

SOVIET PHYSICS

JETP

A translation of Zhurnal Éksperimental'noi i Teoreticheskoi Fiziki.

Vol. 11, No. 4, pp. 733-979

(Russ. orig. Vol. 38, No. 4, pp. 1017-1360, April, 1960)

Oct., 1960

LONG-LIVED LUTETIUM ISOTOPES

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Submitted to JETP editor August 7, 1959

J. Exptl. Theoret. Phys. (U.S.S.R.) **38**, 1019-1026 (April, 1960)

The conversion spectrum of the long-lived lutetium isotopes Lu^{173} and Lu^{174} was investigated. The relative conversion-line intensity in the soft region of the Lu^{173} spectrum (up to 250 keV) was measured. The per cent ratio of the M1 + E2 mixture for the 78.6- and 100.6-keV transitions is determined. The Lu^{174} spectrum contains conversion lines which can be ascribed to transitions connected with the isomer state of Lu^{174} ($E_\gamma = 44.7$ keV — M1 transition and $E_\gamma = 59.0$ keV — M3 transition). The half-life of the isomer state is ~ 90 days.

IN the study of the lutetium fraction separated from a tantalum target bombarded by fast (660 MeV) protons, a series of lines with a half-life considerably in excess of 32 days was observed in the conversion spectrum after the decay of the lutetium isotopes with relatively small half-lives ($T_{1/2} < 8$ days), and after the decay of the Yb^{169} present in the source ($T_{1/2} = 32$ days). The measurements were carried out on a double-focusing spectrometer with an approximate transmission of 0.2% ($R_0 = 24$ cm., half-width $\sim 0.3\%$). In many measurements the transmission was reduced to 0.1% in order to reduce the half-width of the lines. In the soft region (< 30 keV) the width of the lines was greater than the instrument width, owing to the slowing down of the electrons in the source material.

The measurements were carried out with two sources. Source 1 was obtained by bombarding the tantalum target in the cyclotron for approximately 4 hours and separating the lutetium fraction 10 to 12 hours after the bombardment. To produce source 2, the target was irradiated for approximately three months, and the lutetium fraction was separated a week after the end of the irradiation. Source 2 was employed previously by Dzhelepov et al.^{1,2}

The considerable number of the observed lines with long half-lives is due to the presence in the source of Lu^{173} , the spectrum of which has been relatively well studied.¹⁻³ Lu^{173} lines were observed in the spectra obtained with both sources. The relative line intensities obtained were in good agreement with those given by Bobrov et al.¹ with the exception of the Auger lines, whose intensities were obviously overestimated in their paper.

Figure 1 shows the strongest lines and the spectrum of the long-lived isotopes of lutetium, obtained with source 1. The figure shows the conversion lines without corrections for $H\rho$ and for absorption in the source.

Table I lists the energies and relative intensities of the lines (without corrections for the absorption in the source) in the Lu^{173} spectrum. The intensities given were obtained with source 2. The relative intensities obtained with the thinner source 1 are the same, within the limits of errors, for all lines with the exception of $K\gamma$ 78.6 keV. This shows that the correction for absorption in the source and in the counter films is small, starting with an electron energy ~ 30 keV.

When measurements with source 1 were made at a reduced aperture ratio, the conversion lines from the L subshells were sufficiently well separated.

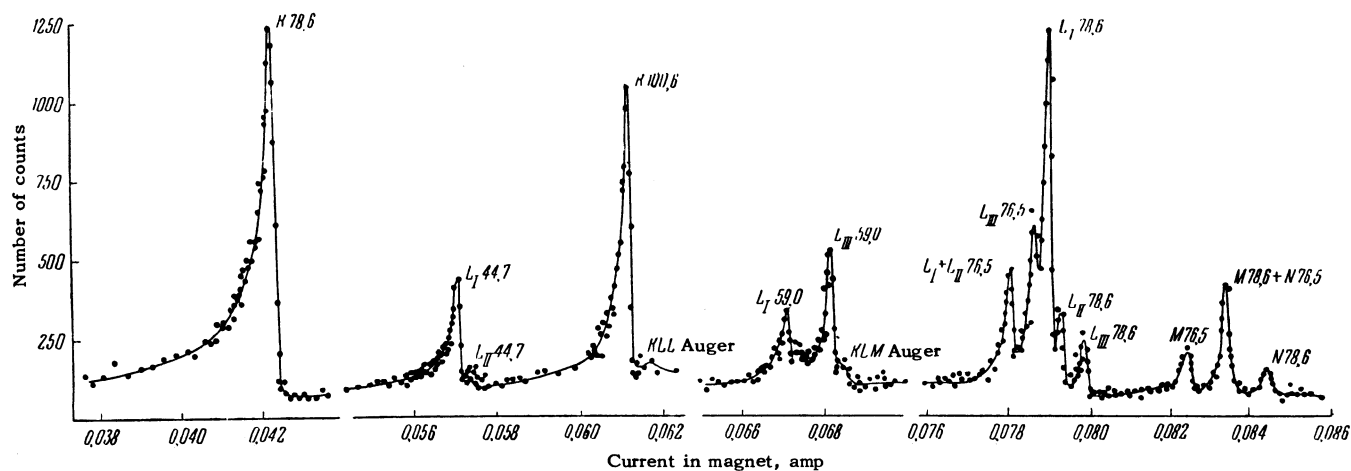


FIG. 1. Section of spectrum of long-lived lutetium isotopes.

TABLE I. Energy and relative intensity of the conversion lines in the Lu^{173} spectrum

No. of line	Identification of line	Line energy, kev	Relative intensity*	No. of line	Identification of line	Line energy, kev	Relative intensity*
1	$K\gamma$ 78.6	17.33	345 ± 10	11	$L_I + L_{II}\gamma$ 100.6	90.14	19.3 ± 0.5
2	$K\gamma$ 100.6	39.32	100	12	$L_{III}\gamma$ 100.6	91.70	
3	KLL -Auger	—	33 ± 1	13	$M\gamma$ 100.6	98.21	3.4
4	KLM -Auger	—	21.2 ± 0.7	14	$N\gamma$ 100.6	100.43	1.4
	KLN -Auger	—	4.0 ± 0.7	15	$K\gamma$ 171.3	110.0	1.4
5	KMM -Auger	—	4.0 ± 0.7	16	$K\gamma$ 179.3	118.0	2.1
6	$L_I\gamma$ 78.6	68.15	98 ± 3	17	$L\gamma$ 171.3	161.1	< 0.3
7	$L_{II}\gamma$ 78.6	68.64		18	$L_I + L_{II}\gamma$ 179.3	169.5	1.1
8	$L_{III}\gamma$ 78.6	69.69		19	$L_{III}\gamma$ 179.3	170.6	
9	$M\gamma$ 78.6	76.22	18.9 ± 0.6	20	$K\gamma$ 272.5	211.2	2.8 ± 0.1
10	$N\gamma$ 78.6	78.15	6.0 ± 0.2				

*The errors characterize the statistical dispersion and do not include the possible systematic errors.

rated for the different transitions. For the 78.6-keV γ transition the ratio obtained was $L_I : L_{II} : L_{III} = 1 : (0.24 \pm 0.005) : (0.164 \pm 0.003)$.

A comparison of the relations obtained with the theoretical values makes it possible to establish that this is a mixed transition, 95.5% M1 + 4.5% E2. For such radiation we have a theoretical ratio $(K/L)_{\text{theor}} = 5.1$. An experimental ratio $(K/L)_{\text{exp}} = 4.1$ was obtained with source 1 and $(K/L)_{\text{exp}} = 3.5$ was obtained with source 2. The deviation from the theoretical values is explained by absorption in the source material. A ratio $L_{III} : (L_I + L_{II}) = 0.075 \pm 0.002$ was obtained for the 100.6-keV γ transition. The transition is of the mixed type, 96% M1 + 4% E2. In this case $(K/L)_{\text{theor}} = 5.62$. The experimental ratio $(K/L)_{\text{exp}} = 5.4 \pm 0.2$ agrees, within the limits of errors, with the theoretical one. We see from a comparison of the theoretical and experimental values that the correction for absorption is small in this case.

For the 171.3-keV γ transition, an estimate of $K/L > 4.5$ was obtained. Such a K/L ratio is possessed by the transitions E1, M1, and M2. In the case of the 179.3-keV γ transition, the resultant ratios $L_{III} : (L_I + L_{II}) = 0.60$ and $K : L = 1.9$ indicate that this is an E2 transition. The theoretical ratios of the internal conversion coefficients for an E2 transition of the same energy are $L_{III} : (L_I + L_{II}) = 0.54$ and $K/L = 1.73$.

Gorodinskiĭ et al.³ measured the relative intensities of the γ rays and the spectrum of Lu^{173} . Using the relative γ -ray and the relative conversion K-line intensities obtained in the present work, one can determine the K-shell internal-conversion coefficients. In this case it is best to normalize the internal-conversion coefficients against the 78.6-keV $L\gamma$ line (the multipolarity of the transition has been well established, and the line energy is sufficiently high).

Table II lists the relative γ -ray intensities

TABLE II. Relative intensities of the transitions in the spectrum of Lu¹⁷³

Transition energy, keV	I _γ [%]	I _k , rel. units	I' _γ , rel. units	α _k from I _k and ref. 3	(α _k) _{theor} , from ref. 4			I _{tot}
					E1	M1	E2	
78.6	1	—	90.3					713
100.6	0.52	100	38.3	2.13 (0)	2.80 (-1)	2.81 (0)	9.89 (-1)	166
171.3	0.31	1.4		5.0 (-2)	7.0 (-2)	6.58 (-1)	2.70 (-1)	22
179.3	0.11	2.1	9.4	2.1 (-1)	6.65 (-2)	5.88 (-1)	2.23 (-1)	13
272.5	1.85	2.8		1.7 (-2)	2.14 (-2)	1.77 (-1)	7.1 (-2)	134
351	0.11							10
570	0.15							14
630	0.26							24

I_γ, obtained in reference 3, the relative intensities I_k of the conversion K lines, the γ-ray intensities I'_γ, obtained from the conversion data and from the Sliv and Band tables,⁴ and also the calculated K-shell internal-conversion coefficients α_k. The 100.6-keV Kγ line intensity is taken to be 100. A comparison of the calculated internal-conversion coefficients with the theoretical ones for the 100.6- and 179.3-keV γ transitions confirms the previously-established multipolarity and shows that 171.3- and 272.5-keV γ transitions are E1. Table II lists also the total relative intensities of the transitions, I_{tot}, calculated from our conversion data and the Sliv and Band tables.⁴ The intensities of the 351-, 570-, and 630-keV γ transitions were taken from the paper by Gorodinskii et al.³ and it is assumed that in these cases I_k << I_γ.

The level scheme of Lu¹⁷³ has been sufficiently well investigated.^{1-3,5} The total intensities and the decay scheme yield the "population" of the excited levels of Yb¹⁷³. Figure 2 shows the level scheme of Yb¹⁷³. In our case the number of captures in the excited levels was 760 (I_k (γ100.6) = 100 units). The number of vacancies freed after the conversion was ΣI_k = 605. The total number of vacancies was 1365. If we take Gray's⁶ Auger electron yield for Yb, α_k = 0.064 ± 0.010, and the experimental number of Auger K electrons to be 58 ± 2, a total number of 906 ± 165 K captures is obtained, which is less than a number of vacancies obtained from the transition intensities, even if it is assumed that there are no transitions to

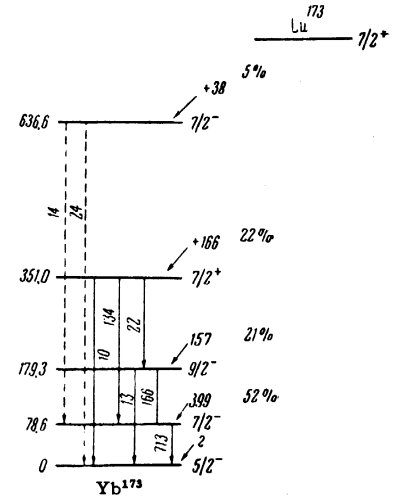


FIG. 2. Decay scheme of Lu¹⁷³.

the Yb¹⁷³ ground level. This may be due either to the comparatively large L capture, or to the inaccuracy in determination of the Auger-electron yield.

The spectrum of the long-lived isotopes produced from source 1 disclosed a series of lines not belonging to Lu¹⁷³, shown upon repeated measurements to have diverse half lives, all different from the half life of Lu¹⁷³. Table III lists energies of these lines. Two lines were observed in the region of the L lines of the 78.6-keV γ transition with energies 66.47 and 67.53 keV, identified as L_I + L_{II} and L_{III}. M and K lines of the same transition were also found. The K-L energy distance indicates that the conversion occurs in an atom with Z = 70. Table IV gives a comparison of the experimental and theoretical ratios of the

TABLE III. Energies of lines connected with the decays of Lu^{174*} and Lu¹⁷⁴

No. of line	Identification of line	Line energy, keV	No. of line	Identification of line	Line energy, keV	
1	Kγ	76.5	8	L _{II} γ	59.0	
2	L _I γ	44.7	9	L _{III} γ	59.0	49.75
3	L _{II} γ	44.7	10	Mγ	59.0	—
4	L _{III} γ	44.7	11	Nγ	59.0	58.56
5	Mγ	44.7	12	L _I + L _{II} γ	76.5	66.47
6	Nγ	44.7	13	L _{III} γ	76.5	67.53
7	L _I γ	59.0	14	Mγ	76.5	74.34

TABLE IV. Comparison of experimental and theoretical ratios of internal-conversion coefficients for the 76.5-keV γ transition of Lu^{174}

	Experiment	E1	E2	E3	M3
$K:L$	$0.23 \pm 0.06^*$	5.7	0.263	0.012	0.42
$L_I + L_{II} : L_{III}$	1.05 ± 0.03	3.34	1.01	1.06	0.745
$L:M$	3.6 ± 0.1	—	—	—	—

*The correction for absorption in the source is neglected.

coefficient of secondary conversion for this transition. The comparison identifies the transition to be E2. The transition energy was measured and found to be 76.5 keV. Chupp et al. observed in an investigation of Coulomb excitations⁷ a transition with energy 76.46 ± 0.01 keV, identified as a transition from the first excited state to the ground state of Yb^{174} . The transition energy and multipolarity obtained in the present work are in good agreement with the data of Chupp et al.⁷ The conversion lines of this transition were observed by Mihelich et al.⁸ and were also associated with the spectrum of Lu^{174} .

The 33.79-, 34.27-, 42.14- and 44.06-keV lines were identified as the L_I , L_{II} , M, and N lines of the 44.7-keV γ transition. The distance between the L_I and M lines is 8.35 keV. A comparison with the distance between L_I and M as obtained by x-ray data⁹ [$Z = 70$ (8.09 keV), $Z = 71$ (8.38 keV), $Z = 72$ (8.68 keV)] indicates that the conversion takes place in an atom with $Z = 71$. The ratios $L_I:L_{II}:L_{III} = 1:(0.156 \pm 0.004):(0.038 \pm 0.007)$ obtained indicate unequivocally that the transition is of the M1 type. The theoretical value for the M1 transition is $L_I:L_{II}:L_{III} = 1:0.093:0.013$. If there is an E2 admixture, it does not exceed 1% (approximately 0.75% for L_I/L_{II} and approximately 0.3% for L_{III}/L_I).

Lines 7, 8, 9, 10 and 11 in Table III have been identified as L, M, and N lines. It is difficult to determine the Z of the atom in which the conversion takes place directly from the L-M energy distance, since the M line falls on the KLM Auger lines. To obtain a better value of Z we measured the distance between the L_I and L_{III} lines. The source was isolated from the housing of the spectrometer and a certain voltage was applied to it. With a constant field in the spectrometer, we plotted the lines L_I and L_{III} several times while varying the voltage. The distance between the lines was read from the plot directly in volts and was found to be 1631 ± 12 v. The x-ray data for different Z yield the following values:

	Tm	Yb	Lu	Hf
Z	69	70	71	72
Distance between lines, v	1460	1544	1625	1716

Thus, the distance obtained between the L_I and L_{III} lines is in good agreement with the distance in the atom with $Z = 71$. The transition energy is $E_\gamma = 59.0$ keV. The experimentally obtained ratio $L_I:L_{II}:L_{III} = (0.49 \pm 0.02):(0.07 \pm 0.01):1$ is close to the tabulated one for the M3 transition ($L_I:L_{II}:L_{III} = 0.46:0.060:1$). For comparison, we give the theoretical value of the L-shell secondary conversion coefficient for transitions with a different multipolarity ($Z = 71$, $k = 0.10$, where k is the energy in m_0c^2 units):

E4	$L_I:L_{II}:L_{III} = 0.02:0.99:1$
E5	$L_I:L_{II}:L_{III} = 0.02:0.01:1$
M4	$L_I:L_{II}:L_{III} = 0.15:0.02:1$
M5	$L_I:L_{II}:L_{III} = 0.08:0.01:1$

To obtain more exact values of the half-lives of the intensities of the conversion lines, two series of measurements were made with source 2, and three series of measurements with source 1. Each series consisted of two measurements. The first series with source 2 was performed 2.5 to 3 years after the preparation of the source, while the second was performed 150 days after the first. In the case of source 1, the first series of measurements was made 300 days after separation, the second was made 100 days after the first, and the third 100 days after the second. In measurements with source 2, the half-lives of the lines in the Lu^{173} spectrum were found to be approximately 160 days. The intensity of the lines $L\gamma 44.7$ keV was found to be so low as to require special measurement. A rough estimate of the half-lives, based on the data of Bobrov et al.¹ and on our measurements, yields a half-life of about 100 days for $L\gamma 44.7$ keV. The $L\gamma 59.0$ keV lines cannot be separated from the stronger KMM Auger lines in these measurements. The lines $L\gamma 76.5$ keV and $L\gamma 78.6$ keV are very close to each other, and therefore the combined area of the two lines was measured. The

half-life of the entire group is approximately the same as for the Lu^{173} lines. The half-life of $M\gamma$ 76.5 keV does not differ greatly from that of the Lu^{173} lines, and is perhaps somewhat shorter. A rough estimate of the half-life of $M\gamma$ 76.5 keV, from the data of Bobrov et al.¹ and our measurements, yields a value of approximately 200 to 250 days.

The measurements with source 1 were performed three times. Since the measurement conditions (the aperture of the instrument) varied, it is impossible to calculate the half-life directly from these measurements, and it has become necessary to determine it from its ratio to $T_{1/2}$ for Lu^{173} , the value of which (107 days) was taken from reference 1 (our measurements with source 2 gave nearly the same value). Taking the $L\gamma$ 78.6-keV line of Lu^{173} as a reference, the half-lives of the remaining lines were found to be:

Line	$T_{1/2}$
$K\gamma$ 78.6 keV Lu^{173}	195 days
$L_I\gamma$ 44.7 keV	97 days
$K\gamma$ 100.6 keV	166 days
$L_{III}\gamma$ 59.0 keV	87 days
$L\gamma$ 76.5 keV	309 days
$M\gamma$ 76.5 keV	271 days
$M\gamma$ 78.6 keV $\pm N\gamma$ 76.5 keV	182 days

The lines of Lu^{173} have nearly the same half-life as the reference line $L\gamma$ 78.6 keV ($T_{1/2} = 170$ days). The half-life of $L_I\gamma$ 44.7 keV and $L_{III}\gamma$ 59.0 keV are relatively close to each other ($T_{1/2} \sim 85 - 95$ days). The half-life of the γ 76.5-keV lines amounts to 250 - 300 days and is clearly much greater than that of the reference line. The 44.7- and 59.0-keV γ transitions belong to the same isotope. This is confirmed by the fact that they occur in an atom having the same value of Z , that their lines have nearly equal half-lives, and that they have practically the same intensity:

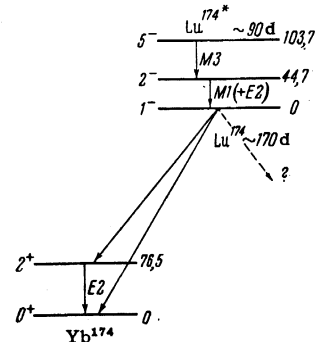
$$I_{\text{tot}} (\gamma 59.0 \text{ keV}) : I_{\text{tot}} (\gamma 44.7 \text{ keV}) = 0.94 \pm 0.03.$$

An attempt can be made to establish the particular lutetium isotope to which these transitions belong. Lutetium has a large number of known isotopes with different half-lives.¹⁰ The isotopes Lu^{169} , Lu^{170} , Lu^{171} , and Lu^{172} are obviously eliminated, for once the nucleus goes over from the isomer state to the ground state, the decay has a relatively small half-life, and the strongest transitions following the decay of these isotopes would have been observed. The level schemes of Lu^{173} , Lu^{175} , and Lu^{177} have been investigated and do not contain such transitions. This leaves only Lu^{174} and

Lu^{176} . The isotope Lu^{176} is found in the natural isotope mixture (2.6%) and decays with $T_{1/2} \approx 2.1 \times 10^{10}$ years¹⁰ to high-spin Hf^{176} levels. The spin of the ground state of Lu^{176} is 7.¹¹ Rotational levels with spins 8 and 9 have been obtained above the ground state by Coulomb excitation, with values $3\hbar^2/J \approx 66$ keV. In addition to the Lu^{176} in the ground state, an isomer state with $T_{1/2} = 3.7$ hours is known. No transition from the isomer state to the ground state has been observed. Lu^{176*} decays to the 0^+ and 2^+ levels of Hf^{176} . Its spin is small (it is assumed to be 1). The energy difference between the ground and excited states of Lu^{176} (based on the energies of the β^- decay) amounts to approximately 170 keV. It is thus seen that the low-lying excited levels of Lu^{176} have been sufficiently well investigated, but the levels corresponding to the transitions we observed did not appear. In addition, compounds obtained by bombardment with fast protons do not display, as a rule, sufficiently large amounts of neutron-excess isotopes.

It is most probable that the transitions belong to Lu^{174} . One of the possible versions of the decay scheme, which would agree with all the measured values of the half-lives, is shown in Fig. 3. To

FIG. 3. Proposed decay scheme of Lu^{174*} .



reconcile the half-lives of $M\gamma$ 76.5 keV in the measurements with the various sources, it is necessary to assume that when the lutetium fraction is first liberated it contains more Lu^{174*} than Lu^{174} . Then the amount of Lu^{174} in the ground state (γ -line intensity 76.5 keV) will first increase to a maximum (approximately 150 days from the instant of irradiation) and will then decrease. By the time measurements with source 1 were performed (300 to 500 days after irradiation), the apparent half-life of Lu^{174} should have been approximately 210 to 250 days. In the measurements with source 2 (approximately 1000 days after irradiation), the apparent half-life was close to the real one (obviously, 160 to 170 days). Wilkinson and Hicks¹² give a value $T_{1/2} = 165 \pm 5$ days for the half life of Lu^{174} .

The possible spins of the ground state of ${}_{71}\text{Lu}_{103}^{174}$ can be obtained by considering the neighboring odd nuclei. The neighboring nucleus, ${}_{70}\text{Yb}_{103}^{173}$, has spin and parity $5/2^-$ in the ground state, and $1/2^+$ in the first excited state ($E_\gamma = 351.0$ keV). The spin and parity of ${}_{71}\text{Lu}_{104}^{175}$ are $1/2^+$ in the ground state and $5/2^+$ in the first excited state ($E_\gamma = 343$ keV). Thus, the characteristics of the ground state of Lu^{174} can be either 6^- or 1^- . The characteristics of the excited states are 7^+ , 0^+ , 5^- , and 0^- . Of the two possible ground-state characteristics, it is apparently 1^- which is realized, since decay to the first rotational level 2^+ of Yb^{174} is observed. A search for a transition from the second excited level of Yb^{174} to the first level produced no results. The intensity of this transition amounts to not more than 2% of the total intensity of the γ transition, 76.5 keV.

If the ground state of Lu^{174} is 1^- , then for the resultant multipolarities M1 and M3 the excited states have characteristics 2^- and 5^- . The Lu^{174*} goes from the isomer state through an intermediate level (2^-) to the ground state. It remains unclear why the M3 transition is so greatly hindered and why there are no transitions to the rotational levels of Yb^{174} with spin 4^+ or 6^+ directly from the isomer state (the M3 transition is hindered by an approximate factor 2×10^7 compared with single-particle estimates).

The first excited level of Lu^{174} (2^-) can be interpreted as the first rotational level. It is possible to determine the moment of inertia of the odd-odd nucleons Lu^{174} . (It is more convenient to deal with the quantity $3\hbar^2/J$.) In this case $3\hbar^2/J = 67.2$ keV. The moment of inertia of Lu^{174} is greater than the moment of inertia of the neighboring nuclei with odd A. The energy of the second rotational level is 112 keV, i.e., the level should lie above that with the level with characteristics 5^- and does not appear in this case.

Recently Dillman et al.¹³ indicated that an 84-keV transition was observed in the spectrum of Lu^{174} . This transition is interpreted as one from the first excited state of Hf^{174} to the ground state. They did not notice any conversion lines corresponding to such a transition. If this transition exists, its intensity is considerably greater than that of the 76.5-keV γ transition, namely, $K_\gamma 76.5 \text{ keV} : K_\gamma 84 \text{ keV} \geq 5$.

Assuming the scheme shown in Fig. 3 and values of 59.0 and 76.5 keV for the relative intensities of the γ transitions, the intensity of the transitions to the ground and first-excited states of Yb^{174} can be estimated by considering that the majority of

transitions are to these levels (several transitions with greater energy were observed in reference 13, but their total intensity is less than the total intensity of the 76.5-keV γ transitions). The number of β^- decays to the levels of Hf^{175} , as indicated earlier,¹¹ amounts to approximately 20%. Under these conditions, an estimate shows that the number of transitions to the ground state of Yb^{174} is approximately 4 times the number of transitions to the first excited state: $\log ft \sim 8.4$ and ~ 9.0 respectively (at $\Delta E = 1.5$ MeV).

The ratio of the intensity of the K x-rays to the intensity of the 76.5-keV γ transition is found to be approximately 50, which agrees, within the limits of errors, with the results given in reference 13.

The authors express their sincere gratitude to Professor V. M. Kel'man for interest in the work and for valuable remarks made in the discussion of the results.

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