_2O_3, after an irradiation of two weeks in the Argonne National Laboratory *CP*5 facility, was dissolved in 3.0*N* HCl and the solution evaporated to dryness over a steam bath. The chloride thus formed was dissolved in Pyridine to form the bath for electrodeposition. With a potential difference of 6 volts, free ytterbium was deposited from this bath onto a 10-mil Pt wire which served as the cathode. This wire was then used as the spectrograph source.

Figure 1 is a reproduction of the electron spectra obtained. Table I presents the conversion line energies, intensities, and assignments. Table II illustrates the consistency of the individual transition energies and their sums as compared to the appropriate crossover transitions.

III. ESTABLISHMENT OF MULTIPOLE ORDERS

In general, one attempts to determine the multipolarity of γ-ray transitions in a radioactive decay process by means of internal conversion data. In particular, these data are the absolute internal conversion coefficients (henceforth abbreviated as ICC) for the K and L shells and the relative ICC for the three L subshells (the L ratios) and the relative ICC for the M subshells, if such data are available. If one is able to compare these experimental data with accurate theoretical values of ICC,[15] then in essentially all cases, one can specify a unique multipole assignment. Such a procedure is feasible, at least for nuclei of sufficiently high Z and transition energies which are not greater than, say, 200 or 300 kev.

Unfortunately, it has become evident that there is an error in the theoretical values of the ICC for transitions of multipole order $M1$,[9] this error being due to the finite size of the nucleus. There is apparently a correction needed for the K, $L_{\rm I}$, and $L_{\rm II}$ conversion of this multipole order, and there are possibly corrections required for other multipole orders. We have assumed that, if there is a correction to the $L_{\rm III}$ ICC, it is small and can be neglected. These corrections, only roughly determinable at present, make the precise, or even correct, determination of dipole-quadrupole mixtures difficult. We have attempted to make our interpretation by adjusting the correction applied to the K, $L_{\rm I}$, and $L_{\rm II}$ shell ICC and the dipole-quadrupole mixing ratio so that the absolute ICC and the L ratios are internally consistent.

If the L ratios are not sufficiently sensitive to the unknowns, or perhaps not obtainable, then a directional correlation measurement would be useful in order to establish the multipole order (including mixing) and hence, the correction to the theoretical ICC.

In our calculations, we have assumed that the only multipole order requiring correction is $M1$, and in particular that the $E2$ ICC are correct. If one considers the case of $M1+E2$ mixtures, and makes the assumptions stated in the last paragraph, then the following procedure is possible. Let us call the correction to the K, $L_{\rm I}$, and $L_{\rm II}$ shells "f," "g," and "h," and the fractional part of the photons which go via an electric multipole transition "δ," the fraction going via a magnetic multipole transition being $(1-\delta)$. Then the

[13] Murray, Boehm, Marmier, and DuMond, Phys. Rev. **97**, 1007 (1955).

[14] Thanks are due Mr. Charles Armbruster who prepared the sources for us.

[15] Rose, Goertzel, Spinrad, Haar, and Strong, Phys. Rev. **83**, 79 (1951); M. E. Rose in *Beta and Gamma Ray Spectroscopy*, edited by Kai Siegbahn (North Holland Publishing Company, Amsterdam, 1955), Appendix IV, p. 905; Rose, Goertzel, and Swift (privately circulated tables).

correct value of the ICC, which we label with an asterisk, is equal to the appropriate correction factor times the uncorrected theoretical value. Thus,

$$\beta_K^* = f\beta_K,$$
$$\beta_L^* = g\beta_{L\mathrm{I}} + h\beta_{L\mathrm{II}} + \beta_{L\mathrm{III}}.$$

TABLE I. Conversion electron data for Tm^{169}.

Electron energy (kev)	Shell	Energy sum (kev)	Transition energy (kev)	Visual intensity estimate[a]	Intensity (photometer)	Remarks
5.84	M_{I}	8.15		vvw		Or $L_{\mathrm{I}}-M_{\mathrm{II}}M_{\mathrm{II}}$
6.10	M_{II}	8.19	8.17	vvw		Or $L_{\mathrm{I}}-M_{\mathrm{II}}M_{\mathrm{III}}$
18.43	M_{I}	20.74		vw		
20.51	N_{IV}	20.71	20.73	vvw		
42.98	M_{II}	45.07		vvvw		
44.62	N_{II}	45.00	45.0	vvvw		
3.65	K	63.05		vvw	(1900)	(Obtained using $\alpha_1^K=0.80$)
52.78	L_{I}	62.90		m-s	250	
53.30	L_{II}	62.92		m	90	
54.28	L_{III}	62.94		m	110	
60.78	M_{I}	63.09		m }	70	
61.20	M_{III}	63.09		vw }		
62.72	N_{I}	63.19	63.02	w	~20	
34.13	K	93.53		m-s	>270	
83.42	L_{I}	93.54		m	87	
83.96	L_{II}	93.58		vw	~16	
84.92	L_{III}	93.58		vvw		≤ 10
91.28	M_{I}	93.59		w }	33	
93.25	N_{III}	93.59	93.57	vw }		
50.20	K	109.6		vs	1750	
99.69	L_{I}	109.8		s	340	
100.2	L_{II}	109.8		w	60	
101.7	L_{III}	109.9		vw	~20	
107.5	M_{I}	109.8		m }	80	
107.8	M_{II}	109.9	109.8	vw }		
55.88	K	118.0		m	90	
108.7	L_{II}	118.3		w	54 }	$110-N$ is between these two.
109.5	L_{III}	118.2		w	50 }	
116.4	M_{III}	118.2	118.2	vw	(20)	Estimate. On low-energy tail of $177-K$
71.05	K	130.5		s	410	
120.5	L_{I}	130.6		w	50	
121.0	L_{II}	130.6		m-s	228	
122.0	L_{III}	130.6		m	180	
128.6	M_{II}	130.7		w }	83	
128.9	M_{III}	130.8		w }		
130.6	N_{II}	131.0	130.7	w	27	
117.9	K	177.3		s	680	
167.8	L_{I}	177.9		m	118	
169.1	L_{III}	177.8		vw		about 1/10 of L_{I}
175.4	M_{I}	177.7		vw }	28	
175.8	M_{III}	177.7		w }		
177.4	N_{III}	177.7	177.7	vvw		≤ 10
139.1	K	198.5		s	928	
188.5	L_{I}	198.6		m	140	
190.0	L_{III}	198.7		vw		about 1/10 of L_{I}
196.5	M_{II}	198.6		w	35	
198.4	N_{III}	198.7	198.6	vvw		≤ 10
202.7	K	262.1	262.1	vvvw		Estimate 3
249.0	K	308.4		w	34	
298.3	L_{I}	308.4	308.4	vvw		≤ 10

Auger lines

Experimental electron energy (kev)	Vacancies	Theoretical electron energy (kev)	Remarks
5.84	$L_{\mathrm{I}}-M_{\mathrm{II}}M_{\mathrm{II}}$	5.94	Or $8.15-M_{\mathrm{I}}$
6.10	$L_{\mathrm{I}}-M_{\mathrm{II}}M_{\mathrm{III}}$	6.14	Or $8.19-M_{\mathrm{II}}$
38.98	$K-L_{\mathrm{I}}L_{\mathrm{I}}$	39.16	
39.48	$K-L_{\mathrm{I}}L_{\mathrm{II}}$	39.66	
40.41	$K-L_{\mathrm{I}}L_{\mathrm{III}}$	40.62	
40.88	$K-L_{\mathrm{II}}L_{\mathrm{III}}$	41.12	
41.86	$K-L_{\mathrm{III}}L_{\mathrm{III}}$	42.08	
47.21	$K-L_{\mathrm{I}}M_{\mathrm{III}}$	47.39	
47.66	$K-L_{\mathrm{II}}M_{\mathrm{III}}$	47.89	
48.69	$K-L_{\mathrm{III}}M_{\mathrm{III}}$	48.85	
55.66	$K-M_{\mathrm{III}}M_{\mathrm{III}}$	55.62	
57.16	$K-M_{\mathrm{III}}N_{\mathrm{III}}$	57.17	

[a] v ≡very, w ≡weak, m ≡medium, s ≡strong.

TABLE II. Consistency of energy sums (kev).

109.8+8.2	118.0
Crossover	118.2
109.8+20.7	130.5
Crossover	130.7
177.7+20.7	198.4
Crossover	198.6
198.6+109.8	308.4
130.7+177.7	308.4
Crossover	308.4
198.6+63.0	261.6
Crossover	262.1
130.7+ 8.2	138.9
118.2+20.7	138.9
109.8+20.7+8.2	138.7
308.4+ 8.2	316.6
198.6+109.8+ 8.2	316.6
198.6+118.2	316.8
177.7+118.2+20.7	316.6
177.7+130.7+ 8.2	316.6

If one has determined the experimental K and L shell ICC (ϵ_K and ϵ_L), and the L ratios, then one has a sufficient number of independent data to determine the four unknowns (f, g, h, and δ), provided these experimentally determined quantities are sufficiently sensitive to the unknowns and the experimental data are sufficiently accurate.

We shall discuss our analysis of the multipolarities of the various transitions in some detail. The 130-kev transition has been taken to be pure $E2$ and we have assumed its correct ICC to be the theoretical value. We obtain the ϵ_i (experimental ICC for shell i) by normalizing our electron intensity data to the photon intensities of Johannson (assumed to be good to within 10%) at the 130-kev transition, and thus obtain ratios of conversion electron intensity for shell i to the photon intensity for each transition.

In general, the following relationship holds for a mixed transition:

$$\epsilon_i = \beta_i^* - \delta(\beta_i^* - \alpha_i),$$

where ϵ_i, β_i^*, and α_i are the experimental, corrected theoretical magnetic and theoretical electric conversion coefficients for electron shell i, and

$$\delta = \gamma_E/(\gamma_M + \gamma_E),$$

where γ_M and γ_E are the intensities of the magnetic and electric unconverted photons and $\gamma_M + \gamma_E = 1$. The theoretical conversion coefficients for the transitions under discussion appear in Table III.

(a) 63-kev Transition ($E1$)

The experimental value of $\epsilon_L = 0.19 \pm 0.04$ and the experimental L ratios 2.3/0.8/1.0 are consistent with the theoretical $E1$ ICC of 0.15 and theoretical L ratios

TABLE III. Theoretical internal conversion coefficients (ICC).

Transition energy (kev)	K shell α_1	α_2	β_1	β_2	L shell α_1	α_2	β_1	β_2	L ratios $E1$	$E2$	$M1$	$M2$
63	~0.80[a]	~2.5[a]	~4.0[a]	~58.0[a]	0.152	13.67	1.747	47.1	2.2/0.8/1.0	0.04/0.97/1.0	93/7.5/1.0	3.4/0.3/1.0
94	0.30[b]	0.89[b]	3.7[b]	35.0[c]	0.0513	2.08	0.535	8.3	3.3/0.85/1.0	0.13/1.1/1.0	94/7.7/1.0	4.4/0.5/1.0
110	0.20[b]	0.68[b]	2.4[b]	20.0[c]	0.0345	1.043	0.349	4.54	3.7/0.85/1.0	0.18/1.1/1.0	97/7.8/1.0	5.4/0.6/1.0
118	0.16[b]	0.58[b]	1.9[b]	15.0[c]	0.0281	0.780	0.294	3.48	3.9/0.86/1.0	0.20/1.1/1.0	98/7.8/1.0	5.7/0.66/1.0
131	0.13[b]	0.47[b]	1.5[b]	10.0[c]	0.0215	0.556	0.220	2.25	4.3/0.86/1.0	0.24/1.17/1.0	99/7.8/1.0	6.3/0.75/1.0
178	0.062	0.233	0.60	3.15	0.0095	0.138	0.091	0.69	5.4/0.92/1.0	0.48/1.17/1.0	100/7.9/1.0	8.8/1.0/1.0
199	0.046	0.168	0.44	2.1	0.0071	0.079	0.065	0.45	6.0/0.93/1.0	0.65/1.26/1.0	100/7.9/1.0	9.8/1.16/1.0
308	0.015	0.049	0.13	0.47	0.0022	0.014	0.019	0.09	9.0/1.0/1.0	1.87/1.64/1.0	110/8.1/1.0	16.4/1.85/1.0

[a] ICC obtained through extrapolation to threshold value obtained from B. I. Spinrad, Phys. Rev. **98**, 1302 (1955) and Church, Herbst, and Monahan Argonne National Laboratory Report ANL-5174, 1954 (unpublished), pp. 69–71.
[b] ICC taken from J. R. Reitz, Phys. Rev. **77**, 10 (1950).
[c] ICC obtained through extrapolation of results of Rose *et al.* (reference 14). All other ICC also come from Rose *et al.*

of 2.2/0.8/1.0. A small admixture of $M2$ (at most a few parts in 10 000) would be consistent with our data, as such an admixture would tend to increase the ICC and slightly enhance the L_{I} line. Such a mixture is compatible with the measured half-life of the level from which this transition proceeds.

One may exclude an $M1+E2$ mixture since ϵ_L is much too low for any possible such mixture. Therefore, we conclude that this transition is predominantly $E1$ with a small admixture of $M2$ possible.

(b) 94-kev Transition ($M1+E2$)

Intensity measurements were made on the L_{I} and L_{II} electron lines and along with the $L_{\mathrm{II}}/L_{\mathrm{III}}$ ratio reported by Cork,[2] the total L shell ICC was obtained, which is then 0.34 ± 0.05. Assuming that the theoretical $E1$ and $M2$ ICC's are correct, this value would correspond to a mixture of $E1+M2$ (3.5%). However, this mixture would make the L ratios 4.3/0.56/1.0 which is incompatible with the observed ratios of 10/1.8/1.0. On the other hand, neither can one reconcile the L ratios and L ICC with $M1+E2$, unless a correction is made to the $M1$ ICC. It is found that $g=0.55\pm0.09$, $h=0.63\pm0.08$ and $\delta=0.023\pm0.005$ are compatible with the ϵ_L and L ratios. In fact, only the stated values are consistent with these measured data.

We then conclude that this transition is $M1+E2$ (2.3%). In addition, it appears that this transition is one for which one is able to obtain the required corrections to the $M1$ ICC.

(c) 110-kev Transition ($M1+E2$)

For this transition, the values $\epsilon_K=1.27_{-0.25}^{+0.40}$, $\epsilon_L=0.30\pm0.06$, and the L ratios (10/1.8/0.6) indicate that the only logical assignment is $M1+E2$. Here again, unless one makes a correction to the $M1$ ICC, no satisfactory agreement with the experimental data is possible for any mixing ratio. The following values make all data internally consistent: $f=0.54_{-0.10}^{+0.15}$, $g=0.77\pm0.16$, $h=1.23\pm0.22$, and $\delta=0.024\pm0.006$. This multipole order is that expected if this transition proceeds between two levels of a rotational band.

(d) 178-kev and 199-kev Transitions

The establishing of the multipole orders of the three transitions de-exciting level E is difficult. Transitions ED and EC are certainly dipole plus quadrupole ($D+Q$) mixtures, but whether or not there is a parity change is difficult to ascertain.

By reference to Table IV it may be seen that the value of $\epsilon_K(178)$ lies between that for pure $M1$ and pure $E2$. If one takes the 178-kev transition as pure $M1$, a minimum value of f would be 0.65. This is somewhat higher than that which was found for the lower energy 110-kev transition. Any $E2$ admixture

TABLE IV. Transition data.

Transition energy (kev)	Photon intensity[a]	Transition intensity N_e+N_γ	ϵ_K[b]	Experimental ϵ_L	$L_{\mathrm{I}}/L_{\mathrm{II}}/L_{\mathrm{III}}$	Multipole order
63	250	4806[c]		0.19 ± 0.04	23/8/10	$E1+(M2)$
94	34	746	>0.83	0.34 ± 0.05	100/18/10[d]	$M1+E2$(2.3%)
110	144	3650	$1.27_{-0.25}^{+0.40}$	0.30 ± 0.06	167/30/10	$M1+E2$(2.4%)
118	···	379	···	···	–/110/100	$E2$
131	91	1828	0.47	0.53 ± 0.05	28/128/100	$E2$
178	146	2226	0.49 ± 0.10	$>0.084\pm0.017$	100/?/~10	$M1+E2$(10%)
199	214	3173	0.45 ± 0.09	$>0.068\pm0.014$	100/?/~10	$M1+E2$(13%)
308	60	619	0.059 ± 0.012	···	···	$E2$

[a] Obtained from Johannson (reference 7).
[b] All experimental ICC have been normalized to $\alpha_2^K(130)=0.47$.
[c] Obtained using $\alpha_1^K=0.80$.
[d] L ratio obtained using $L_{\mathrm{II}}/L_{\mathrm{III}}$ ratio of Cork *et al.* (reference 2).

would require an increase in the value of f since α_2^K is considerably smaller. However, our estimated L_I/L_{III} ratio of approximately 10 indicates an $E2$ admixture of $10\pm5\%$, where we have taken $0.5\leq g\leq1.0$. Such a mixture is consistent with the measured half-life from which this transition proceeds (see Sec. VI). For the extreme values of ϵ_K and δ the value of f is between 0.66 and 1.09. If a mixture of $E1+M2$ is considered, the data indicates an $M2$ admixture of $15\pm4\%$.

A similar situation exists for the 199-kev transition. Here again a minimum value of f is 0.81, which is higher than both the 110-kev and 178-kev transitions. The $E2$ admixture indicated by our data ($L_I/L_{III}\sim10$) is $13\pm6\%$, where we have considered g over the same range as in the 178-kev transition. The extreme values of ϵ_K and δ in this case yielded an f value between 0.85 and 1.43. A mixture of $E1+M2$ would require an $M2$ admixture of $20\pm5\%$. It may be noted that in both cases within the limits of our accuracy, the L_I/L_{III} ratio is compatible with either assignment.

In the absence of a good determination of the L ratio for both the above transitions, a clear-cut choice cannot be made.

(e) 308-kev Transition

We obtain $\epsilon_K=0.059\pm0.012$ which again indicates $D+Q$. However, if the 316.6-kev level has a (+) parity, it has, almost certainly, a spin $\geq7/2$, since the 316.6-kev ground state transition is, as an upper limit, 10% of the 308-kev transition. If the spins of the rotational band are correct, then the 308-kev transition must be pure $E2$ since an $M1$ transition cannot carry off the required two units of spin. The theoretical value of α_2^K is 0.049.

The observed ϵ_K is also compatible with an assignment of $E1+M2(10\%)$. In this case the 316.6-kev level has a (−) parity, and it has a spin=5/2, since all de-exciting transitions are $D+Q$ (change in parity).

IV. DELAYED COINCIDENCE MEASUREMENTS

The fast—slow apparatus consisted of two scintillation spectrometers and a moderately fast coincidence circuit which employed as input pulses the outputs from two linear amplifiers (Atomic Instruments Model 518). After suitable amplification and shaping, the pulses were sent down a tapped delay line which was grounded at some point. Pulses were picked off from each side of the delay line, added and then passed through a discriminator which rejected singles. Both the delay introduced and the resolving times could be varied. The delay introduced between two incoming pulses was variable in steps of 0.066 μsec. With the "prompt" radiation of Ta^{182}, the time delay curve had a slope corresponding to a decrease in counting rate by a factor of 50 per delay section introduced.

If one counter were made to view photons of 63 kev and the other viewed an integral spectrum of photons above 110 kev, the slope of the delayed coincidence curve (delaying 63-kev γ rays) corresponded to a half-life of 0.64 ± 0.04 μsec. This lifetime is undoubtedly that of the level at 316.6 kev, in agreement with the conclusion of Johannson.

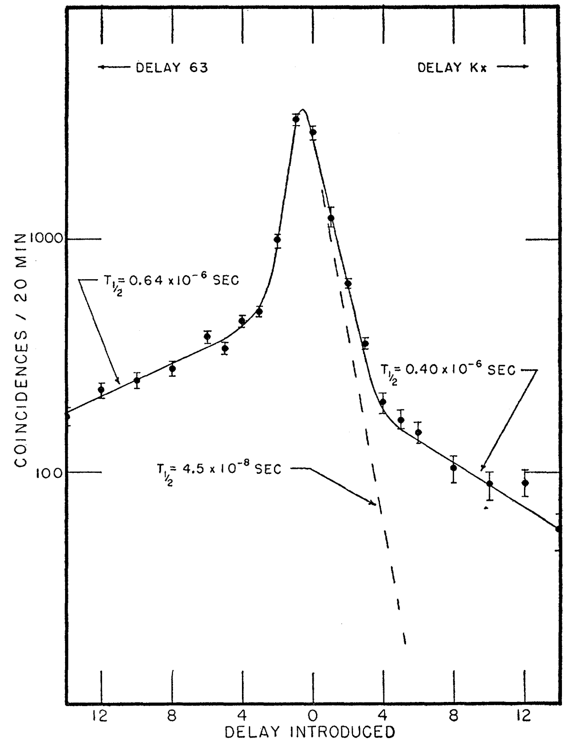

FIG. 2. Delayed coincidence curve with energy selection on K x-rays and 63-kev photons. Each section of delay line introduced corresponds to a delay of 0.066 μsec.

The evidence for the two additional metastable levels was obtained as follows. First, a thin (2 mm) NaI crystal (thin in order to minimize the Compton background due to the higher energy photons) viewed the 63-kev photons and a crystal of $1\times1\frac{1}{2}$ inches viewed K x rays. Figure 2 presents the results obtained when either counter's pulses are delayed with respect to the other. The delaying of the pulses from the x-ray counter results in a curve resolvable into two components corresponding to half-lives of 0.4 and 0.045 μsec. One should note that K-capture branches feed both the 63- and 94-kev transitions. The delaying of pulses from the 63-kev counter results in a decay component attributable to the x-rays arising from internal conversion occurring below the 0.64-μsec level at 316.6 kev. Interchange of amplifiers and delay lines was made in order to insure that the slopes obtained were real and not instrumental.

Secondly, the thin crystal viewed the 63-kev γ ray and the larger viewed the 94-kev γ ray. The fact that the 94-kev "window" also included some 110-kev photons was not deleterious since the 110-kev γ rays are already delayed with respect to the 63-kev γ rays

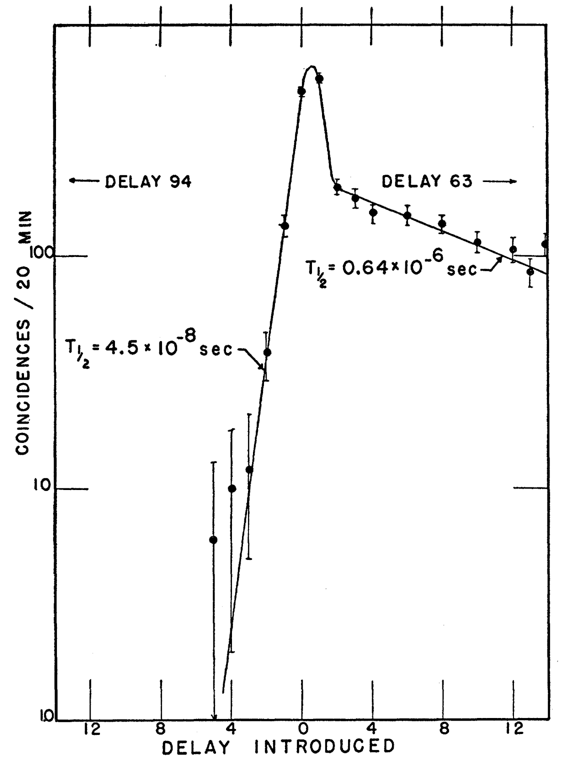

FIG. 3. Delayed coincidence curve with energy selection on 94- and 63-kev photons. Each section of delay corresponds to 0.066 μsec.

because of the metastable level at 316.6 kev. Figure 3 is the delayed coincidence curve. When 63-kev γ rays are delayed, a decay component of 0.64 μsec is observed, this being due to the fact that the 110-kev γ rays are included in the 94-kev window. The delaying of the 94-kev pulses results in a slope corresponding to a decay component of 4.5×10^{-8} sec, which should then be the half-life of the level from which the 63-kev transition proceeds. The premise that the 63-kev gate is free of x-rays is borne out by the fact that any component of 0.64-μsec decay is vanishingly small on the 94-kev delay side.

Therefore, we conclude that the levels G and H have half-lives of 0.4 and 0.045 μsec respectively. A discussion of the γ-ray transition probabilities will be presented in another section of this paper.

V. CONSTRUCTION OF THE DECAY SCHEME

It would seem that the levels at 8.4, 118.2, and 138.8 kev are the levels belonging to a rotational family with $K=\frac{1}{2}$ and ground state spin of $\frac{1}{2}$ (predicted +parity).[6] The Coulomb excitation experiments of Huus and of Heydenburg and Temmer[5] are in accord with this interpretation, as are the multipolarities of the transitions involved. Taking the energies of the first two excited states as 8.4 and 118.2 kev, one finds that the value of $\hbar^2/2\mathcal{I}$ (where $\mathcal{I}$ is the moment of inertia) is 12.39 and the value of a (coupling parameter) is -0.774. With these constants,[4] one finds that the predicted energy of the 7/2 level is 137.9 kev (as compared with the experimental value of 138.9) and of the 9/2 level is 335.7 kev. This indicates that a positive second order correction is needed to give our experimental value. There is some evidence for the existence of the 9/2 level, which if present, is very weakly populated. With regard to the multipolarity of the various transitions, apparently those of 118 and 131 kev are pure $E2$. The transition of 110 kev between the levels of spin 5/2 and 3/2 is $M1$ with 2.4% $E2$ admixture. There is no data available regarding the multipolarity of the 8.4-kev transition, while the 20.7 kev is predominantly $M1$ since Cork has observed only the $L_{\rm I}$ conversion line. We have obtained an estimate of the unconverted photon intensity of this latter transition by performing absorption measurements on the escape peak of the K x-ray which is at this energy. Since the escape peak will absorb like 50-kev photons, one is able to break the absorption curve into two components corresponding to 50 and 21 kev. In this way, we are able to estimate the 21-kev photons to be 2% as intense as the K x-rays.

The next level (316.6 kev) is not yet fully understood. As was mentioned in the section on multipolarities, one cannot unambiguously determine the parity of this state. However, one can say that if the parity is negative (assuming now that the parity of the ground

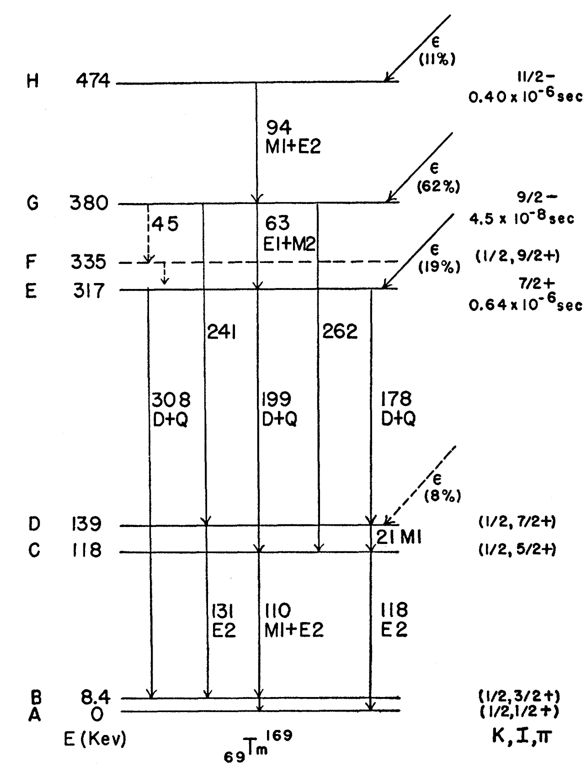

FIG. 4. Level scheme diagram for Tm^{169}. The half-lives for levels E, G, and H are shown beneath the preferred values of spin and parity.

TABLE V. Experimental and theoretical comparative lifetimes.

Transition	Assignment	Partial half-life[a] (sec)	Mixing of photons[b]	Comparative lifetimes $E1$ (−14.176)[c]	$M2$ (−8.079)[c]	$M1$ (−13.447)[c]	$E2$ (−8.204)[c]
178	$E1+M2$	1.78×10^{-6}	86% $E1$+14% $M2$	−6.244	−6.954		
	$M1+E2$		90% $M1$+10% $E2$			−7.752	−5.423
199	$E1+M2$	1.25×10^{-6}	80% $E1$+20% $M2$	−6.248	−7.054		
	$M1+E2$		90% $M1$+10% $E2$			−7.790	−5.267
308	$E1+M2$	6.42×10^{-6}	90% $E1$+10% $M2$	−5.163	−5.229		
	$M1+E2$		100% $E2$				−4.744

[a] Partial half-life calculated by using experimental ICC.
[b] The $E1+M2$ mixing has been obtained through a comparison of ϵ_K and theoretical K ICC, and the $M1+E2$ from the L_I/L_{III} ratio and an assumed value of $g=0.75$.
[c] The numbers in parentheses are the theoretical comparative lifetimes for single proton transitions.

state is positive), then the spin must be 5/2, since the three main transitions depopulating it are dipole-quadrupole mixtures and go to levels of spin 3/2, 5/2, and 7/2, and the selection rule on conservation of angular momentum is a rigorous one.

On the other hand, if the parity is positive, then one is almost forced to assign a spin of 7/2, since an upper limit of any 316.6-kev ground state transition is 10%, and one would find it difficult to explain why this $E2$ transition does not exist, as it should if the spin of the upper level were less than 7/2.

Accordingly, we shall discuss the levels above the one of 316.6, employing both alternatives of spin and parity. First, let us take this level as one of 7/2(+). Since the 63-kev transition is $E1$, the parity of the level G is (−). Transitions GD(241 kev)[8] and GC(262 kev) are both observed. The ratio of the intensities of the 262-K line and 199-K line is ~1/400. On the other hand, when one analyzes the photon spectrum in coincidence with photons of 110 kev, it is found that the photons of 262 kev are ~2% as intense as the 199-kev photons. Therefore $\epsilon_K(262)$ is roughly 0.06, which is consistent with either an $E2$ or $E1+M2$ assignment. No multipolarity data are available for the transition of 241 kev. Hence, the level G would most likely be 9/2(−). In view of the absence of transition HE, level H should be one of 11/2(−).

Level H has a measured half-life of 0.4 μsec. Taking the transition probability for the $M2$ crossover (HE) as 1/100 of the single-particle estimate, one finds that the crossover transition should be ~1% as intense as the 94-kev transition. We would not have observed a transition of this intensity.

The half-life of level H precludes the possibility of levels H and G being members of a rotational band, $K=9/2$. If such were the case, for the spins postulated, the value of $\hbar^2/2\mathcal{I}$ would be 10.4, which is to be compared with the value of 12.39 for the $K=1/2$ band.

Let us consider the case where the transitions ED, EC, and EB entail a change in parity. Then the spin and parity of level E are 5/2(−). Level G is 7/2(+) and level H is 9/2(+). This possibility does not appear to be as likely, since it is difficult to explain the absence of transition GB(371.4 kev) which should be of $E2$ character. Hence, we prefer the first alternative.

There is some evidence for the 9/2(+) level of the $K=1/2$ rotational band. We observe electron lines which apparently belong to a transition of 45 kev (transition GF). Johannson reported the existence of a photon of 194 kev. which could proceed between levels F and D. One may note that the energy of this postulated level is in very good agreement with that predicted.

On the decay scheme, (Fig. 4) we have indicated the intensity of the electron branches feeding the various levels. The branch to level D is in some doubt.

VI. GAMMA-RAY COMPARATIVE LIFETIMES

(a) 63 kev

The measured half-life of level G is 4.5×10^{-8} sec, and taking the total conversion coefficient as ~1.0, $\tau_\gamma=9\times10^{-8}$ sec. Then the logarithm of the comparative lifetime,[16] $\log\{\tau_\gamma A^{\frac{2}{3}}E_\gamma^3\}$, is −9.26 (considering pure $E1$), which is to be compared with the theoretical value[17] of −14.18. If one considers a possible admixture of $M2$ of 10 parts or 1 part per 10 000, then the log of the comparative lifetime for $M2$ radiation, ($\log\{\tau_\gamma A^{\frac{4}{3}}E_\gamma^5\}$), is −8.56 and −7.56 respectively, as compared with the theoretical value of −8.08. Hence a very small admixture of $M2$ radiation is consistent both with the conversion data and the lifetime measurements.

(b) 94 kev

This transition is $M1+E2$ (2.5%) and the half-life of the level which it depopulates is 0.4×10^{-6} sec. Here the value of $\log\{\tau_\gamma E_\gamma^3\}$ is −8.99, which is to be com-

[16] M. Goldhaber and A. W. Sunyar in *Beta and Gamma Ray Spectroscopy*, edited by Kai Siegbahn (North Holland Publishing Company, Amsterdam, 1955), Chap. XVI (II), p. 463.
[17] S. A. Moszkowski in *Beta and Gamma Ray Spectroscopy*, edited by Kai Siegbahn (North Holland Publishing Company, Amsterdam, 1955), Chap. XIII, p. 371.

pared with the theoretical single-particle value of -13.45. Hence, the $M1$ radiation is considerably slowed down. The $E2$ radiation has a value of $\log\{\tau_\gamma A^{4/3}E_\gamma^5\}$ of -6.48; the single-particle value is -8.20. The comparative lifetime is normal for an unenhanced $E2$.

(c) 178, 199, and 308 kev

The dipole component of these mixtures, regardless of whether it is of $E1$ or $M1$ character, is 10^6 to 10^8 times slower than the theoretical single-particle value, whereas the quadrupole component is 10^2 to 10^3 times slower which is not unreasonable for transitions which are not enhanced. The comparative lifetimes for both components of the above mixtures are shown in Table V.

VII. AUGER LINE SCREENING CORRECTION

For $Z=69$, the screening correction ΔZ was found to be 0.51 ± 0.15 for the L_{I} shell, 0.58 ± 0.15 for the L_{II} shell and 0.76 ± 0.20 for the L_{III} shell. These values are to be compared with those obtained by Bergstrom and Hill[10] for mercury ($Z=80$). They found $\Delta Z=0.55$ for L_{I} and L_{II} shells and 0.76 for the L_{III} shell.

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Angular Distribution of Fragments from Neutron-Induced Fission of U^{238} and Th^{232}†

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The angular anisotropy of neutron-induced fission in Th^{232} and U^{238} has been investigated with a double fission chamber whose common center high-voltage electrode was also a collimator. Ratios of $\sigma_F(0°,E_n)/\sigma_F(90°,E_n)$ were measured at a number of neutron energies from the thresholds near 1 Mev to 20 Mev. Larger variations were found in the anisotropy of fission fragments for U^{238} and Th^{232} than in earlier measurements for other nuclei, particularly at neutron energies immediately above the fission thresholds. For Th^{232}, there seems to be direct correlation between the variations in the anisotropy and those in the fission cross section. At 1.6 Mev, corresponding to a peak in the Th^{232} fission cross section, the angular distribution of fission fragments showed a maximum at 90° to the neutron direction. All previous measurements have shown maxima along the neutron direction.

INTRODUCTION

IN earlier studies[1-3] it was shown that the fragments of fission induced by high-energy neutrons were anisotropically distributed in the center-of-mass system. In the first experiments[1,2] the anisotropy was observed in varying degree for different nuclei bombarded with 14-Mev neutrons. These data were represented by angular distributions of the form $1+A\cos^2\theta$. More recent measurements[3] showed that the anisotropy changed considerably when the bombarding neutron energy was changed. In addition, more careful angular distribution measurements were fitted by $1+A\cos^2\theta+B\cos^4\theta$ with the $B\cos^4\theta$ term being dominant. The only study of angular anisotropy as a function of neutron energy[3] was for U^{235} which has a negative neutron energy threshold. It seemed useful to extend the work to include fissionable nuclei having positive neutron energy thresholds for fission. It has been suggested[4] that near the threshold the number of energy levels involved might be small enough to make possible quantitative theoretical analysis. The present work gives the measurements with U^{238} and Th^{232} in an effort to furnish pertinent data to assist in the theoretical interpretation of the fission process.[5-7]

EXPERIMENTAL METHOD

The collimated double fission chamber used in the present measurements was the same as that used in the previous work.[3] The details of the chamber, the associated electronic apparatus, the testing and calibration of the chamber, are all described in detail in reference 3, as well as experimental details concerning the neutron sources and background measurements. Since this information is identical for the present experiment, it will not be repeated here.

The Th^{232} foil having a thickness of 0.9 mg/cm² was evaporated on a 0.9-mg/cm² gold foil backing. Similarly, a 1.06-mg/cm² foil of depleted U^{238} was evaporated on a 1-mg/cm² gold foil backing. The angular distribution data were obtained by counting the relative number of fissions produced for different rotational positions of the chamber while the foils were

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1 W. C. Dickinson and J. E. Brolley, Jr., Phys. Rev. **90**, 388 (1953).
2 J. E. Brolley, Jr., and W. C. Dickinson, Phys. Rev. **94**, 640 (1954).
3 Brolley, Dickinson, and Henkel, Phys. Rev. **99**, 159 (1955).
4 Breit, Bohr, and Wheeler (private communications).
5 D. L. Hill and J. A. Wheeler, Phys. Rev. **89**, 1102 (1953).
6 Peter Fong, Phys. Rev. **89**, 332 (1953).
7 A. Bohr, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, June 1955* (United Nations, New York, 1956), Vol. II, p. 151.

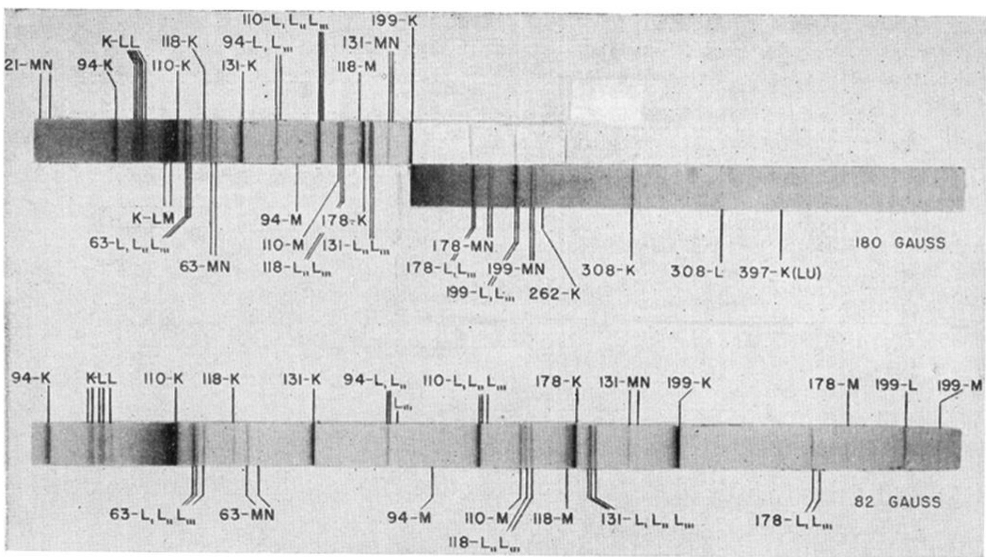

FIG. 1. Reproduction of internal conversion electron spectra of Yb^{169} as obtained with photographic recording permanent magnet spectrographs.