Zr⁹⁷ DECAY SCHEME

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The γ -ray spectrum and $\gamma\gamma$ -coincidence spectra for the $Zr^{97} \rightarrow Nb^{97} \rightarrow Mo^{97}$ decay are investigated with the aid of a coincidence scintillation spectrometer and single-crystal γ -spectrometer with a well-type NaI(Tl) crystal. It is found that excited states with energies 1.15, 1.35, 1.75, 1.84 and 2.1 MeV exist in the Nb⁹⁷ nucleus besides the familiar 0.745-MeV isomeric state. Possible characteristics of the levels are discussed.

1. INTRODUCTION

T was shown earlier in our laboratory^[1], and also by Nordhagen and Thomassen^[2], that in the decay of Zr^{97} there are excited, in addition to the well known 745-keV Nb⁹⁷ level (isomeric state with $T_{1/2} = 60 \text{ sec}$) also the higher levels, with intensity ≤ 10 per cent. Coincidences between the transitions excited were found to exist in the decay of Zr^{97} and not in the decay of the daughter nucleus Nb^{97[1]}. However, the energies and intensities of the gamma lines obtained in our laboratory^[1] and in ^[2], and also the decay schemes proposed there for Zr^{97} , are in poor agreement with each other. In order to resolve this contradiction we have undertaken a more detailed investigation of the decay of Zr^{97} .

2. SOURCE

The Zr^{97} source was obtained by bombarding zirconium, both natural and enriched by a factor of 16 with the Zr^{96} isotope, with neutrons in a reactor for about twenty hours. In view of the fact that the enriched source contained small Na²⁴ impurities, we employed an unenriched Zr^{97} source, in the form of metallic foil containing no noticeable Na²⁴ impurities for the investigation of the hard region of the single spectrum of Zr^{97} .

To investigate the coincidences we used an enriched source, so as to greatly reduce the contribution of soft gamma quanta from Zr^{95} , Hf^{175} , and Hf^{181} . The Na²⁴ impurity gave rise to an extensive background of true coincidences, seen on Figs. 2-4 (see below).

The Zr and Nb were not separated; thus, the measured gamma lines pertain to a mixture of the parent and daughter isotopes in equilibrium.

3. SINGLE SPECTRUM

The single gamma-ray spectrum of $Zr^{97} + Nb^{97}$ was investigated with the aid of a NaI(Tl) crystal measuring 80×80 mm, with a well 26 mm in diameter and 40 mm deep. The crystal was used in conjunction with an FÉU-24 photomultiplier, the pulses from which were fed to a 100-channel type AI-100 analyzer. The resolution at the 660-keV line was 13.5 per cent in the well and 12.8 per cent at a distance 8 cm from the surface of the crystal without collimation of the beam. Plexiglas 1-cm thick was used to absorb the beta radiation. The crystal was shielded on its sides with 5 cm of lead. A lead filter of 1 cm thickness was sometimes used to obtain a relative increase in the count in the hard region of the spectrum.

The gamma spectra obtained with and without a lead absorber were resolved using the standard gamma lines of Zn^{65} (1.12 MeV), Te^{208} (2.62 MeV), and Na²⁴ (2.76 MeV), taken under the same conditions as the measurements.

The efficiency of a well-type crystal for gamma rays from Co^{60} was determined by the method described by Schmidt-Ott^[3]. The value obtained for the efficiency, within the limits of the experimental error (~ 10 per cent) is in good agreement with the data of Gunnink and Stoner^[4], obtained for an analogous crystal. We therefore used the efficiency curve from that paper.

Figure 1 shows the gamma-ray spectrum of Zr^{97} (unenriched source) measured with a distance R = 8 cm from the source to the crystal and with the source placed in the well. The intensities of all lines in the hard region of the spectrum decrease with a half-life close to 17 hours, which makes it possible to ascribe them to the $Zr^{97} + Nb^{97}$ decay. In the soft region of the spectrum, gamma rays were also observed from the



FIG. 1. Gamma ray spectrum of Zr^{97} (unenriched source), measured with an 80×80 mm NaI(T1) crystal with well: O_{-} spectrum measured at distance R = 8 cm between the sources and the crystal; \bullet_{-} gamma spectrum obtained by placing the source in the well. The background has been subtracted. The shaded areas show the spectrum intervals separated in the control channel when measuring the coincidence spectra of Figs. 2a, b.

long-lived impurities Hf^{175} , Hf^{181} , and $Zr^{95} + Nb^{95}$. The maximum energy of the gamma rays due to the impurities is 750 keV ($Zr^{95} + Nb^{95}$). As can be seen from the figure, in the case of measurements in the well the relative intensities of some peaks (1150 and 1750 keV) decreased compared with the single spectrum, and at the same time the intensity of the 2100 keV peak increased appreciably. This result indicates that the 1150 and 1750 keV gamma rays are components of cascades whose summary energy is 2100 keV. This conclusion was confirmed by measurements of the $\gamma\gamma$ -coincidence spectra.

From the spectrum measured at R = 8 cm, we determined the relative intensities of the hard lines. They are given in the second column of the table.

The previously reported^[1] 2.58-MeV transition was not observed in the present research. The upper limit of its intensity amounts to less 0.02 per cent.

4. $\gamma\gamma$ COINCIDENCES

The spectra of the $\gamma\gamma$ coincidences were measured with the aid of a scintillation coincidence spectrometer. In the first series of measurements a transmitter with 80×80 mm crystal was connected in the control channel and a transmitter with a 40×40 mm crystal in the analyzing channel (FÉU-13, resolution ~ 10 per cent by the Cs¹³⁷ line). The source was clamped between the crystals. A lead filter 1 cm thick was placed in front of the thick crystal.

The shaded areas in Fig. 1 are the spectral regions separated in the control channel during measurements of the coincidence spectra (Fig. 2a, b).



FIG. 2. Spectra of $\gamma\gamma$ coincidences measured with the control channel set to the following intervals: a) 1050-1240 keV (region I on Fig. 1); b) 1650-1950 keV (region II on Fig. 1). An 80×80 mm crystal shielded with a 1 cm lead filter was placed in the control channel, and a 40×40 mm crystal in the analyzing channel. The dashed curves show the single spectrum in the analyzing channel. An enriched source was used. The random-coincidence background is practically negligible. Both spectra show extended tails due to the background of the coincidences of the 1.38- and 2.76-MeV Na²⁴ gamma rays. The shaded areas on spectrum a are the intervals separated during the measurements of the spectra in the Figs. 3 and 4.

Region I corresponds to energies from 1050 to 1240 keV; region II covers the range from 1650 to 1950 keV. The spectrum of coincidences with region I is shown in Fig. 2a, while that with region II is shown in Fig. 2b, from which it is seen that the line with energy 1150 keV gives coincidences with the lines 250, 340, 600, and 690 keV, while the 1.8-MeV line (1750-1840 keV) gives coincidences with the 250 and 340 keV lines.

Starting from the results obtained on coincidences with lines lying in the hard region of the spectrum, we made a second series of measurements, in which the windows of the control channel were set to the soft lines obtained from the coincidence spectrum. A small crystal replaced the large one in the control channel during this series of measurements. The large crystal was accordingly connected in the analyzing channel. The shaded areas on Fig. 2a show the windows of the control channel for the second series of measurements.

Figure 3 shows the spectra of the coincidences with regions I and II (with the 250- and 340-keV gamma rays). A single 1750-keV line is seen in the coincidence with the 340-keV gamma rays. In the coincidences with the 250-keV gamma rays there is observed a line at 1800 keV, broader than the single line. Its resolution has shown that it consists of two components, 1750 and 1840 keV. It was shown that the 1750-keV component is determined in this case by the contribution of coincidences with a Compton distribution from the 340-keV gamma transition. Thus, this measurement establishes the presence of 340-1750 keV and 250-1840 keV cascades. The intense 1150-keV line which appears in the spectra of Fig. 3 is in coincidence with both the 340 and the 250-keV transition.



FIG. 3. Coincidence spectra: •- with 340-keV gamma rays (region I on Fig. 2a), 0-with 250-keV gamma rays (region II on Fig. 2a). The measurement time in the second case was half that of the first. Broadening of the 1.80-MeV line in the spectrum of the coincidences with the 250-keV gamma rays, compared with the 1.75-MeV line in the coincidences with the 340-keV gamma rays, is noticeable. An enriched source was used. A 40×40 mm crystal was used in the control channel and an 80×80 mm crystal, shielded with 1 cm of lead, was used in the analyzing channel. Extending beyond the seventieth channel is the coincidence background due to Na²⁴. The ledge at ~ 1350 keV is partially due to the 1380-keV gamma rays of Na²⁴, to the Compton maximum from the 1750-keV gamma quanta, and apparently also to the 480-1350 keV cascade.





FIG. 4. Coincidence spectra measured with the control channel set for the intervals 440-520 (curve a), 550-630 (curve b), and 650-740 keV (curve c) (regions III, IV, and V of Fig. 2a, respectively). The remaining conditions are the same as in measurement of the spectra of Fig. 3. A relative increase is seen in the intensity of the ledge at 1350 keV when the control channel is set to the portion corresponding to region III. These measurements point to the existence of a 1350-480 keV cascade.

Figure 4 shows the spectrum of coincidences with regions III, IV, and V. Regions III, IV, and V correspond to the energy intervals from 450 to 530, from 550 to 620, and from 650 to 740 keV, respectively. It is seen from this figure that only a line with energy 1150 keV appears in the coincidences with the 690- and 600-keV gamma rays. When the window is shifted to the region $\sim 500 \text{ keV}$ a hump appears on the skirt of the 1150 keV line, corresponding to the 1350-keV energy. In the coincidence spectrum with the 1150 keV gamma rays, a 480-keV peak appears (Fig. 2a). Both results can be explained by the presence of a weak 480-1350 MeV cascade. (When setting the control channel to the 1150-keV peak we simultaneously register the pulses from the Compton distribution of the 1350-keV quanta). The observed gamma-ray cascades are shown in the fourth column of the table.

From the coincidence spectra shown in Figs. 2a and b we determined the relative intensities of the 250, 340, 600, and 690 keV lines, which owing to their low intensity did not manifest themselves in the single spectrum even when an enriched source was used. It was assumed in the intensity determination that the gamma transitions indicated

E _Υ , keV	Relative gamma-ray intensity, %		Gamma transitions	Nucleus in which
	Single spectrum	Coincidence spectrum	with which the given line is in cascade	the transition takes place
$\begin{array}{c} 250\pm10\\ 340\pm10\\ 480\\ 600\pm10\\ 665\\ 690\pm10\\ 745\\ 1000\pm20\\ 1150\pm15\\ \sim\!1350*\\ 1750\pm20\\ 1840\pm30\\ 2100\pm30\\ \end{array}$	$ \begin{array}{c} 100\\ 100\\ 2.6\pm0.6\\ 3.1\pm0.5\\ 2.7\pm0.5\\ 1.7\pm0.3\\ \leqslant 0.06 \end{array} $	$1.6\pm0.33.0\pm0.5weak1.7\pm0.31.3\pm0.31.3\pm0.30.4\pm0.2$	$(690), 1150, 1840 (600), 1150, 1750 (1350) (340), 1150 no coincidence with \gamma(250), 1150no coincidence with \gammano coincidence with \gamma250, 340, 600, 690(480)340250$	Nb ⁹⁷ Nb ⁹⁷

Data on the gamma ray spectrum of $Zr^{97} + Nb^{97}$

*Complicated line. Consists of at least two components in the 1.3-1.4 keV region. **Isomeric transition M4.

***According to $[^2]$, only approximately half of the 1.0-MeV gamma-ray intensity is connected with the transition in Mo⁹⁷. Our investigations, however, cannot place satisfactorily a transition with such energy between the Nb⁹⁷ levels.

in the table satisfy the Zr^{97} decay scheme shown in Fig. 5, and that the levels with energy 1840, 1750, and 1150 keV are populated essentially on account of the gamma transitions from the higher 2100 keV level.

5. DISCUSSION OF RESULTS. DECAY SCHEME OF Zr⁹⁷

The $Zr^{97} \rightarrow Nb^{97}$ decay scheme, which can be constructed on the basis of the results of the present measurements, is shown in Fig. 5.

The 2.1-MeV energy level can be immediately introduced on the basis of measurements of the spectrum for the summation of the cascade gamma quanta in the crystal with the well. The de-excitation of this level proceeds principally via the cas-



FIG. 5. Proposed scheme of the $Zr^{97} \rightarrow Nb^{97}$ decay.

cade transitions 0.34-1.75 and 0.25-1.85 MeV, and also 0.34-0.60-1.15 and 0.25-0.69-1.15 MeV. (We did not measure the triple coincidences, but double coincidences were observed between their components individually. In addition, the sum of the energies of these cascades is also equal to 2.1 MeV with good accuracy.) A weak direct transition from this level to the ground state with intensity < 1.5 per cent relative to the summary intensity of the cascades is also observed.

Delyagin et al^[1] observed in the beta spectrum of Zr^{97} a component with 0.48 MeV energy, giving coincidences with the hard (>1 MeV) gamma rays, and on this basis a level ~ 2.2 MeV was introduced in Nb⁹⁷. This level apparently corresponds to the 2.1 MeV level observed in the present work. The new value of its energy is somewhat in discord with the energy of the soft partial beta spectrum (the energy of the $Zr^{97} \rightarrow Nb^{97}$ decay is 2.65 MeV). However, the large thickness of the source used in [¹] (25 mg/cm²) could have lead to a considerable error in the determination of the energy of this beta-spectrum branch.

No other beta spectra giving coincidences with gamma rays were observed in ^[1] (except for the beta spectrum of 1.21 MeV of Nb⁹⁷, which gives coincidences with the 0.665 MeV gamma transition). On this basis we have assumed that the intermediate Nb⁹⁷ levels are not excited by beta decay (except for the 0.745 MeV level). Under this assumption we calculated the intensities of the cascade components listed in the table.

To accommodate the observed cascades, the levels 1.15, 1.75, and 1.84 MeV were introduced in the decay scheme of Fig. 5. Generally speak-

ing, the cascades can be placed in reverse order (spaced at 0.25, 0.34, and 0.95 MeV). However, such a placement of the levels seems to us less probable. First, the states $g_{9/2}$ (ground state) and $p_{1/2}$ (the 0.745-MeV level) are neighbors in accordance with the Mayer scheme and it would then be difficult to explain the nature of the intermediate levels. Second, if the spins of the 0.25 and 0.34 MeV levels are less than $\frac{9}{2}$ it would be more probable for the 0.745 MeV level to become de-excited via a cascade transition (as for example in Tc⁹⁹), and not by a direct M4 transition to the ground state, something not observed in fact. On the other hand, high spin values of the possible 0.25-0.34 MeV levels $(\geq \frac{9}{2})$ can likewise find no satisfactory explanation within the framework of the existing models.

The observed weak 0.48-1.35 MeV cascade can be located between the 1.84 MeV level and the ground state. The intensity of this cascade is much smaller than that of the 1.35-MeV gamma rays, determined from the single spectrum. It can be assumed that there is still one more 1.35-MeV transition, which produces no coincidences with the other gamma rays. It can be located between the 2.1-MeV level and the 0.745-MeV isomeric state.

The 1.00-MeV gamma transition observed in the single spectrum does not appear in any of the coincidence spectra. It was shown in [2] that this transition pertains at least partially to the Nb⁹⁷ decay.

On the basis of the shell-model predictions and in analogy with Zr^{95} , it has been previously assumed that the ground state of Zr^{97} is $d_{5/2}$. In this case the 1.9 MeV beta transition to the 0.745 MeV level $(\frac{1}{2})$ should be unique ($\Delta I = 2$, yes), which contradicts the relatively small value of log ft = 7.2 for this transition. However, the recent investigations of the stripping reactions on Zr nuclei^[5] have shown that the ground state of Zr^{97} is $\frac{1}{2}^+$. The 1.9-MeV β^- transition is in this case firstforbidden (non-unique) and the indicated value of log ft is well explained.

The summary intensity of the gamma transitions which de-excite the 2.1-MeV level amounts to ~ 0.07 quantum per decay, which agrees with the summary intensity of the cascade gamma transition and with the intensity of the soft partial beta spectrum determined in ^[1] by a different method. If we assume the energy of this gamma spectrum to be 0.55 MeV, as would follow from the energy of the $Zr^{97} \rightarrow Nb^{97}$ decay, we obtain a value log ft = 6.3, which corresponds to an allowed or firstforbidden spectrum, that is, $\Delta I = 0$ or 1. Consequently, the spin of the 2.1-MeV level should be $\frac{1}{2}$ or $\frac{3}{2}$.

The soft partial beta spectrum was separated in coincidences with gamma rays using a coincidence circuit with resolution time $\tau = 4 \times 10^{-8}$ sec^[1]. Consequently, the lifetime of the 2.1-MeV level is smaller than (or of the same order as) this quantity and the multipolarity of the 0.25 and 0.34 MeV transitions that de-excite it cannot be higher than E2. Thus, the 1.75- and 1.84-MeV levels should have spins $\leq \frac{7}{2}$. The absence of beta transitions to them, and of gamma transitions from them to the 0.74 MeV level, apparently limits the values of their spins from below to $\frac{5}{2}$.

The characteristics of the 1.15- and 1.35-MeV states cannot be determined for the time being. The absence of transitions from them to the 0.745-MeV level $(\frac{1}{2})$ indicates that their spin values are apparently larger than $\frac{3}{2}$.

Approximately 2 MeV above the $g_{9/2}$ and $p_{1/2}$ states there should be located the states $g_{7/2}$, $d_{5/2}$, $d_{3/2}$, and $s_{1/2}$ ^[6]. It is possible that they correspond to the levels 1.75 and 1.84 MeV ($d_{5/2}$ and $g_{7/2}$) and 2.1 MeV ($s_{1/2}$ or $d_{3/2}$). In addition, there can appear levels corresponding to the connection between the motion of an odd particle and the excitation of an even-even core (the core multiplet)^[7].

It is possible that the 1.15 and 1.35 MeV levels are members of the core multiplet, corresponding to $J_C = 2$ and $j_p = \frac{9}{2}$ (the notation is that of ^[7]). Then the 2⁺ level in Zr^{96} should lie approximately in this energy region. Unfortunately, there is no information whatever on the excited states of Zr^{96} in the literature. In Zr^{92} and Zr^{94} there are known first excited levels with energies 0.93 and 0.91 MeV respectively ^[8].

It is frequently possible to obtain interesting data by comparing the level schemes of neighboring odd isotopes which differ in the number of particles in the even group. However, for the Nb isotopes the material for the comparison is quite skimpy. Levels lying above the isomer state $\frac{1}{2}^{-}$ in Nb⁹¹ (1.31 and 1.64 MeV) and Nb⁹⁵ (0.724 and 0.757 MeV) are known^[8], but the characteristics of these levels are almost unknown.

In conclusion it must be noted that the present investigation has eliminated the contradictions between the results of our earlier work^[1] and the work of Nordhagen and Thomassen^[2]. The proposed decay scheme combines the principal features of the schemes proposed in these papers.

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