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THE SPECTRUM OF INTERNAL CONVERSION ELECTRONS ACCOMPANYING ALPHA DECAY OF U^{233}, AND THE LEVEL SCHEME OF Th^{229}

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The spectrum of internal conversion electrons emitted by Th^{229} (daughter nucleus of U^{233}) was studied in a β spectrometer with a toroidal magnetic field in conjunction with an e-α coincidence circuit. The existence of a rotational band starting from the ground state of Th^{229} was confirmed. The energies of the rotational levels were determined more accurately. Gamma transitions from some new levels (29.1, 71.4, 320.0, and 366.0 kev, and apparently 131 kev) were discovered and studied. The α spectrum of U^{233} was improved. The level scheme of Th^{229} is discussed.

1. INTRODUCTION

It is now well known[1,2] that the α decay of U^{233} results in the populating of the whole rotational band of levels of the daughter nucleus Th^{229}, starting from the ground state. Transitions to these levels are enhanced. The intensities of α transitions to the rotational levels of Th^{229} show a very definite anomaly;[2,3] they agree with the theory for the transitions to the first four levels (having spins of the successive levels equal to $5/2$, $7/2$, $9/2$, and $11/2$) and are in marked disagreement with theory for the next two levels ($13/2$? and $15/2$?). The conclusion that the last two levels are rotational then followed only from the agreement of the energy with that calculated from the rotational formula. Such agreement could, of course, be accidental.

The main purpose of the present work was to study the γ transitions between levels of Th^{229}. The study of the intensity and multipolarity of γ transitions enables one to determine the nuclear spin and the parity of the wave function in excited states, and thus to establish whether the level belongs to a particular rotational band.

Investigation of the γ transitions in Th^{229} is also of interest in another connection. The neighboring odd nuclei show a complex system of levels, resulting from the overlap of several rotational bands. In Th^{229} only one band was known, starting from the ground state. It was of interest to find out whether this simple picture is the result of the absence of other independent levels (in the low energy region) or is due to the weakness of α transitions to these other levels. There was every reason to suppose that levels which do not show up (or appear weakly) in the α-decay, will be populated by γ transitions from higher levels.

The investigation was carried out in a β spectrometer with a toroidal magnetic field.[4] The electronic circuit selected electrons emitted immediately following α decay (e^--α coincidences). We could use pulse height discrimination in the electron channel, and thus lower the counter background. An α spectrometer[1] was used to improve the α-spectrum of U^{233}.

2. PREPARATION OF SOURCES. CHEMICAL PURIFICATION OF URANIUM

The sources were prepared by vacuum deposition of uranium (in the form of the oxide) onto a thin sheet of mica (0.8 mg/cm^2). The deposition was carried out from a tungsten strip, onto which we first baked some tungsten powder. Depending on the intensity of the lines to be studied, the source activity was varied from 0.05 to 1 microcuries.

The first pictures of the conversion electron spectrum showed a large number of lines, which could not be explained on the basis of the known[1,2] α spectrum of U^{233}. The problem arose whether these lines actually belonged to the radiation of Th^{229}, following from α decay of U^{233}. To check this we carried out a purification of the material of possible radioactive impurities. First we knew that the U^{233} sample contained U^{232}, and consequently the whole radioactive series starting from it. It also contained Th^{229}, though in small quantity. We determined the U^{232} activity from the 238.6-kev γ transition which follows the α decay of the Pb^{212} daughter. This transition has been well investigated (cf. for example, reference 5). The activity proved to be extremely small, so that conversion lines appearing as the result of the α decay of the U^{232} itself could easily be eliminated from the U^{233} spectrum. We first purified the material of Th, and consequently of all its short-lived daughters. The separation of Th from U was done by precipitating the Th in a lanthanum carrier by the fluoride method.[6] Threefold repetition of this cycle gives a purification down to some hundredths of a percent of the initial content of Th in the U. By this same method we simultaneously also rid the U of Ac.[7] The intensity of the K-line of the 238.6-kev transition (Bi^{212}) served as an indicator of the degree of purification of the uranium from thorium. After purification, the intensity of this line was reduced by at least a factor of 20.

Later we carried out a purification of the uranium of protactinium as well as thorium and actinium. The thorium and actinium were separated from the uranium by using a lanthanum carrier by this same method. The protactinium was precipitated with a barium carrier by the following method. A solution of uranyl sulfate was made 1N in H_2SO_4 and 2N in HCl, and then 2 mg of barium carrier were introduced. $BaSO_4$ was precipitated, carrying with it radium and 70 – 80% of the protactinium.[8] After threefold precipitation, only 2 – 3% of the Pa remained in the solution. In order to get rid of all the Pa and to rid the uranium of weighable amounts of iron and aluminum, a Zr carrier was added to a sulfuric acid solution of uranyl sulfate, and a twofold precipitation with cupferron was performed (cf. reference 6). After removal of the zirconium and iron cupferrate from the Pa, the filtrate was steamed several times in concentrated HNO_3 to get rid of organic matter, the uranium was reduced to U^{4+} with amalgamated zinc in a reductor, and precipitated with cupferron.[9] The aluminum remained in the solution. The uranium cupferrate was burned to U_3O_8 and dissolved in nitric acid. The uranyl nitrate was converted to uranyl chloride and used for preparing sources. When the filaments were heated, the chloride was converted to oxide.

Chemical purification of the uranium resulted in no noticeable change in the appearance of the spectrum (except for the disappearance of the 238.6-kev conversion line from Bi^{212}). We thus demonstrated that the remaining electron lines in the spectrum were not part of the radiation associated with the presence in the sample of protactinium, actinium, thorium or the thorium series.

3. RESULTS OF MEASUREMENT

The most intense γ transitions in Th^{229} are in excellent agreement with the principal lines of the α spectrum, as found in references 1 and 2. According to our measurements, the excitation energies of the first three levels of Th^{229} which are known from the α decay are 42.4 ± 0.2, 97.3 ± 0.3, and 163.4 ± 0.4 kev. In measuring the energy, the following electronic lines were used for calibration: the L_{III} line of 51.7 (Pu^{239}); the L_{III} of 43.5 and the L_{II} and L_{III} of 100.0 (Pu^{238}); the L_{III} of 44.1 (Cm^{242}). (The conversion radiation follows the alpha decay of the nucleus listed in parentheses.) We found the following transitions between these levels (the complete list of lines is given in the table)

42.4±0.2(42.4—0)	80% $M1$+20% $E2$;
54.7±0.5(97.3—42.4)	$M1$+$E2$;
97.3±0.3(97.3—0)	$E2$;
66.0±1.0 (163.4—97.3)	—
121.0±0.3 (163.4—42.4)	$E2$.

The energies of the levels are in agreement with the rotational formula: the measured values of the energies are 42.4, 97.3, and 163.4 kev; the values calculated from the rotation formula (with $K = 5/2$) are 42.4, 96.8, and 163.5 kev. The energies of these three levels of Th^{229} and the multipolarities of the transitions between them show that together with the ground state they belong to a rotational band with $K = 5/2$.

If the level scheme of Th^{229} were exhausted by the levels found in references 1 and 2, and if all these lines were rotational, then the maximum energy of the conversion electrons would be 170 kev. But there are many lines with greater energy in the spectrum. (the conversion electron spectrum of Th^{229} is shown in Fig. 1). Analyzing this spectrum, we found various transitions which could not be explained by the scheme proposed in reference 2: 29.1, 71.4, 185, 245.3, 248.6, 277.8, 291.5, 317.3, 321, and 366 kev. The K-conversion lines of the 245.3, 291.5, and 317.3 kev transitions have an intensity of the order of 0.01%. From their L/K ratio, these transitions are most likely M1, though we cannot completely exclude E1 or M2, because of the poor statistics.

It is difficult to set up a level scheme for Th^{229} on the basis of these data because the last excited level, which was found in reference 1 and assigned to the ground rotational band in reference 2, has too high an energy (333 ± 5.0). In reference 11, the excitation energy of this level was given as 316 kev. A transition with this energy (317.3 ± 1.0) was also found in our conversion spectrum. The 245.3 and 71.4 γ lines (giving a cascade 71.4 + 245.3 = 316.7) are explained by a transition from this level. It also turned out that the α transition to the 240-kev level (237 kev in reference 2) has an intensity of about 0.007%. A check of our old data showed that the value of 333 kev[1] was the result of a numerical error. When this error was corrected, the energy became 320 kev.

The intensities of the 317.3 and 245.3 kev transitions (0.01%) are considerably less than the intensity of the 71.4 kev transition (0.3%). It was therefore suggested that the 71.4 kev level is populated mainly by α-decay. In this connection we undertook a new investigation of the α spectrum of U^{233} in the neighborhood of the main α peaks. For this purpose we had to use a very thin source and take long runs. Alpha lines corresponding to the transition to the 71.4 kev level of Th^{229} were actually observed (cf. Fig. 2).

We know from the α spectrum of U^{233} that the population of the 317 kev level is very small and amounts to only 0.04%. In order to find transitions from this level, we had to work with a thick source ($\sim 100\ \mu g/cm^2$) and reduce the resolving power of the spectrometer. Poor resolution and still insufficient statistics prevented us from determining directly the spin and parity of the 317-kev level from the conversion spectrum. (The ratios of the L and K conversion coefficients for E1, M1, and M2 transitions are not very different.) More definite conclusions concerning spin and parity can be made by comparing the intensities of the transitions with the population of the levels by α decay.

Conversion lines and γ transitions of Th^{229}*

Line Number	Electron energy, kev	Intensity, in % per α decay	Conversion	Energy of γ ray, kev		Multipolarity	Intensity, in % per α decay	Transition between levels
2	9.2	0.51	Auger					
3	12.0	0.51	L-$2M$					
4	12.75	0.42						
5	15.35	0.21	L-$2N$					
6	18.40	<0.05						
1	8.5	0.54	partly L_1	29.0				
9	24.2	0.27	M	29.3	29.1±0.2	$M1$?	0.7±0.4	29.1—0
11	28.1	0.15	N	29.4				
7	21.8	2.3	L_I	42.3				
8	22.6	4.5	L_{II}	42.4				
10	26.1	4.3	L_{III}	42.4	42.4±0.2	80% $M1$ 20% $E2$	16.0	42.4—0
13	38.0	4.0	M	42.5				
14	41.2	1.2	N	42.5				
12	34.6	0.6	L_I+L_{II}	54.7				
15	49.5	0.32	M	—	54.7±0.5	$M1+E2$	1.0±0.3	97.3—42.4
17	53.2	0.12	N	—				
19	61.6	—0.01	M	—	66.0±1.0		0.05	163.4—97.3
16 18 20 21	51.7 55.0 66.5 70.8	0.12 0.08 0.07 0.03	L_I+L_{II}? L_{III} M N	71.4 71.3 71.0 71.9	71.4±0.6		0.30	71.4—0
23	77.5	0.25	L_{II}	97.2				
24	81.0	0.18	L_{III}	97.3				
27	92.8	0.12	M	97.4	97.3±0.3	$E2$	0.6	97.3—0
28	95.7	0.03	N	96.8				
22	73.5	<0.005						

Conversion lines and γ transitions of Th^{229}* (continued)

Line Number	Electron energy, kev	Intensity, in % per α decay	Conversion	Energy of γ ray, kev		Multipolarity	Intensity, in % per α decay	Transition between levels
25	83.5	<0.005	L_{II}	103.2	103.0±1.0		0.01	(131—29.1) ?
26	86.6	<0.005	L_{III}	102.9				
29	101.2	0.012	L_{II}	120.9	121.0±0.3	$E2$	0.03	163.4—42.4
30	104.7	0.010	L_{III}	121.0				
31	116.4	0.007	M	120.9				
32	120.8	0.003	N	121.9				
33	124.7	0.001						
34	135.7	0.01	K	245.3	245.3±0.5	$M1$	0.015	317.0—71.4
42	225.2	0.003	L	245.7				
35	139.0	0.004	K		248.6±0.8	$M1$?	0.008	320—71.4
36	151.7	0.001						
37	168.2	0.002	K		277.8±1.5	$M1$?	0.005	320—42.4
38	181.9	0.008	K	291.5	291.5±0.5	$M1$	0.012	320—29.1
44	271.0	0.002	L	291.5				
39	198.0	<0.001						
40	207.7	0.012	K	317.3	317.0±1.5	$M1$	0.02	317.0—0
45	298.0	0.003	L	318.5				
41	212.0	0.004	K		321.0±1.5	$M1$?	0.008	320—0
43	256.5	0.0015	K	366.1	366.0±2.0	$M1$?	0.005	366—0
48	348.0	<0.001	L	368.0				
46	313.0	0.0015						
47	325.0	<0.001						

*Intensities of lines with energies below 30 kev are corrected for counter sensitivity.

The intensity of the transitions from the 317 kev level is 0.03%. This immediately excludes the possibility of an E1 transition, because of the smallness of the conversion coefficients ($e_K/\gamma = 0.05$). The only remaining possibilities are M1 or M2.

If we assume that the 317.3-kev transition is M2, then the level must have spin $1/2$ and parity opposite to that of the ground state. If the spin value were large, E1 transitions would be possible with greater intensity than the 317.3 kev transition (0.02% for M2), and the population of the level, as computed from the conversion spectrum, would exceed that obtained from the α spectrum by several times ten. We cannot ascribe a spin of $1/2$ to the 317-kev level because of considerations related to the intensity of α decay. In this case the α transition to the 317-kev level would be very highly forbidden, since the forbiddenness associated with the change of parity would be added to the forbiddenness associated with the large amount of angular momentum carried off by the α particle ($l \geq 3$). However experiment shows that the decay coefficient to this level is not too small ($F = 0.03$), while the coefficient for the known favored transition to the ground state is $F = 0.5$. Thus the additional forbiddenness amounts to 17. (For comparison we note that, for the α decay of Am^{241} the transition with change of parity and $l = 1$ is suppressed by a factor around 400.)

Therefore it is most probable that the parity of the 317-kev level is the same as that of the ground state of U^{233} and Th^{229}, and that the transition from this level to the ground state is M1.

In the conversion electron spectrum, only one

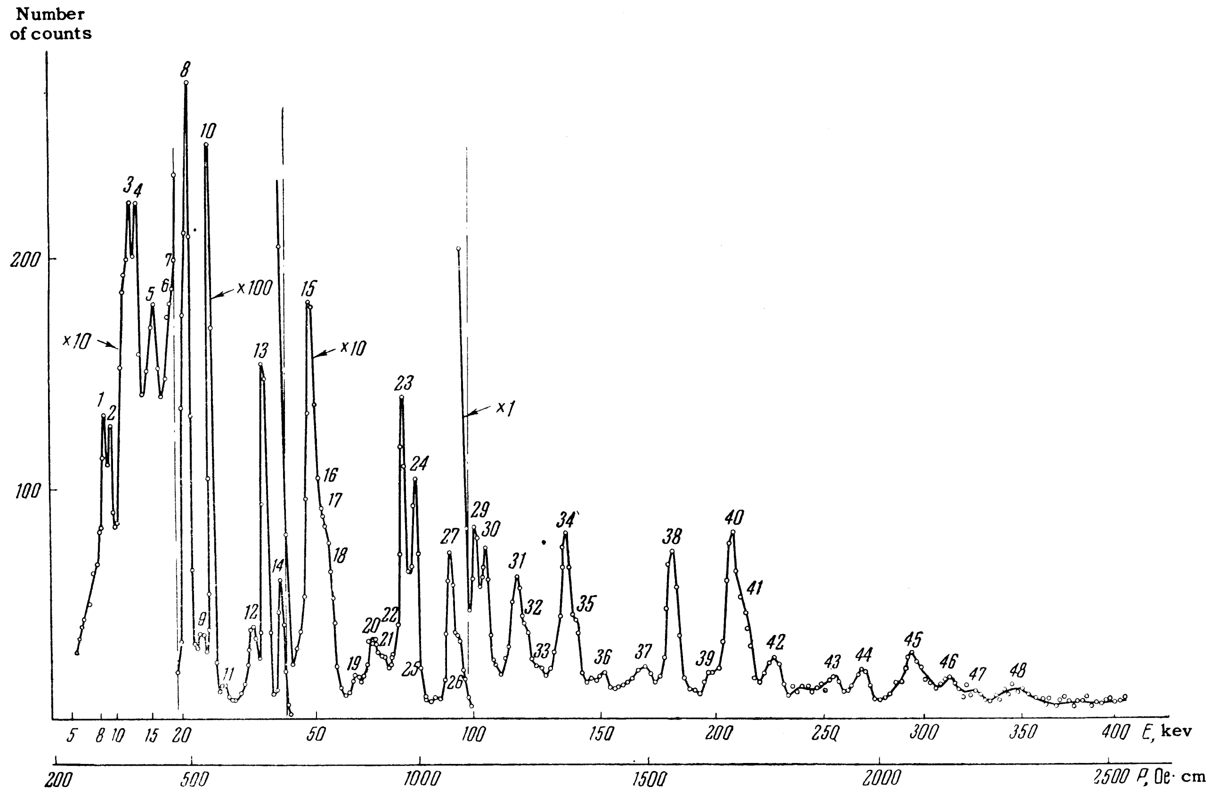

FIG. 1. Spectrum of conversion electrons of Th^{229}.

strong transition from the 317-kev level to a level of the rotational band was observed, namely the transition to the $5/2^+$ ground state. This should be the case if the spin of the 317-kev level is $3/2$. In this case the transitions to other levels of the rotational band will be E2, and their absence from the spectrum is not surprising (because of the lower probability of E2 transitions compared to M1, and their lower conversion coefficient).

In the conversion electron spectra, the K lines of the 245.3- and 317.3-kev transitions are broadened on the high energy side. We suggest that this broadening is caused by the presence of a level with excitation energy 320 kev, near to the 317-kev level. The following transitions are observed from this level: 1) 321.0 ± 1.5 kev; 2) 277.8 ± 1.5; 3) 248.6 ± 0.8; 4) 291.5 ± 0.5 kev, with a total intensity of about 0.02%. From the same arguments as for the 317-kev level, the parity of this level is positive. Just as for the 317-kev level, we can assert on the basis of the intensities of the conversion lines that the 321.0-, 248.6-, and 277.8-kev transitions are M1. The most probable value of the spin of the 320-kev level is $5/2$. The intensities of M1 transitions to the ground and first excited states of the rotational band should then be in the ratio 1 : 0.4.[10] This is in good agreement with our data on the intensities of the 321-kev (320 — 0) and 277.8-kev (320 — 42.4) transitions.

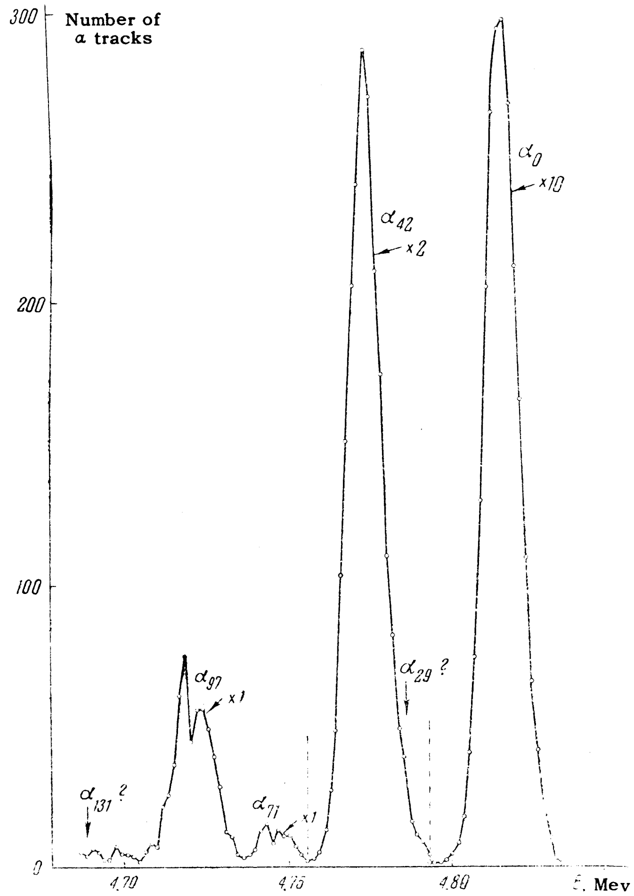

FIG. 2. Alpha spectrum of U^{233} in the region of the principal peaks.

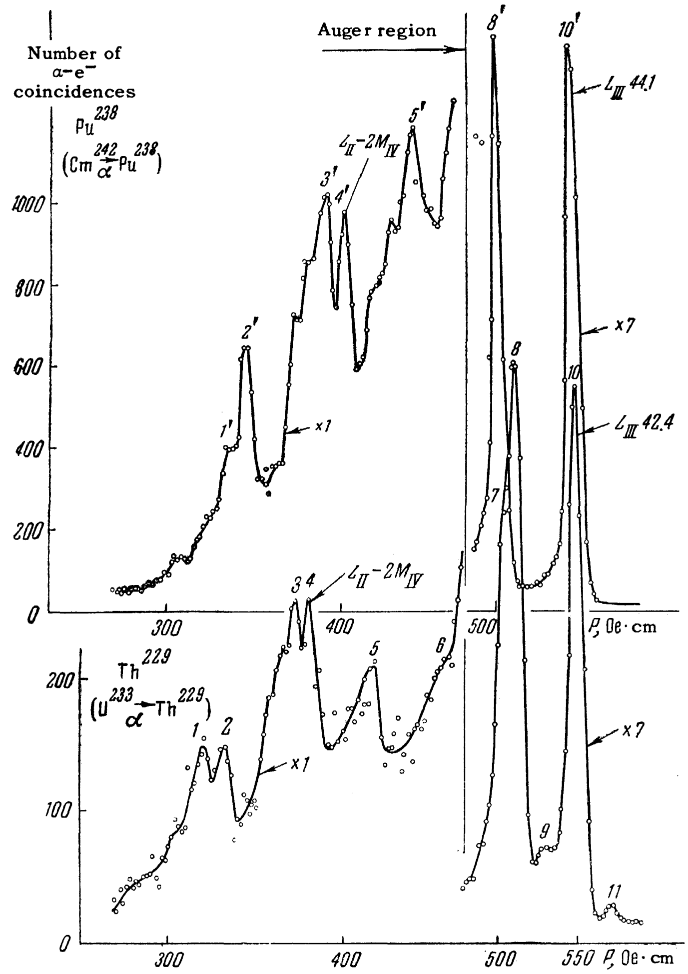

FIG. 3. Electron spectra of Th^{229} and Pu^{238} in the region of Auger transitions.

In the conversion electron spectrum there are relatively strong lines with energies 181.9 kev (0.008%) and 271.0 kev (0.002%). They are interpreted by us as K and L conversion of the 291.5-kev γ transition from the 320-kev level to the 29.1-kev level. The transition energy of 29.1 kev was determined from the electron lines (cf. Fig. 3): 1) L_I, 8.5 kev (0.5%); 9) M, 24.0 kev (0.27%); 11) N, 27.7 kev (0.15%). Line 1 at 8.5 kev lies in the region of the (L-2M) Auger electrons and could easily be interpreted incorrectly. To clarify the picture we made a comparison of the Auger spectra of the parent nuclei U^{233} and Cm^{242}. The corresponding spectra are shown in Fig. 3. As we see from the figure, the spectra are very similar everywhere, except in the region where line 1 is found, and there the intensity is considerably greater than at the corresponding line 1′ in Cm^{242}.

The intensity of the 29.1 transition is 0.7% as calculated from line 1, and is somewhat greater than 1% as calculated from lines 9 and 11. It is possible that there are γ transitions of 44.5 or 48.2 (M1), whose L lines increase the intensity of peaks 9 and 11. The shape of the α spectrum (cf. Fig. 2) also supports such a reduction in intensity of the 29.1-kev transition. Judging from the line α_{42}, the α decay intensity to the 29.1-kev level can hardly be greater than 0.5%. The transition at 48.2 kev (or 44.5) with an intensity no greater than 0.2% could be a transition from the first rotational satellite of the level with energy 71.4 kev. (The 71.4 kev level may be only the ground level of a band, since there is a strong transition going from it to the lowest state.)

Now we discuss possible assignments of spin and parity of the 71.4- and 29.1-kev levels. The parity of the 71.4- and 29.1-kev levels must be the same as the parity of the levels at 317 and 320 kev. In fact, the transitions from the 317 level to 71.4, and from 320 to 71.4 and 29.1 kev can only be M1 or M2 (from the intensities of the conversion lines). M2 transitions, which involve a change in parity, could not compete with the M1 transition to the ground state, and would simply not be seen. But in the experiment they have intensities of the same order as the transitions to the ground state.

The spin of the 71.4-kev level cannot be determined uniquely on the basis of our data. Transitions of type M1 go to the 71.4-kev level from the levels at 317 ($I = 3/2^+$) and 320 kev ($I = 5/2^+$). Thus the spin of the 71.4-kev level can be $3/2^+$ or $5/2^+$.

Transitions to the 29.1-kev level occur only from the 320 level. The energy of the transition is 291.5 kev (0.012%) and its multipolarity is M1. The spin of the 29.1-kev level must be great enough to forbid an M1 transition from the 317 kev level, for example, $7/2^+$.

The electron lines 43 and 48 are explained on the basis of the 366-kev transition with intensity 0.005% (for M1). Recently we received a report[11] that a line was observed in the α-spectrum of U^{233}, corresponding to a transition to the 364-kev level, and having an intensity of the order of 0.004%. This level is probably the rotational satellite of the 320 or 317 kev level. In this case, the spin of the 366 kev level should be either $7/2^+$ or $5/2^+$.

We also made measurements of the electron spectra for electron energies below 6 kev. (An accelerating voltage of 4 kev was applied to the source.) No difference from the usual Auger spectrum (M — 2N) could be detected.

In the α spectrum of the principal lines there is an indication of a level with excitation energy 130 kev and intensity about 0.05%. The existence of such a level is entirely possible. Electron lines

25 and 26 can be interpreted as the L_{II} and L_{III} conversion lines of the transition from the 131 level to the 29.1-kev level. Part of the intensity of line 37 at 168.2 kev could be interpreted as the L conversion line of a 185-kev transition from the 317 level to the level at 131 kev.

4. CONCLUSION

1. A study of the conversion electrons emitted by Th^{229} has shown that the levels: 0, 42.4, 97.3, and 163.4 kev belong to a rotational band with K equal to $^5/_2$. As pointed out in reference 3, the intensities of these lines are in good agreement with the theory of α decay. We found no γ transitions from the level at 240 kev (given as 237 kev in reference 2), which could be assigned to this same rotational band. The population of this level was apparently badly overestimated. According to the data of reference 11, it amounts to only around 0.007%. If this level is actually rotational and belongs to the main rotational band, the absence in our spectrum of transitions from this level is not surprising. The transitions would have energies of 77 kev (240 − 163) and 143 kev (240 − 97). To confirm or deny the presence of transitions with such low energy and so small an intensity (of the order of 0.001%) is not possible for us. According to our data, the levels at 317 and 320 kev do not belong to the main rotational band.

Thus the discrepancy between the calculated and experimental values of the intensity of α transitions to the levels at 237 and 317 (given as 333 in reference 2), which was pointed out in references 2 and 3, has been cleared up.

2. The level scheme of Th^{229} has proved to be much richer in excited levels than was apparent at first. A large number of the excited states of Th^{229} appear weakly in the α decay, since transitions to such levels must compete with the enhanced transitions to levels of the main rotational band.

3. A comparison of data from α and γ spectrometry enabled us to establish the energy, spin, and parity of various levels of Th^{229}. The excitation energies of levels in Th^{229} are: 29.1 ± 0.2, 42.4 ± 0.2, 71.4 ± 0.5, 97.3 ± 0.3, (131.0 ± 2.0)?, 163.4 ± 0.4, 240 (from reference 11), 317.0 ± 1.0, 320.0 ± 1.5, and 366 ± 2 kev. The level scheme is shown in Fig. 4.

The authors consider it their duty to express their deep gratitude to their co-workers who took part in the work of investigating the conversion spectrum of Th^{229}: G. I. Grishchuk, L. N. Kondrat'ev, Yu. N. Chernov, V. F. Konyaev, and S. V. Kalashnikov.

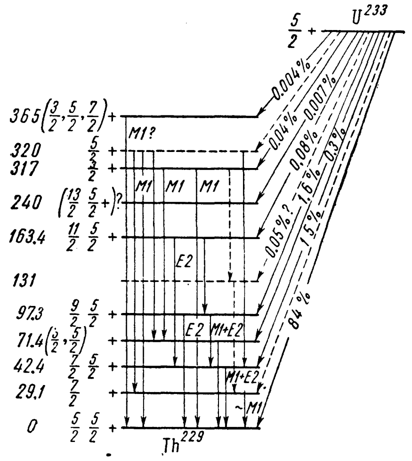

FIG. 4. Level scheme of Th^{229}.

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184