INTERNAL CONVERSION ELECTRON STUDY OF THE LOWER EXCITED LEVELS OF U^{235}

E. F. TRET' IAKOV, G. I. GRISHUK and L. L. GOL' DIN

Submitted to JETP editor November 15, 1957

J. Exptl. Theoret. Phys. (U.S.S.R.) **34**, 811-819 (April, 1958)

The internal conversion electrons emitted by excited U^{235} nuclei after α decay of Pu^{239} were investigated. Multipolarities of the transitions were established and the spins and parities of the first 6 levels were determined by comparing the results with the theoretical internal conversion coefficients for the L_I, L_{II} and L_{III} shells. It was shown that the first 5 levels form a rotational band with $K = \frac{1}{2}$. More precise values of the energies of the excited states were obtained.

INTRODUCTION

THE α spectrum of Pu^{239} is very well known at present and indicates that the daughter nucleus U^{235} possesses a number of excited levels. Table I is a list (taken from Refs. 1 to 3) of the first few excited levels of U^{235} as known from α decay and the probabilities of α decay to these levels.

TABLE I

No. of level	Excitation energy (kev)	Intensity of α decay (%)
0	0	72
I	13.2	16.8
II	51.7	10.5
III	84	$3.7 \cdot 10^{-2}$
IV	151	$1.3 \cdot 10^{-2}$
V	172	$5 \cdot 10^{-3}$
VI	234	$5 \cdot 10^{-3}$

These excitation energies are computed from the "zero" level of U^{235} (the lowest U^{235} level to which α decay goes). As was shown by Novikova et al.,[3] this level does not coincide with the ground level of U^{235}. α spectroscopy provides a reliable determination of the energies of the excited levels but does not generally establish their spins and parities. The γ transitions between levels must be studied for additional information.

The conversion and γ emission of U^{235}* have not been studied as thoroughly as the α decay of Pu^{239}. In Refs. 4 and 5 the internal conversion electrons of U^{235} were investigated by means of electron-sensitive plates. Only the most intense transitions of 35 and 53 kev were found (II — I and II — 0 in our notation). West and Dawson[6] obtained strong L lines, and also γ lines with the energies 37.7, 52.3, and 59.2 kev. The first two lines are due to the transitions II — I and II — 0; the origin of the last line is uncertain. Freedman, Wagner and Engelkemeir[7] observed only conversion electrons with the energies 31.4, 35.3, 47.1, and 50.2 kev, i.e., L_{II}, L_{III}, M and N + 0 electrons from the II — 0 transition. They also found γ rays with energies of 17.5 kev (100%), 35 kev (0.4%), 100 kev (1.1%), 124 kev (0.5%) and 384 kev (0.3%). These will be discussed later. Finally, Shliagin[8] detected a number of U^{235} conversion lines which can be explained completely by the transitions I — 0, II — I and II — 0. He also found strong 2.2-kev emission which he erroneously ascribed to K conversion of a 117-kev γ transition.

It is thus clear that we have extremely incomplete data as well as poor agreement in the case of weak lines. This induced us to make a detailed investigation of the conversion electron spectrum which accompanies the α decay of Pu^{239}

EXPERIMENTAL METHOD

The internal conversion electrons which accompany the α decay of Pu^{239} were studied by means of an iron-free β spectrometer with a toroidal magnetic field.[9] We studied the spectrum of electrons ejected immediately following α emission ($\alpha - \beta$ coincidences). The transmission of the instrument for electrons is 7% with resolution of 1% (the transmission for $\alpha - \beta$ coincidences is 2.5%).

In the study of low-energy electrons a potential of 10 kev was applied to the source. Acceleration did not cause broadening of the lines of even the slowest electrons. The α detector was a stilbene crystal placed directly behind the source and con-

*Here and hereinafter we are concerned with the radiation from U^{235} nuclei left in excited states after the α decay of Pu^{239}.

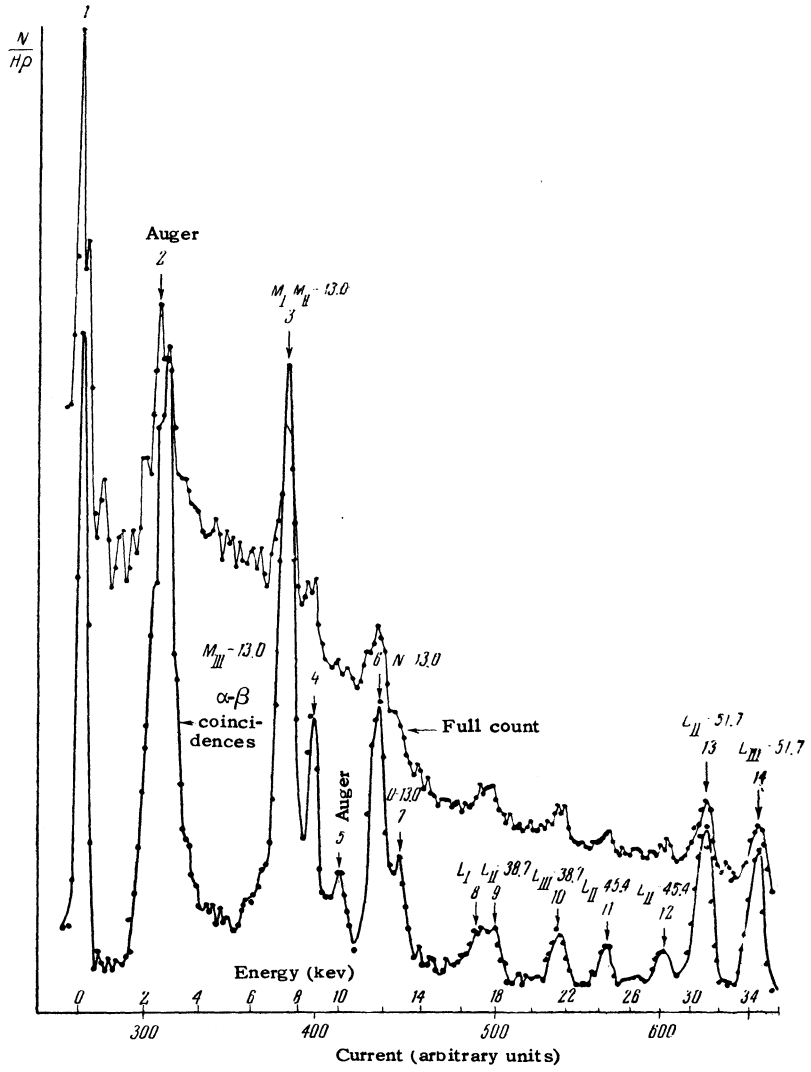

FIG. 1. Electron spectrum of U^{235} in the range 0 – 35 kev with accelerating voltage 7.0 kev.

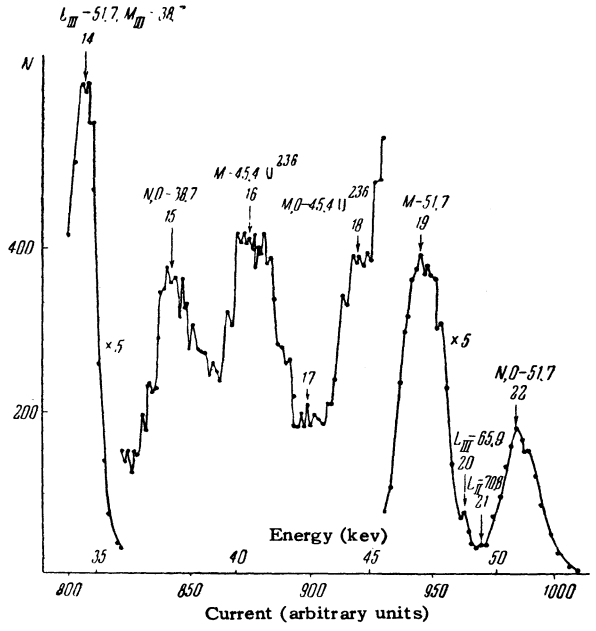

FIG. 2. Electron spectrum of U^{235} in the range 35 – 52 kev.

nected by a light pipe to a FEU-29 photomultiplier. We also used a scintillation counter to detect electrons, with a CsI(Tl) crystal for electrons with energies below 20 kev and stilbene at higher energies.

The source was prepared by vacuum deposition of the active material on a thin mica backing (0.8 mg/cm^2) which was coated with aluminum on the reverse side.

EXPERIMENTAL RESULTS

We investigated the conversion spectrum of U^{235} up to electron energy of 350 kev, but only in the 0 — 105 kev range were conversion lines found which appreciably exceeded the background energy level.

Figures 1, 2 and 3 show portions of the conversion spectrum, which was not fully corrected for the counter efficiency. We measured the counter efficiency in the range 0 — 35 kev and assumed it to be unity above 35 kev.

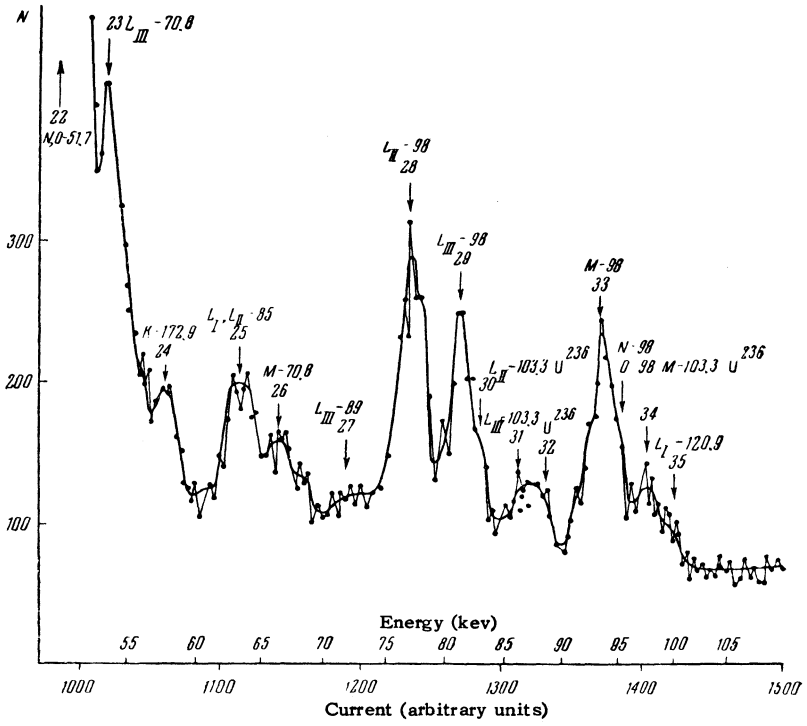

FIG. 3. Electron spectrum of U^{235} in the range 52 – 105 kev.

The electron energies and conversion line intensities are given in Table II. The line intensities were corrected for counter efficiency and are expressed as percentages relative to the number of Pu^{239} α decays. In determining conversion electron energies our reference lines were the L_{II} and L_{III} lines from the II – 0 transition with energy taken to be 51.7 kev and the L_{II} and L_{III} lines from the γ transition between the 143.3 and 43.5-kev levels of U^{234} (the daughter nucleus of Pu^{238}).

I. Levels I and II (13.0 and 51.7 kev)

Novikov et al.[3] have shown that levels I and II with excitation energies 13.0 and 51.7 kev (relative to the "zero" level of U^{235}) can be regarded as the first excited levels of a rotational band with $K = \frac{1}{2}$, beginning with the "zero" level. With this interpretation levels 0, I and II must have spins $\frac{1}{2}$, $\frac{3}{2}$, and $\frac{5}{2}$ and identical parity.

Almost all of the conversion lines corresponding to the transitions II – 0, II – I and I – 0 are clearly seen in the spectrum (Figs. 1 and 2). We determined the multipole orders of these γ transitions. Our experimental ratio of the conversion coefficients of the II – 0 γ transition in the L_{III} and L_{II} subshells is 0.88, which is in excellent agreement with the theoretical value of 0.87 (private communication from L. A. Sliv) for an E2 transition at the given energy. The II – I transition is mixed: 87% M1 + 13% E2 (for γ-emission probability). The multipole orders of the transitions provide acceptable confirmation of the rotational nature of levels II, I and 0

The M-conversion coefficients are unknown, but if we make the usual assumption that the conversion ratio for M subshells is equal to the corresponding ratio for L subshells it is possible to calculate the M1 and E2 contributions to the I – 0 transition. (In this case L radiation cannot be excited because the γ-ray energy is below the electron binding energy in the L shell.) For the I – 0 transition the experimental ratio is $(M_I + M_{II})/M_{III} = 10\%/3.6\%$, which corresponds to 99.5% M1 + 0.5% E2. (The L-conversion coefficients supplied to us by Sliv were extrapolated to 13 kev.)

II. Level III (83.8 kev)

According to Ref. 3, the 83.8-kev level is the third excited rotational level of a band with $K = \frac{1}{2}$. The spin must be $\frac{7}{2}$ and the parity must agree with that of the other levels of the same band.

We detected only one transition beginning at this level (the III – I transition). The transition energy of 70.8 ± 0.2 kev was determined from the L_{III} line (peak 23, Fig. 3). We were unable to determine the multipole order of this transition because its weak L peaks fall on the slope of the strong N + 0 peak of the II – 0 transition and reliable measurements could not be obtained. We were also unable to detect III – II transitions; this is

TABLE II. Internal conversion electrons which accompany the α decay of Pu^{239}

No. of peak	Electron energy (kev)	Intensity*	Transition type and energy		Levels between which transition occurs
1	0	~100	"Zero" peak		
2	2.59	~15	Auger M – 2N		
3	7.48	10	$M_I M_{II}$	13.0	13.0—0
4	8.62	3.6	M_{III}	12.92	13.0—0
5	9.79	1.0	Auger M – 2N		
6	11.68	4.4	N	13.1	13.0—0
7	12.88	1.6	O	13.2	13.0—0
8	16.70	0.65	L_I	38.5	51.7—13.0
9	17.83	0.76	L_{II}	38.8	51.7—13.0
10	21.63	0.76	L_{III}	38.8	51.7—13.0
11	24.43	0.61	L_{II}	45.4	45.4—0 U^{236}
12	28.28	0.54	L_{III}	45.4	45.4—0 U^{236}
13	30.82	2.8	L_{II}	51.7	51.7—0
14	34.47	2.46	L_{III}	51.6	51.7—0
15	37.5	0.14	N, O	38.8	51.7—13.0
16	40.7	0.19	M	45.4	45.4—0 U^{236}
17	42.8	0.01	?		
18	44.5	0.07	NO	45.4	45.4—0 U^{236}
19	47.0	1.4	M	51.7	51.7—0
20	48.7	0.02?	L_{III}	65.9	149.7—83.8
21	49.4	0.02?	L_{II}	70.3	83.8—13.0
22	51.0	0.6	N, O	51.7	51.7—0
23	53.6	0.0050	L_{III}	70.8	83.8—13.0
24	57.9	0.002	K	172.9	172.6—Ground level of U^{235}
25	63.3	0.003	$L_I L_{II}$	85	234.7—149.7
26	66.6	0.002			
27	71.9	0.0007	L_{III}	89	172.6—83.8
28	77.0	0.006	L_{II}	97.9	149.7—51.7
29	81.0	0.004	L_{III}	98.2	149.7—51.7
30	82.7		L_{II}	103.6	148.7—45.4 U^{236}
31	85.9	0.001	L_{III}	103.1	148.7—45.4 U^{236}
32	87.9	0.0008			
33	93.6	0.004	M	98.0	149.7—51.7
34	97.4	0.002	N, O	98.0	149.7—51.7
35	99.2	0.001?	L_I	120.9	172.6—51.7

*The intensity is corrected for the transition of the counter in the 0 – 35 kev region and given in percentages relative to the total number of P^{239} α decays.

easily explained by the fact that the conversion lines of these transitions fall in the region of the strong peaks of Fig. 1.

There is a noteworthy absence of the III – 0 transition, which we would easily have observed if its intensity were comparable with that of the III – I transition. This absence provides strong confirmation that the spin of level III is $\frac{7}{2}$. Indeed, the III – 0 transition must thus be of the M3 type and cannot compete with the E2 and M1 transitions from level III to levels I and II.

III. Level IV (149.7 kev)

This level, like the preceding level, was first found by Novikova et al.[3] through α rays and was interpreted as the fourth excited level of a rotational band with $K = \frac{1}{2}$. Its spin must be $\frac{9}{2}$ and its parity must agree with the parity of lower-lying levels.

We have found two transitions beginning at this level. Fig. 3 shows the L_{II} (peak 28) and L_{III} (peak 29) conversion lines which belong to the transition IV – II. From these lines the transition energy was determined to be 98.0 ± 0.3 kev. The experimental L_{III}/L_{II} intensity ratio is 0.76, which is somewhat higher than the theoretical ratio of 0.56 for an E2 transition but fits no other transition. The absence of conversion in the L_I subshell is additional evidence favoring an E2 transition.

Figure 2 shows a weak line (peak 20) of the IV – III transition (L_{III} with 65.9 kev). By a fortunate coincidence this line falls into the valley between the M and N peaks of the II – 0 transition. The other L lines of this transition are hidden by the M peak of II – 0. We cannot see conversion lines corresponding to IV – I and IV – 0 transitions, and if they exist their intensity is less than $\frac{1}{10}$ of the IV – II intensity. On the basis of the foregoing data, we can confidently

state that level IV belongs to the rotational band which we are considering and has spin $^9/_2$.

As already mentioned, a 100-kev γ line in the γ spectrum of U^{235} was reported in Ref. 7. This line is easily explained by the IV — II transition; its intensity compared with the 17.5-kev line is highly exaggerated in Ref. 7.

IV. Level V (172.6 kev)

This level was first observed by Asaro and Perlman through $\gamma-\gamma$ coincidences (private communication). According to Ref. 3, the intensity of the α transition to level V is 5×10^{-3}%. We observed a few weak conversion lines which can be ascribed to transitions from this level, which has an energy of 172.6 ± 0.5 kev. In considering transitions from level V, we are struck by the absence of L-conversion electrons from V — I transitions and the considerable number of such electrons from V — II. The absence of L electrons from V — I cannot be explained by reduction of the L-conversion coefficient with increasing transition energy; higher probabilities of γ transitions with increasing energy compensate the reduction of the conversion coefficient. The most reasonable explanation is the large spin of level V.

Our results can be interpreted clearly by assigning spin $^7/_2$ to level V and a parity which is opposite to that of the rotational band and agrees with the parity of the ground level (but not zero level) of U^{235}. The transition to level I must then be of type M2 and cannot compete with the E1 transition to levels II and III. We interpret peak 27 to be an L_{III} peak of the V — III transition. The L_I and L_{II} lines fall into the region occupied by peak 26 and cannot be seen. Peak 35, which is on the slope of peak 34, can be ascribed to a V — II transition (a L_I line). In the E1 transition the L_{II} and L_{III} lines are considerably weaker than L_I and are thus not observed. The small conversion coefficient for E1 transitions (K + L = 0.27) explains the strong 120-kev γ radiation which has been noted by a number of investigators (Ref. 7 and a private communication from Asaro and Perlman).

We ascribe peak 24 to the K conversion of the transition from level V to the ground level of U^{235}. E0 and M1 transitions are possible here, and estimates show that the first of these possesses considerable probability. The large conversion coefficient for M1 transitions and, possibly, the considerable contribution of the 100% converted E0 transition explain why no one has observed thus far the corresponding γ rays. The absence of L electrons is due to the large K/L ratio for E0 and M1 transitions.

We note in conclusion that our argument in favor of spin $^7/_2$ is based on an interpretation of very weak and inadequately investigated transitions. This interpretation cannot therefore be considered final.

V. Level VI (234.7 kev)

Peak 25 of Fig. 3 (electron energy 63.4 kev) cannot be ascribed to any one of the foregoing transitions. This peak is wider than the peaks of single lines. It could be explained as resulting from a transition between level III (83.8 kev) and the ground level (not the zero level) of U^{235}. However, this seems very unlikely since the intensity of this transition must be small as a result of nuclear readjustment and peak 25 is only one-half smaller than peak 23 (of the L_{III} transition III — I).

We believe that peak 25 is most probably associated with a transition between levels VI and IV. The energy obtained for level VI is 234.7 kev. We find no transitions from level VI to other levels of the rotational band; this may be ascribed to a large spin of level VI (such as $^{11}/_2$).

SEARCH FOR AN ISOMERIC TRANSITION

We know from Ref. 10 that the spin of U^{235} in the ground state is $^7/_2$, whereas the α decay of Pu^{239} goes to the "zero level" of U^{235}, which undoubtedly has spin $^1/_2$. The large spin difference and close energies of the ground and zero levels suggest that the "zero state" is an isomeric state of U^{235} (Ref. 3).

In Shliagin's[8] study of the conversion spectrum of U^{235}, he found an intense 2.2-kev line which he ascribed to γ radiation of 3 kev. On the basis of Shliagin's work, Novikova et al.[3] suggested that the "zero peak" of U^{235} is separated by 3 kev from the ground level.

We have carefully investigated the conversion radiation from U^{235} in the soft energy region and have actually found a strong peak in the region 2.4 — 2.6 kev. However, we have found similar peaks with comparable intensities for Pu^{238}, Cm^{242}, Am^{241} and even Po^{210}, where there are certainly no 3-kev transitions. This peak is undoubtedly associated with the Auger effect, which occurs in emission from excited levels (the larger part of the effect) and in auto-ionization of atoms accompanying α decay (the smaller part of the effect).

We were unsuccessful in our search for electrons from the isomeric transition. These electrons are most likely concealed by the strong (~100%) peak of "zero"-energy electrons which

was always present in our experiments and was even observed in control sources lacking the active material (aluminum-coated mica). The energy of electrons from the isomeric transition is probably so small that they practically do not emerge from our sources of 0.8 mg/cm^2 thickness. This was confirmed by Asaro and Perlman, who reported[12] that they succeeded in finding soft radiation from U^{235} which was almost completely absorbed in a source of 0.7 mg/cm^2 thickness.

EXCITED LEVELS OF U^{236}

An admixture of Pu^{240} (6.5% of the activity) in our sources enabled us to investigate the conversion electrons emitted by the daughter nucleus U^{236} The L_{II} and L_{III} lines (peaks 11 and 12, Fig. 1) permit a reliable determination of the energy and spin of the first excited level of U^{236}. The energy of the level is 45.4 ± 0.2 kev. The emission in question is of type E2, so that the 45.4-kev level has spin 2 and positive parity. The L_{III} line for the transition from the second to the first excited level gives 148.7 ± 0.4 kev for the energy of the second excited level.

Our source also contained some Pu^{241}. We observed only a continuous β spectrum associated with the β transition $Pu^{241} \rightarrow Am^{241}$.

CONCLUDING REMARKS

Our results definitely show that the zero, I, II, III, and IV levels of U^{235} (see Fig. 4) belong to a rotational band with $K = 1/2$. We observed a number of electromagnetic transitions between these levels and transitions from the nonrotational level V ($7/2^-$) to various levels of the rotational band.

The probabilities of transitions of the same multipole order to levels in a single rotational band are related by the simple theoretical formula:[13]

$$\frac{B(L,\ I_i - I_f)}{B(L,\ I_i - I_f')} = \frac{\left| C^{I_f K_f}_{I_i K_i;\ L,\ K_f - K_i} \right|^2}{\left| C^{I_f' K_f}_{I_i K_i;\ L,\ K_f - K_i} \right|^2},$$

where B is the reduced probability of a γ transition, which depends on the multipole order L of the γ radiation and the difference between the spin I_i of the initial nucleus and I_f of the final nucleus. The right-hand side of this equation contains the Clebsch-Gordan coefficients C. The probabilities B are related to ordinary probabilities by the formula

$$T = \frac{8\pi\,|\,L+1\,|}{L\,[(2l+1)!!]^2}\,\frac{1}{\hbar}\left(\frac{\omega}{c}\right)^{2L+1} B(L).$$

Our experimental results permit a comparison

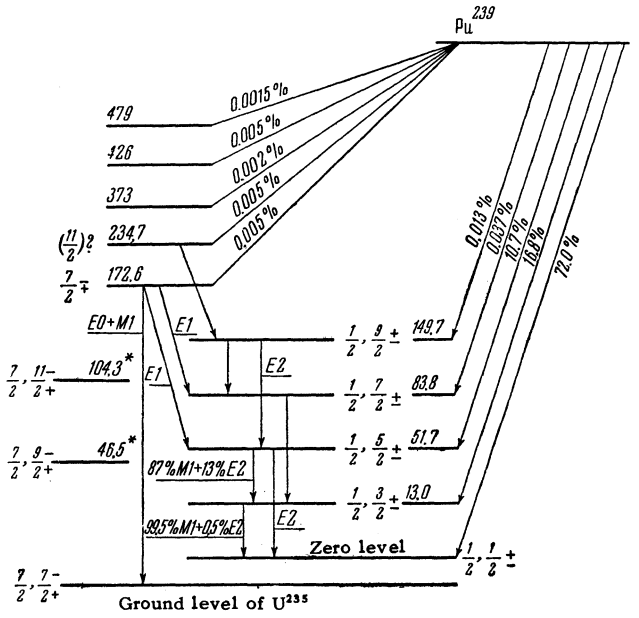

FIG. 4. Low-lying excited levels of U^{235} (from α decay data in Ref. 3). * – marks levels known from the Coulomb excitation of U^{235}. The energies of all levels are expressed in kev and computed from the "zero level."

with the theoretical intensities of E2 transitions from level II to levels I and 0. The theoretical ratio is

$$B\left(E2;\ \frac{5}{2} - \frac{3}{2}\right) \Big/ B\left(E2;\ \frac{5}{2} - \frac{1}{2}\right) = \frac{2}{7} = 0.29.$$

The experimental ratio is

$$\frac{B(E2;\ 5/2 - 3/2)}{B(E2;\ 5/2 - 1/2)} = \frac{N_{L_{III}}(38.7)}{N_{L_{III}}(51.7)}\,\frac{\alpha_{L_{III}}(51.7)}{\alpha_{L_{III}}(38.7)}\left(\frac{51.7}{38.7}\right)^3 = 0.36.$$

The good agreement of the theoretical and experimental intensity ratios is additional confirmation of the rotational nature of levels 0, I, and II. Transitions from level V to levels of the rotational band cannot be compared with the foregoing theory because these transitions involve violation of the rules of K forbiddenness, making the formulas inapplicable.

We note, finally, that the investigation of the α spectrum of Pu^{239} points to the existence of a series of higher excited levels of U^{235}, although electromagnetic transitions between these levels have thus far escaped detection.

Figure 4 summarizes the present data on the lower excited levels of U^{235}.

In conclusion we gratefully acknowledge assistance with the measurements by L. N. Kondrat'ev, I. I. Agapkin, and G. Chernov. We also wish to thank L. A. Sliv for values of the L-conversion coefficients which he supplied to us before publication.

[1] F. Asaro and I. Perlman, Phys. Rev. **88**, 828 (1952).
[2] Gol' din, Tret' iakov and G. I. Novikova, Reports of Conference of the U.S.S.R. Academy of Sciences on the Peaceful Uses of Atomic Energy, Div. of Phys. and Math. Sciences, p. 266, 1955.
[3] Novikova, Kondrat' ev, Sobolev, and Gol' din, J. Exptl. Theoret. Phys. (U.S.S.R.) **32**, 1018 (1957); Soviet Phys. JETP **5**, 832 (1957).
[4] D. Dunlavey and G. T. Seaborg, Phys. Rev. **87**, 165 (1952).
[5] G. Albouy and I. Teillac, Compt. rend. **232**, 326 (1951).
[6] D. West and J. K. Dawson, Proc. Phys. Soc. (London) **A64**, 586 (1951).
[7] Freedman, Wagner, and Engelkemeir, Phys. Rev. **88**, 1155 (1952).
[8] K. N. Shliagin, J. Exptl. Theoret. Phys. (U.S.S.R.) **30**, 817 (1956), Soviet Phys. JETP **3**, 663 (1956).
[9] Tret' iakov, Gol' din and Grishuk, Приборы и техника эксперимента (Instruments and Meas. Engg.) **6**, 22 (1957).
[10] K. L. Van der Sluis and J. R. McNally, J. Opt. Soc. Am. **45**, 65 (1955).
[11] Bohr, Froman and Mottelson, Kgl. Danske Videnskab. Selskab, Mat.—fys. Medd., **29**, 10 (1955).
[12] F. Asaro and I. Perlman, U.C.R.L. 3774 (1957).
[13] Alaga, Alder, Bohr and Mottelson, Kgl. Danske Videnskab. Selskab, Mat.—fys. Medd., **29**, 9 (1955).

Translated by I. Emin
166

SOVIET PHYSICS JETP VOLUME 34 (7), NUMBER 4 OCTOBER, 1958

STRUCTURE OF SUPERCONDUCTORS: XII
INVESTIGATION OF BISMUTH — RUBIDIUM ALLOYS

N. N. ZHURAVLEV, T. A. MINGAZIN, and G. S. ZHDANOV

Moscow State University

Submitted to JETP editor November 15, 1957

J. Exptl. Theoret. Phys. (U.S.S.R.) **34**, 820-826 (April, 1958)

A provisional melting diagram is constructed for the Bi — Rb system on the basis of thermal, microscopic, and x-ray data. Four compounds have been established for the Bi — Rb system: Bi_2Rb, $BiRb_3$, and two others which presumably are Bi_2Rb_3 and $BiRb_2$. The superconducting compound Bi_2Rb crystallizes in the cubic system with a lattice constant a = 9.609 A and has a structure of the Cu_2Mg type.

ACCORDING to data obtained by Alekseevskii,[1] heterogeneous alloys of bismuth with rubidium, having a large excess of bismuth, go over into the superconducting state at a temperature of 4.25°K. In spite of the non-uniformity of the alloys, the curves showing the superconducting transition displayed relatively little scatter, thus indicating the existence of a superconducting bismuth — rubidium compound. One of the purposes of the present investigation was to ascertain the composition and the atomic-crystalline structure of this compound.

The study of the bismuth — rubidium system involved the solution of a series of experimental problems. The principal difficulties were conditioned by the large chemical activity of metallic rubidium and by the markedly different physiochemical natures of bismuth and rubidium. All the investigations were carried out with a small quantity of rubidium (~3 gm), so that we had to develop micromethods for preparing the alloys and for analyzing them physiochemically. A certain quantity of metallic rubidium was obtained by one of the authors from the vacuum reduction of rubidium — iodide by calcium.[2]

PREPARATION OF THE ALLOYS

The large chemical activity of metallic rubidium required the creation of conditions which would prevent the oxidation of the metal during the proc-