DECAY OF NEUTRON-DEFICIENT NEODYMIUM ISOTOPES. A NEW ISOTOPE Nd^{138}

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The spectra of conversion electrons, γ rays, and γ-γ coincidences for the 5.5-hour activity of the neutron-deficient Nd isotope fraction have been studied with a double-focusing $\pi\sqrt{2}$ β spectrometer and a scintillation γ spectrometer. It is shown that most γ transitions observed in this activity are related to the isotope Pr^{139}, excited in the decay of the isomer Nd^{139m}, excited in the decay of the isomer Nd^{139m}. A decay scheme is proposed for $Nd^{139m} \to Pr^{139}$. In the conversion electron spectrum we have also observed the EO transition line in Ce^{138}, excited in the decay $Nd^{138} \to Pr^{138} \to Ce^{138}$. The intensity of this line also fell off with a $\sim$5.5-hour period, which is experimental confirmation of the presence of a roughly 5-hour Nd^{138} activity in the Nd fraction.

1. INTRODUCTION

STOVER[1] in 1951 irradiated praseodymium and cerium oxides with 10-60 MeV protons and observed a number of new Nd and Pr isotopes, including Nd^{139} with a 5.5-hour half-life and Nd^{138} with a 22-minute half-life. Bonch-Osmolovskaya et al,[2] in a study of the positron spectra of neutron-deficient Nd isotopes obtained by irradiating a tantalum target with 660-MeV protons, found positrons whose intensity decayed with a 5.5-hour half-life. They established that the positron spectrum consists of three components with end-point energies of 3300 ± 100 keV, 1000 ± 100 keV, 460 ± 60 keV, and relative intensities 1 : 0.084 : 0.024, respectively. On the basis of the half-life, these positrons were related to a chain beginning with Nd^{139}. It was established that the positrons with the 1000 keV end-point energy arise in the decay of Pr^{139} ($T \approx 5$ hours).[1,2] However, since Bonch-Osmolovskaya et al[2] did not observe an increase in the intensity of this component with the decay of neodymium, most of the intensity of the 1000 keV component must be associated with the decay of the 5.5-hour Nd isotope. On the other hand there were difficulties in the assignment of all three components with the intensities listed to the decay of Nd^{139}. Dzhelepov et al[3] have suggested that Nd^{138} and Nd^{139} both have a decay period of about 5 hours: The Nd^{138} decay results in a short-lived isomeric state of Pr^{138} (with quantum numbers 1^+), and positrons with a 3300 keV end-point energy are emitted in the Pr^{138} decay; the positrons with end-point energies 1000 and 460 keV are associated with the decay of Pr^{139}.[3] These assumptions need experimental verification.

The conversion electron and γ-ray spectra of the 5.5-hour neodymium activity were studied by Kolesov et al,[4] who observed γ transitions with energies of 114, 228, 703, and 950 keV. Earlier, Gromov et al[5] studied the conversion electron spectrum of the neutron-deficient neodymium isotopes with a β spectrograph and determined that the previously known 5.5-hour neodymium activity belongs not to the ground state but to an isomeric state of Nd^{139} with an energy of 232.5 keV and quantum numbers $h_{11/2}$. This isomeric state decays mainly to levels of Pr^{139} and only partially (not more than 18%) by an isomeric transition to the ground state of Nd^{139}.

In the present work we have continued the studies of the Nd^{139m} decay. We have also obtained experimental proof of the existence of the isotope Nd^{138} with a half-life of about 5 hours.

2. EXPERIMENTAL CONDITIONS AND RESULTS

We obtained the neutron deficient neodymium isotopes by irradiation of erbium oxide or tantalum targets with 660-MeV protons. The irradiations were carried out in the Joint Institute for Nuclear Research synchrocyclotron for a period of about 2 hours. The neodymium fraction (all the neodymium isotopes formed in deep spallation reactions on Ta or Er) was separated by the chromatographic method within 4-5 hours after the end of the irradiation.

Separation of the isotopes was not carried out and therefore the neodymium fraction used in the investigations could contain the following known radioactive isotopes (see the tables in Dzhelepov et al [3]):

$$Nd^{141} \xrightarrow{2.5\text{ hour}} Pr^{141}$$
$$Nd^{140} \xrightarrow{3.3\text{ day}} Pr^{140} \xrightarrow{3.4\text{ min}} Ce^{140}$$
$$Nd^{139m} \xrightarrow{5.5\text{ hour}} Pr^{139} \xrightarrow{4.5\text{ hour}} Ce^{139} \xrightarrow{140\text{ day}} La^{139}$$
$$Nd^{138} \xrightarrow{5\text{hour?}22\text{ min.?}} Pr^{138} \xrightarrow{?} Ce^{138}$$
$$Pr^{138} \xrightarrow{2\text{ hour}} Ce^{138}$$

The following considerations show that the presence of other isotopes does not greatly hinder the investigation of the ~5 hour neodymium activity. Nd^{141} decays mainly (more than 96% [3]) to the ground state of Pr^{141}; i.e., the γ radiation from this isotope is of low intensity. Since our measurements were started 6-10 hours after the end of the irradiation, i.e., after 3-5 half-lives of Nd^{141}, we can assume that its contribution must be small. The decay of Pr^{139} also occurs almost completely (99% [3]) to the ground state of the daughter nucleus. The presence of several weak γ transitions which arise in this decay did not actually interfere with our investigations. Nd^{140} has a relatively longer half-life (3.3 days), and therefore the radiation arising in the decay of Nd^{140} and Pr^{140} can be easily distinguished from the 5 hour activity.

Conversion electron spectrum of the neodymium fraction. This spectrum was measured with a double focusing β spectrometer. The measurements were commenced 5-6 hours after the end of the irradiation. The sources for the β spectrometer were prepared by electrolytic deposition on a copper wire 0.2 mm in diameter. The resolution varied from 0.15-0.3% in the different experiments, depending on the quality of the source. In these measurements we set as our goal to improve the identification of the lines observed by us earlier [5] and assigned to the decay of Nd^{139m}, and also to extend the studies of the conversion electron spectrum to higher energies. Our earlier studies of the conversion electron spectrum [5] were made with a β spectrograph in the energy region up to 900 keV. In the present work we have studied the spectral regions from 50-290 keV and from 650-1500 keV.

The results of the measurements are presented in Table I. The energies of the transitions are determined on the assumption that they originate in a Pr nucleus. Exceptions are the 232.5 keV transition, which we previously established [5] as an isomeric transition in Nd^{139}, and the 1478 keV transition, which apparently originates in Ce^{138} (see below).

In the fifth column are listed the measured half-lives of the conversion lines. In the cases where the half-life is not shown, estimates of the half-life allow us to exclude all possibilities other

Table I. Results of conversion electron spectrum measurements

Transition number	Electron energy, keV	Identification	Relative intensity	$T_{1/2}$, hours	Multipolarity
1	72,00	*K*-114,0	5100±200 *		*M*1
2	197.2	*M*-114,0			
3	112.5	*M*-114.0			
4	90.7±0.2	*K*-132.7	~50		
5	158.1±0.3	*K*-200.1	65±20		
6	168,4±0.3	*K*-210.4	25±14		
7	174.6±0.3	*K*-216.6	25±14		
8	188.9±0.4	*K*-232.5	1000	5.5±0.5	*M*4
9	225.8	*L*-232.5	340±40		
10	230,9	*M*-232.5	80±20		
11	285.0±0,5	*K*-327.0	131±15	5.5±0.5	
12	665.8±1	*K*-707.8	152±8	5.5±0.7	*M*2
13	695.4±1	*K*-737.4	~50±7		*M*2
14	701.0	*L*-707.8	17		
15	731±1	*K*-773	7±2		*E*1
16	779±2	*K*-821	6±2		
17	780±2	*K*-822	~6		
18	784±2	*K*-826	6±2		
19	786±2	*K*-828	10±3		
20	941±3	*K*-983	8±2		
21	1437±3	*K*-1478	45±7	5.1±0.6	
22	1472±3	*L*-1478	6±1		

*The intensity of the K-114 line was taken from conversion electron spectrum measurements made in a β-spectrograph.[5] In the measurements made in the present work, its intensity was diminished, since the electron counter window transmitted electrons beginning with an energy of 30 keV.

Table II. Energies and relative intensities of γ lines ($T_{1/2} \approx 5$ hours)

Transition number	Photopeak energy, keV	γ-line intensity in singles spectrum	γ-line intensity in coincidence with 114-keV line	Transition number	γ-line intensity in coincidence with 114-keV line	γ-line intensity in singles spectrum	γ-line intensity in coincidence with 114-keV line
1	115±10	0.8±0.2		11	1030±20	0,30±0.05	0.2±0.1
2	330±30	$\sim$0.5		12	1100±20	0.30±0.1	0,1±0.05
3	510 annihilation	14	3.5±0.5	13	1240±30	0.2±0.05	
				14	1340±40	0.2±0.1	0.15±0.08
				15	1480±30	0.1±0.03	$<$0.01
4	630±30	0,3		16	1580±30	0.08±0.02	0.04±0.01
5	710±10	1	1	17	1720±30	0.04±0.01	0,03±0.01
6	740±10	0,42±0.3	0.3±0.2	18	1850±30	0.03±0.01	
7	770±10	0,68±0.2	0.4±0,3	19	2050±20	0,10±0.03	0.15±0.05
8	820±10	0.7±0,1		20	2170±20	0.05±0,02	
9	900±20	0,25±0.05	0.1±0.05	21	2350±30	0,014±0,005	
10	980±10	0.7±0.1	0.5±0.2	22	2500±50	0.015±0.005	

than assignment of these lines to a $\approx$ 5-hour activity.

Spectrum of γ rays. The γ-ray spectrum was studied with a scintillation γ-spectrometer. Two NaI(Tl) crystals were used: one 40 × 40 mm with an FÉU-11 photomultiplier, and the second 80 × 80 mm with an FÉU-24 photomultiplier. The second crystal had a well 30 mm in diameter and 40 mm deep. The first crystal had a resolution of 10% for the Cs^{137} 662 keV line, and the second—12.5%. An AI-100 multichannel analyzer was used in the measurements.

The γ-ray spectrum measurements on the neodymium fraction were started 10-12 hours after the end of the irradiation. In the low energy region of the spectrum, photopeaks are distinctly observed which correspond to γ-rays of 114 and 510 keV (the annihilation peak). Comparison of the Nd spectrum with the spectrum of Na^{22} measured under the same conditions permits us to detect γ rays with an energy of about 330 keV. The remaining γ rays, which were observed in the study of the conversion spectrum in the region between $h\nu$ = 114 keV and $h\nu$ = 510 keV (Table I), are masked by the Compton spectrum and by the back-scattered γ-ray peak.

Figure 1 shows the γ spectrum of the 5-hour neodymium isotopes in the region from 500-2900 keV (lower curve). The upper curve was obtained using the crystal with the well. It is evident that in this case the γ spectrum is distorted by adding in the crystal. The data obtained on the energies and relative intensities of the γ rays are listed in Table II.

Multipolarities of the transitions. We previously established[5] that the 114-keV transition has a multipolarity M1. Using the value of α_k given in the tables of Sliv and Band for this transition, we related the intensities of the γ rays and the conversion electrons and computed the values of α_k

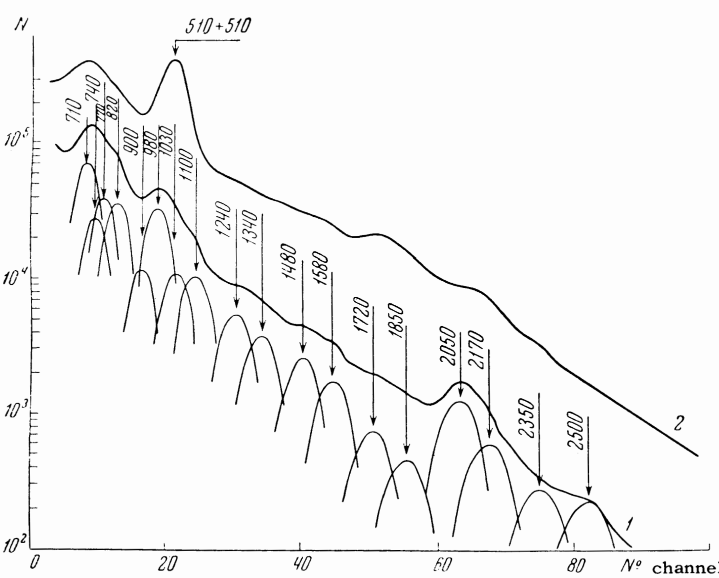

FIG. 1. Curve 1 – γ-ray spectrum of the 5-hour Nd isotopes in the energy region 500-2900 keV. Crystal is 80 × 80 mm, source to crystal distance is 30 cm. Curve 2 – the same spectrum measured with the source located in the well.

Table III. Multipolarity of γ transitions

$E\gamma$, keV	α_k, experimental	α_k, theoretical								Multi-polarity
		M1	M2	M3	M4	E1	E2	E3	E4	
707,8	$(1{,}8\pm0{,}7)\cdot10^{-2}$	$6{,}3\cdot10^{-3}$	$1{,}74\cdot10^{-2}$	$3{,}98\cdot10^{-2}$	$8.7\cdot10^{-2}$	$1.54\cdot10^{-3}$	$3{,}98\cdot10^{-3}$	$8{,}9\cdot10^{-3}$	$2{,}1\cdot10^{-2}$	*M*2
737	$(1{,}36\pm0{,}5)\cdot10^{-2}$	$5{,}6\cdot10^{-3}$	$1{,}55\cdot10^{-2}$	$3.46\cdot10^{-2}$	$7{,}6\cdot10^{-2}$	$1.38\cdot10^{-3}$	$3{,}6\cdot10^{-3}$	$8.1\cdot10^{-3}$	$1.7\cdot10^{-2}$	*M*2*
773	$(1{,}2\pm1{,}0)\cdot10^{-3}$	$4.46\cdot10^{-3}$	$1{,}2\cdot10^{-2}$	$2{,}7\cdot10^{-2}$	$5{,}6\cdot10^{-2}$	$1{,}17\cdot10^{-3}$	$2{,}98\cdot10^{-3}$	$6{,}6\cdot10^{-3}$	$1.58\cdot10^{-2}$	*E*1
1478	$>4{,}7\cdot10^{-2}$	$1{,}07\cdot10^{-3}$	$2{,}4\cdot10^{-3}$	$4.46\cdot10^{-3}$	$7.58\cdot10^{-3}$	$3{,}63\cdot10^{-4}$	$8.13\cdot10^{-4}$	$1{,}5\cdot10^{-3}$	$2{,}63\cdot10^{-3}$	—

*Most probable multipolarity.

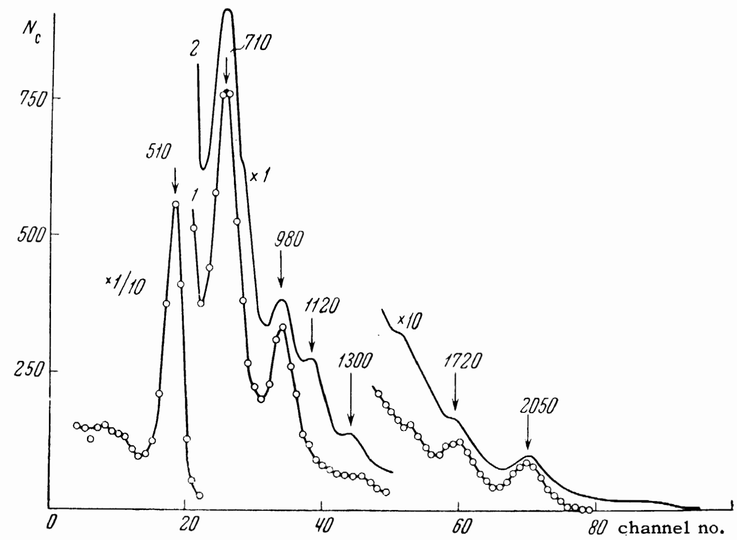

FIG. 2. Curve 1 – spectrum of coincidences with 114 keV γ-rays. Curve 2 – singles spectrum (without points). Crystal is 80×80 mm, shielded by 5 mm Pb and 0.5 mm Cd. The spectral region 80–140 keV was recorded in the controlling channel (crystal 12×30 mm). The angle between counters is 180°.

for the γ transitions at 707.8, 737, and 773 keV. Table III compares the values thus obtained with the theoretical values and also lists our conclusions on the multipolarity of the transitions. The value of α_k obtained for the 1480-keV transition is considerably larger than any of the theoretical values given in Table III. This leads to the conclusion that the observed conversion lines are associated either with a high order multipole γ transition ($L > 6$) or with an E0 transition.

γ-γ coincidences. These events were studied in the coincidence spectrometer described by Sorokin.[6] The same counters were used (crystals and photomultipliers) as in the study of the γ spectrum. The resolving time of the coincidence circuit was $2\tau = 2 \times 10^{-7}$ sec. The measurements were made with angles of 180° and 90° between the counters. Appropriate lead-cadmium shielding was used to absorb photons scattered from one crystal to the other.

The coincidence spectra, particularly in the high energy region, turned out to be extremely complex and frequently could not be interpreted uniquely. The following cascades were established with the greatest reliability: 114—708 keV, 114—980 keV, 114—1720 keV, and 114—2050 keV. Figure 2 shows the spectrum of coincidences with 114-keV γ rays, and the last column of Table II lists the relative γ-ray intensities obtained from decomposition of the spectrum. The cascade 980—740 keV also appeared distinctly (see Fig. 3). In the spectrum of coincidences with hard γ rays (> 1 MeV) the 114-keV line is the most intense. In addition we observed a peak at $\sim$ 330 keV. This peak is also visible in the coincidences with 980-keV γ rays (Fig. 3). However, its intensity de-

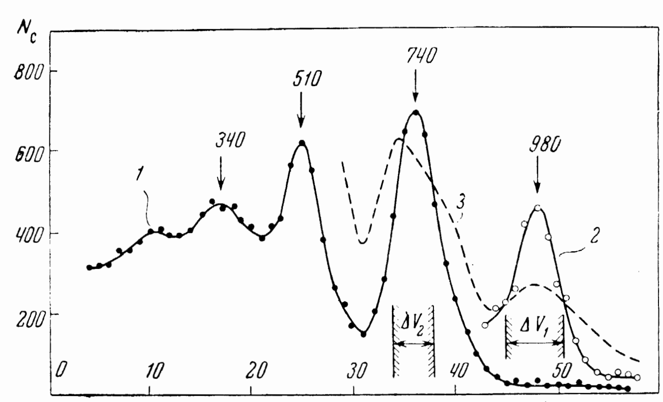

FIG. 3. Curve 1 – spectrum of coincidences with 980-keV γ rays; ΔV_1 - location of controlling channel window. 2 – spectrum of coincidences with 740-keV γ rays (region ΔV_2). Curve 3 – section of the singles spectrum. The crystals and the filters are the same as in Fig. 2.

cayed with a half-life somewhat longer than 5.5 hours, evidently due to some long-lived admixture. There are also indications that the 5-hour component of this peak is produced not by one γ transition (for example, 327 keV, observed in the conversion spectrum) but by two.

In the high energy region no intense coincidences were observed except the 980-740 keV cascade. There are indications of coincidences of 700-800 keV γ rays of the same energy and with γ rays in the 1000—1300 keV region. No coincidences were observed of the 114 keV γ line with γ rays > 2050 keV or with the 980-708 cascade. It should be noted that the intense 708 keV γ transition line in the singles spectrum is extremely weak, if it appears at all, in the coincidence spectrum, except in coincidences with 114-keV γ rays.

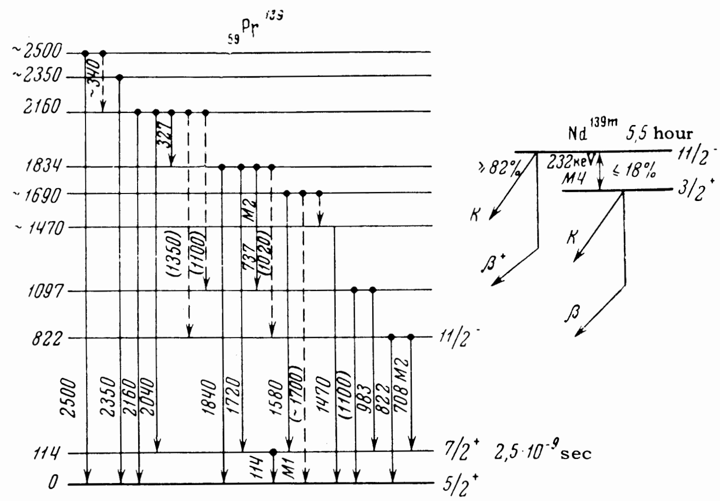

Fig. 4. Decay scheme of Nd^{139m}.

3. DISCUSSION OF RESULTS

Decay scheme of Nd^{139m}. The most intense γ transition in the spectrum of the 5-hour neodymium fraction is the 114-keV transition, which occurs in cascade with most of the other γ transitions of this fraction. As we established earlier,[5] this transition, and consequently also the other γ transitions related to it, originates in a Pr nucleus. This nucleus can be either Pr^{138} or Pr^{139}, since the isotopes Nd^{138} and Nd^{139} both have the ≈ 5-hour half-life. As we will see below, the structure of the level scheme which can be constructed on the basis of our data is more characteristic of an odd-even nucleus than of an odd-odd nucleus. Therefore we consider it most probable that these transitions occur between the levels of Pr^{139}, excited in the decay of Nd^{139m}.

On the basis of our studies of the γ-γ coincidence spectra and from the energy differences of the γ transitions in Pr^{139}, we can establish the following levels with the greatest confidence (see Fig. 4):

114 keV. A considerable number of transitions terminate in this level. An approximate balance of the intensities shows that this level is almost never excited by direct β decay. It is depopulated by an M1 transition to the ground state.

822 keV. This level is depopulated by the 708—114 keV cascade and by a direct transition to the ground state. An intensity balance shows that direct β decay must play an important role in the excitation of this level.

1097 keV. This level is depopulated by the 983-114 keV cascade and, possibly, by a direct transition to the ground state. It is populated by the 737 keV transition and directly by β decay.

1834 keV. This level is depopulated by the 737-983-114 keV cascade (possibly also by the 737-1097 keV cascade; however, as the coincidence spectrum of Fig. 3 shows, this cascade is appreciably weaker than the first), by the 1720-114 keV cascade, and by a direct transition to the ground state. It is apparently filled by the 327 keV transition and possibly by β decay.

2160 keV. This level is depopulated by a direct ground state transition, by the 2040-114 keV cascade, and also by cascades proceeding via the 1834 keV level and possibly the 1097 and 822 keV levels.

2500 keV. This level is introduced on the basis of the direct transition of this energy and of the possible cascade $\sim 340 - 2040$. On the basis of our data we can also introduce levels with energies of ~ 1470, ~ 1690, and ~ 2350 keV. In reality the level scheme in the energy region of 1000 keV and above is considerably more complex.

From the properties of the Pr^{139} β decay we can conclude, as has been done by Dzhelepov et al,[3] that the quantum numbers of the Pr^{139} ground state are $5/2^+$. The ground state of Pr^{141}, in which the filling of the N-82 neutron shell is completed, has these same quantum numbers. The 114-keV state can then evidently be assigned the quantum numbers $7/2^+$, since the $5/2^+$ and $7/2^+$ levels, which correspond to the $d_{5/2}$ single-particle states, are closest to each other in odd nuclei with Z from 51 to 63, and particularly in the isotopes Pr^{141} and Pr^{143} (in Pr^{143}, where the neutrons begin to fill a new shell, the levels overlap and the ground state has the quantum numbers $7/2^+$ [7]). Sorokin[8] has measured the lifetime of the 114 level and has shown that the 114-keV transition has a retardation factor, in comparison with the single-particle value, of $f \approx 300$, which also is characteristic of l-forbidden M1-transitions between $g_{7/2}$ and $d_{5/2}$ states.

The multipolarity of the 708-keV M2 transition determines the quantum numbers of the 822-keV level as $3/2^-$ or $11/2^-$. Since, according to our data, direct β-decay from the $11/2^-$ isomeric state of Nd^{139} must play an important role in excitation of the 822-keV level, the latter choice ($11/2^-$) is to be preferred. In this case the 822-keV transition must have a multipolarity E3 and its intensity should be small in comparison with the cascade. Apparently the main contribution to the 830 keV peak in the scintillation spectrum (see Table II) is due to the 826 and 828 keV transitions observed in the conversion electron spectrum.

The new isotope Nd^{138}. As was stated above, the determination of α_K for the 1480-keV transition leads to the conclusion that this transition must have either a very high multipole order ($L > 6$) or must be a $0 \rightarrow 0$ transition. It is clear that the first possibility can be excluded: transitions of such a high multipolarity are not observed and their appearance, evidently, is unlikely. The only reasonable assumption is that the observed conversion lines are associated with a $0 \rightarrow 0$ transition, and the conversion lines corresponding to γ rays of about 1470 keV are weak and were not observed by us.

Observation of the E0 transition conversion lines, whose intensity decays with a $\approx$ 5-hour half-life, is experimental proof of the existence of the assumed isotope Nd^{138} with this half-life. It is obvious that this transition must be located in the $Nd^{138} \rightarrow Pr^{138} \rightarrow Ce^{138}$ decay scheme as shown in Fig. 5. An additional argument in favor of the existence of the $\approx$ 5-hour Nd^{138} is the following experimental fact. The relative intensities of 710-keV γ rays and the annihilation radiation are considerably different in the singles spectrum of the 5-hour fraction and in the spectrum of coincidences with 114-keV γ rays. The relative intensity of the annihilation peak is several times greater in the singles spectrum than in the coincidence spectrum (see Table II). Since the decay of Nd^{139m} ($11/2^-$) goes almost completely to excited states of Pr^{139} in view of the strongly forbidden β decay to the ground state ($5/2^+$), this means that there is an intense 5-hour β^+ component which is not associated with the decay of Nd^{139m}.

Thus, the decay of Nd^{138} (the 0^+ level) evidently occurs to a Pr^{138} state with low spin

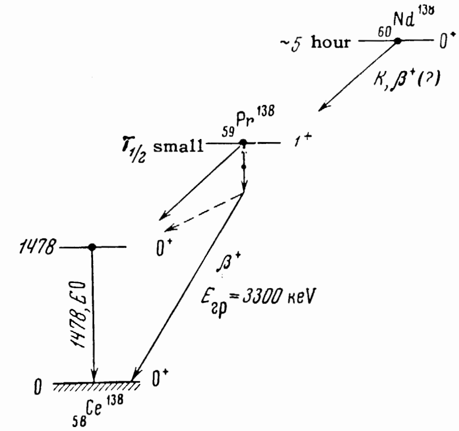

FIG. 5. Decay scheme of the chain $Nd^{138} \rightarrow Pr^{138} \rightarrow Ce^{138}$.

(probably 1^+ [3]); from this level, in turn β-decay occurs to the ground state of Ce^{138} (the component with $E_{max} = 3300$ keV) and to the 0^+ level at 1478 keV. The Pr^{138} state with $\approx$ 2-hour half-life is not formed in the decay of Nd^{138}, since it has a high spin of 7^- or 8^-, as has been shown by Dzhelepov et al. [3]

It remains unclear to which isotope belongs the 22-minute activity observed by Stover. It is possible that it is associated with an Nd isotope lighter than Nd^{138}.

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