Decay properties of neutron-rich niobium isotopes

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The decay of neutron-rich niobium isotopes in the mass region $A \simeq 100$ has been investigated by γ-ray singles and γ-γ coincidence measurements. The isotopes were produced by thermal-neutron induced fission of ^{235}U, ^{239}Pu, and ^{249}Cf, and niobium was separated from fission-product mixtures with an automated chemical procedure. Isomerism was found in the even-mass niobium isotopes with the following half-lives: 1.5 and 3.1 sec for ^{100}Nb, 1.3 and 4.3 sec for ^{102}Nb, and 0.8 and 4.8 sec for ^{104}Nb. Half-lives and γ rays were assigned to the 15.0-sec ^{99}Nb, 7.1-sec ^{101}Nb, 1.5-sec ^{103}Nb, and ~1-sec ^{106}Nb. Revised half-life values are given for the 7.1-sec ^{100}Zr and 2.9-sec ^{102}Zr.

[RADIOACTIVITY ^{99,100,101,102,103,104,106}Nb, ^{100,102}Zr; measured $T_{1/2}$, E_γ, γ-γ-coin. ^{100,102,104}Mo deduced levels. Fission ^{235}U, ^{239}Pu, ^{249}Cf (n_{th}, f); automated chemical separation procedure, Ge(Li) detectors.]

I. INTRODUCTION

In a previous paper,[1] short-lived neutron-rich isotopes of zirconium and their niobium daughters were investigated by rapidly separating zirconium from fission products and successively recording γ-ray spectra of the purified zirconium fraction. In these spectra γ rays of niobium nuclides were characterized by a growth-and-decay curve. The half-lives of their zirconium parents in the mass chains 100 and 102 were deduced from such growth-and-decay curves, since no γ-ray peaks could be assigned unambiguously to the decay of ^{100}Zr and ^{102}Zr. For mass 100 the curve showed a growth component with a half-life of $\approx$1.3 sec and a decay component with a half-life of 7.1 sec[1]; for the mass 102 a growth component with a half-life of $\approx$1 sec and a decay component with a half-life of 2.9 sec were obtained.[1] Since half-lives for β^- decay increase along a β^--decay chain for most of the isobaric mass chains, it seemed reasonable to attribute the shorter half-lives, $\approx$1.3 and $\approx$1 sec, to the decay of the zirconium and the longer half-lives, 7.1 and 2.9 sec, to the decay of the niobium nuclides of mass numbers 100 and 102, respectively. In addition, our half-life value of 7.1 sec for ^{100}Nb was in good agreement with the previously reported values of 6.6 sec found by Eidens, Roeckl, and Armbruster[2] and of 7.1 sec reported by Wilhelmy.[3] Eidens *et al.*[2] used a helium-filled mass separator of moderate mass resolution for the on-line separation of fission products and detected them by β-γ-coincidence techniques. Wilhelmy,[3] who studied the decay of products from the spontaneous fission of ^{252}Cf, determined the nuclear charge via the energy of characteristic x rays emitted in coincidence with γ-ray transitions and the mass via the kinetic energy of the fission fragments.

A different half-life, 2.4 sec, was found for ^{100}Nb by Hübenthal, Monnand, and Moussa[4] and Herzog[5] by means of the ^{100}Mo$(n,p)^{100}$Nb reaction. An activity with this half-life did not grow in the zirconium fraction.[1] This situation could be accounted for with the explanation that the 7.1-sec ^{100}Nb is a low-spin isomer fed in the β^- decay of the even-even nucleus ^{100}Zr with ground state $J^\pi = 0^+$, whereas the 2.4-sec ^{100}Nb is a high-spin isomer not populated in β^- decay of ^{100}Zr. However, both isomers should have been formed in the case of the ^{100}Mo(n,p) reaction.[4,5]

To clear up this discrepancy we investigated the short-lived niobium nuclides formed in fission. A rapid chemical separation technique for niobium was developed.[6] The results obtained show that the assignments[1-5] of half-lives to ^{100}Zr, ^{100}Nb, ^{102}Zr, and ^{102}Nb need revision. In addition ^{103}Nb, ^{104}Nb, and ^{106}Nb were discovered in this work, and further information on the decay of ^{101}Nb was obtained.

II. EXPERIMENTAL

Neutron-rich isotopes of niobium were produced by thermal-neutron induced fission of ^{235}U, ^{239}Pu, and ^{249}Cf. The irradiations were performed in a

rabbit system of the Mainz Triga reactor operated in the pulse mode. After irradiation, niobium was quickly isolated by an automated chemical procedure using an experimental apparatus similar to those used for the chemical separation of zirconium and technetium.[1]

The separation method is based on the sorption of niobium on fiberglass filters from strong nitric acid.[6] The apparatus for this separation procedure and the time schedule is shown in Fig. 1. A solution of ^{235}U or ^{239}Pu in 2 ml of 0.1 N HNO_3, containing SO_2 for the reduction of the halogens, tartaric acid for complexing antimony, and holdback carriers for halogens, alkalines, alkaline earths, lanthanides, zirconium, and technetium, is sealed in a polystyrene capsule and irradiated with a pulse of neutrons. After the pulse the capsule is transported within 0.2 sec into the separation apparatus where it is smashed by impact. Bromine, iodine, and silver fission products are removed from the solution in an exchange reaction by filtration through two layers of preformed silver chloride. Then the layers are washed with 1 ml of 0.1 N HNO_3. The filtrates are collected in 6 ml of concentrated HNO_3 and the resulting 10 N HNO_3 solution is filtered through two fiberglass filters which adsorb niobium. After washing with 10 ml of 10 N HNO_3 the filters containing the niobium are transferred by pressure to Ge(Li) detectors placed in a shielded position. Counting of the sample is started 2.2 sec after the end of irradiation. The decontamination factors are $\geq 10^2$ for zirconium and $\geq 10^4$ for other elements, e.g., halogens, alkalines, alkaline earths, lanthanides, yttrium, molybdenum, technetium, and ruthenium.

In order to increase the decontamination factor for zirconium to $\geq 10^4$, a further purification step was added in a series of experiments. The adsorbed niobium was eluted with 6 ml of a mixture of 0.5 N hydrofluoric acid and 15 N nitric acid, the fluorocomplexes of niobium were destroyed with 3 ml of boric acid solution (50 mg/ml), and the niobium was resorbed on fiberglass filters. The niobium samples were available for counting 4.4 sec after the end of irradiation.

In the experiments performed with ^{249}Cf a solid target deposited on a cylindrical target holder was used. The fission products recoiling out of the target were collected[7] in an easily soluble catcher material such as NH_4NO_3. After irradiation the catcher assembly was quickly transferred into the head of the separation apparatus. The catcher was then dissolved in 0.1 N HNO_3 containing SO_2 and tartaric acid. The subsequent separation procedure and the time schedule are the same as described for ^{235}U and ^{239}Pu.

To record γ-ray spectra a 32-cm^3 Ge(Li) detector [1.75 keV full width at half maximum

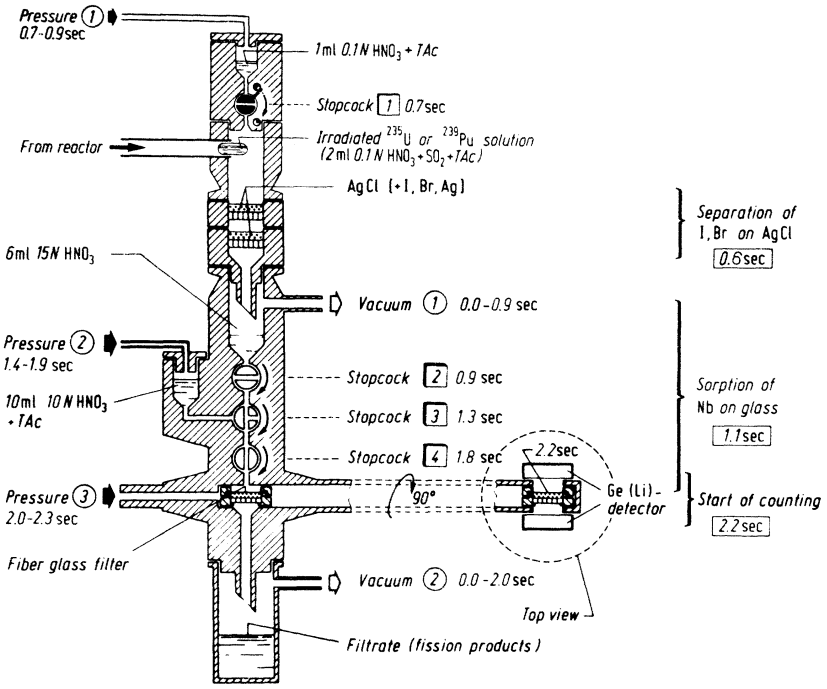

FIG. 1. Apparatus and time schedule for the rapid separation of niobium from fission products. At the right hand side the mean time for each separation step is indicated.

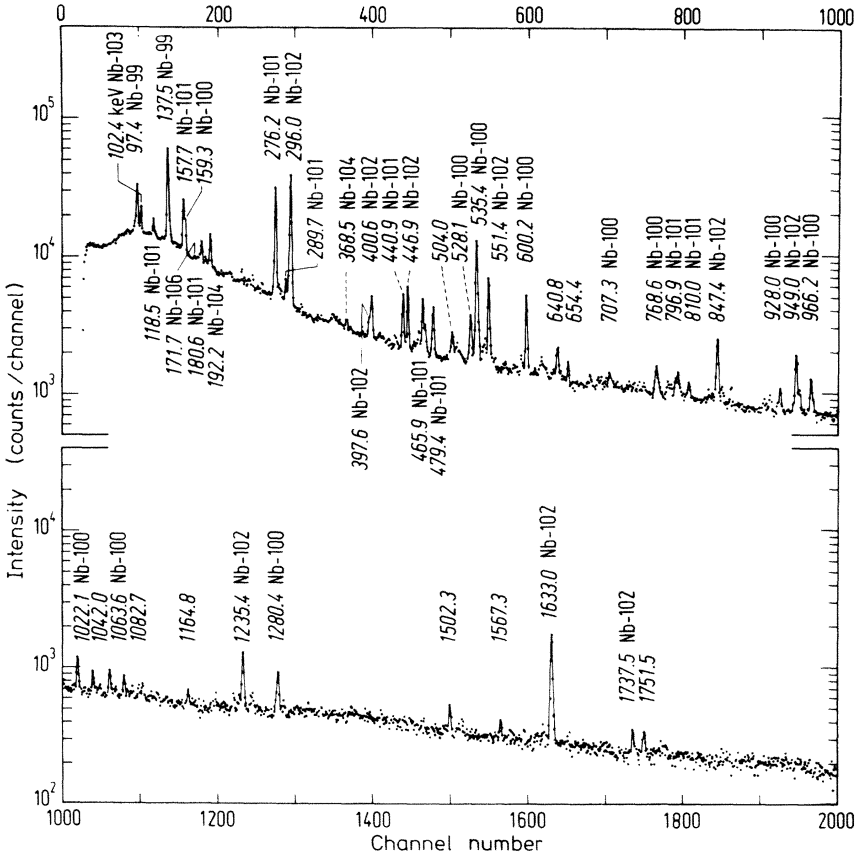

FIG. 2. Spectrum of γ rays of short-lived niobium isotopes from thermal-neutron induced fission of ^{239}Pu.

(FWHM) at 1333 keV] was used in combination with a 16 384-channel analyzer and a magnetic tape recorder. The γ-ray energies were measured to ±0.2 keV for strong and ±0.5 keV for weak γ rays. In some experiments, a 0.14-cm^3 x-ray detector (0.2 keV FWHM at 5.9 keV) was used simultaneously to check the purity of the samples via characteristic x rays. In addition, γ-γ-coincidence measurements were performed with two 35-cm^3 Ge(Li) detectors (2.1 keV FWHM at 1333 keV) in the two-dimensional mode with a 4096×4096 channel matrix; the coincidence data were recorded event by event on magnetic tape. The resolving time of the coincidence setup was about 25 nsec. In most cases 50 to 100 runs were summed up to improve the statistical quality of the spectra.

III. RESULTS

A. γ-ray spectra

A γ-ray spectrum of short-lived niobium isotopes covering the energy region up to 2.0 MeV is shown in Fig. 2. The niobium isotopes were produced by thermal-neutron induced fission of ^{239}Pu and separated from fission products 1.6 sec after the end of irradiation. The samples were counted for 11.5 sec beginning 4.4 sec after the

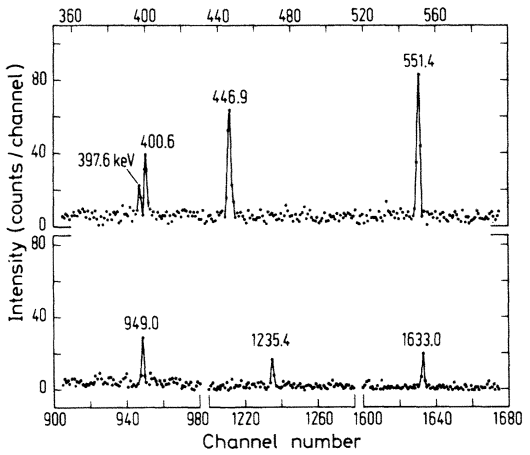

FIG. 3. Spectrum of γ rays in coincidence with the 296-keV peak of ^{102}Nb.

end of irradiation. Contributions of longer-lived isotopes were subtracted using the spectra recorded during the following 11.5 sec. As a specimen spectrum for the γ-γ-coincidence measurements, Fig. 3 shows the γ lines which are in coincidence with the 296.0-keV γ ray of ^{102}Nb.

B. ^{99}Nb, ^{101}Nb, and ^{103}Nb

The 97.4- and 137.5-keV γ rays, already observed in zirconium samples,[1] belong to the decay of the 15.0-sec ^{99}Nb. In the zirconium work,[1] it was possible to assign the 276.2-keV peak to 7.1-sec ^{101}Nb. Additional γ rays at 118.5, 157.7, 180.6, 289.7, 440.9, 465.9, 479.4, 796.9, and 810.0 keV which decay with the same half-life are found in the niobium fraction and are attributed to ^{101}Nb. The γ-ray peak at 102.4 keV decays with a half-life of 1.5 ± 0.2 sec. It does not fit into the decay schemes of the even-mass niobium isotopes discussed below; therefore, it is assigned to the decay of ^{103}Nb. The mass assignment is in agreement with data of Clark, Glendenin, and Talbert,[8] who studied delayed γ rays in the spontaneous fission of ^{252}Cf.

C. ^{100}Zr-^{100}Nb

Figure 4(a) shows the decay curve of the strongest γ-ray transition of ^{100}Nb at 535.4 keV. This energy is known from nuclear reaction data[9-11] as that of the first excited state in ^{100}Mo with $J^\pi = 2^+$. Two isomers of ^{100}Nb with 1.5- and 3.1-sec half-lives can be unfolded from this curve, but there is no evidence for a 7.1-sec component. In the zirconium fraction,[1] however, the growth-and-decay curve of the 535.4-keV peak [Fig. 4(c)] showed, after passing the maximum, a decay with a 7.1-sec half-life. Hence, the 7.1-sec activity is assigned to ^{100}Zr which decays into a shorter-lived niobium daughter. From the growth of the 535-keV γ line in the zirconium fraction, a half-life of 1.4 sec for the niobium daughter nuclide is deduced, in agreement with the shorter-lived component appearing in Fig. 4(a). As the $J^\pi = 0^+$ ground state of the even-even nucleus ^{100}Zr

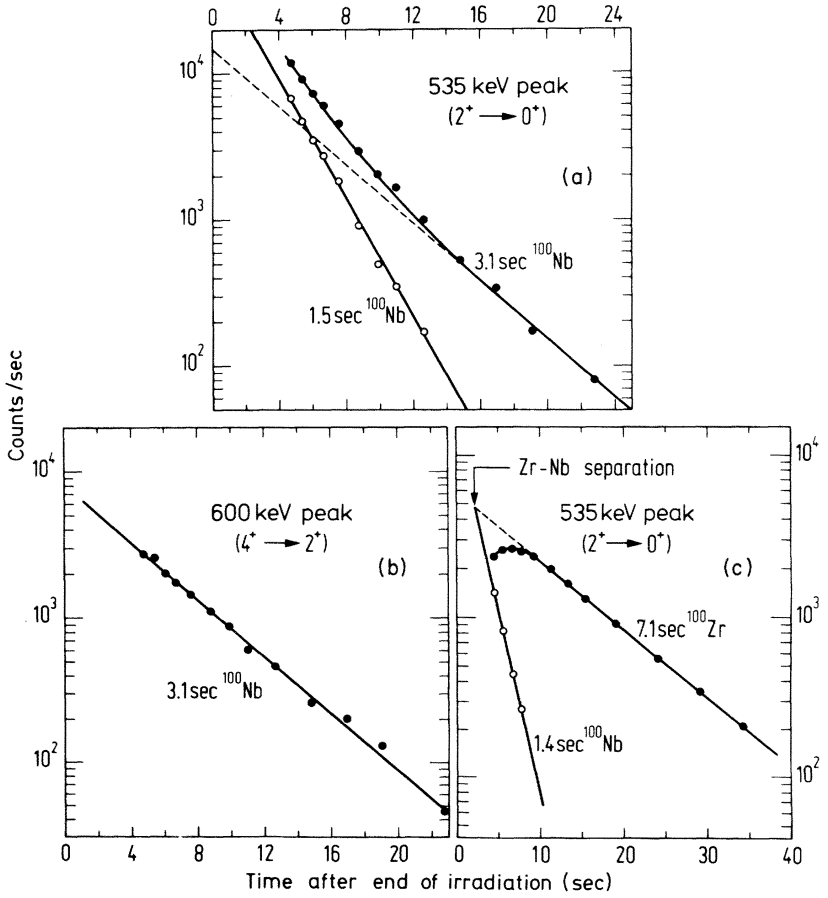

FIG. 4. Decay and growth-and-decay curves of the (a), (c) 535- and (b) 600-keV γ-ray peaks of ^{100}Nb in niobium (a), (b) and zirconium (c) samples (Ref. 1).

TABLE I. Half-lives and γ-ray energies of even-mass zirconium and niobium isotopes.

Isotope	Half-life (sec)	γ-ray energy [a] (keV)
^{100}Zr	7.1 ± 0.4 [b]	
^{100}Nb	1.5 ± 0.2	159.3, 528.1, 535.4, 600.2,[c] 707.3,[d] 768.6,
	3.1 ± 0.3	928.0, 966.2,[c] 1022.1, 1063.6, 1280.4 [c]
^{102}Zr	2.9 ± 0.2 [b]	
^{102}Nb	1.3 ± 0.2	296.0, 397.6,[d] 400.6, 446.9,[c] 551.4, 847.4,
	4.3 ± 0.4	949.0, 1235.4,[c] 1633.0,[c] 1737.5,[c] 2184.4 [c]
^{104}Nb	0.8 ± 0.2	192.2, 368.5 [d]
	4.8 ± 0.4	
^{106}Nb	~1	171.7

[a] The γ-ray energies were measured to ±0.2 keV for strong and ±0.5 keV for weak γ rays.

[b] Indirect half-life measurement by following the growth and decay of the niobium daughter in zirconium samples.

[c] These γ-ray peaks decay with the half-life of the longer-lived high-spin isomer; in all the other cases complex decay curves with two components have been observed or the peaks are too weak for half-life determinations (see d below).

[d] These γ-ray peaks are too weak for half-life determinations. The mass assignment is based on γ-γ-coincidence measurements.

should decay preferably to low-spin states in ^{100}Nb, the 1.5-sec half-life is assigned to a low-spin isomer of ^{100}Nb. Hence, the 3.1-sec component in Fig. 4(a) should represent the decay of a high-spin state in ^{100}Nb. This is confirmed by the feeding of the 4^+ level in ^{100}Mo at 1135.6 keV[9,11] in the β^- decay of the 3.1-sec isomer, as follows from Fig. 4(b), showing the decay of the 600.2-keV γ rays which connect[9] the 4^+ level with the 2^+ level at 535.4 keV in ^{100}Mo. Additional γ-ray peaks attributed to ^{100}Nb are given in Table I. Most of them decay with two half-lives of about 1.5 and 3 sec. The decay of the three peaks at 600.2, 966.2, and 1280.4 keV can only be fitted with a one-component decay curve with 3.1-sec half-life.

Obviously the 2.4-sec activity observed in Refs. 4 and 5 for the ^{100}Mo(n,p) reaction consists of a mixture of the two isomers of ^{100}Nb with 1.5- and 3.1-sec half-lives. The absence of a 7.1-sec component in this reaction[5] is consistent with our assignment of this half-life to the decay of ^{100}Zr.

A partial decay scheme for ^{100}Nb, based on γ-singles and γ-γ-coincidence measurements, is shown in Fig. 5. As it is not possible to decide which of the two isomers belongs to the ground state of ^{100}Nb, they are arbitrarily shown at the same energy. The assignments of spin and parity to the low-lying levels of ^{100}Mo are based on nuclear reaction data.[9-11] The level scheme is in essential agreement with the ^{100}Nb decay scheme of Herzog[5]; an additional level at 2085.6 keV has been deduced in the present work.

D. ^{102}Zr-^{102}Nb

Figure 6(a) shows that half-lives of 1.3 and 4.3 sec result from the decay of the 296.0-keV γ line in the niobium γ-ray spectra. This line has already been assigned to the $2^+ \rightarrow 0^+$ transition of ^{102}Mo in γ-x-ray-coincidence measurements[12,13] and in (t,p) reaction studies.[14] In the growth-and-decay curve of this peak in zirconium samples,[1] a growth with an ≈1-sec and then a decay with 2.9-sec half-life was observed [Fig. 6(c)]. Using the same arguments for ^{102}Nb as for ^{100}Nb the 1.3-sec activity is assigned to a low-spin isomer of ^{102}Nb, and the 2.9-sec activity to the decay of ^{102}Zr. The energy of the $4^+ \rightarrow 2^+$ transition[13] in ^{102}Mo is 446.9 keV; a γ-ray peak of this energy was observed to decay with a half-life of 4.3 sec [Fig. 6(b)]. This half-life is assigned to a high-spin isomer of ^{102}Nb. Additional γ lines of ^{102}Nb are given in Table I. The peaks at 446.9, 1235.4, 1633.0, 1737.5, and 2184.4 keV decay only with the half-life of the longer-lived isomer, whereas the other γ-ray peaks decay with two half-lives of about 1.3 and 4.3 sec.

The γ lines in coincidence with the 296.0-keV γ peak are shown in Fig. 3. A partial decay scheme of ^{102}Nb is given in Fig. 5. Again it is not possible to decide which of the two isomers is the ground state in ^{102}Nb. The spin and parity assignments for the low-lying levels of ^{102}Mo are based on the data of Cheifetz *et al.*[13] and Casten *et al.*[14]

E. ^{104}Nb, ^{106}Nb

Isomers were also found in ^{104}Nb. The decay curve of the 192.2-keV peak in the niobium fraction (Fig. 7) indicates half-lives of 0.8 and 4.8 sec. Cheifetz *et al.*[12,13] have assigned the 192-keV peak to the $2^+ \rightarrow 0^+$ transition in ^{104}Mo. Although we observe the 368.5-keV transition from the 4^+ to the 2^+ level[12,13] in ^{104}Mo in the niobium fraction, the intensity is too weak for an unambig-

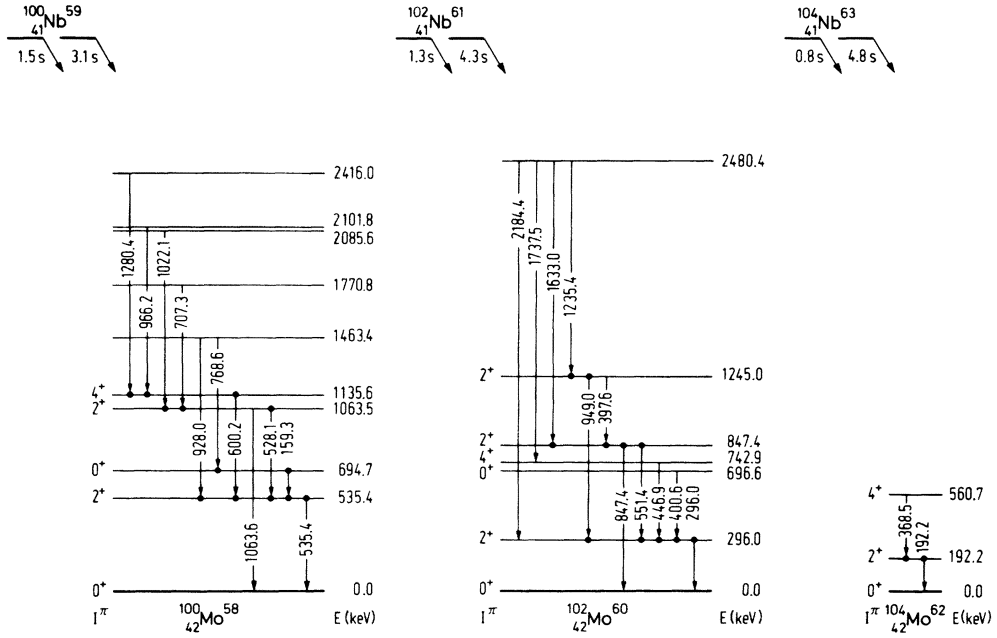

FIG. 5. Partial decay schemes of ^{100}Nb, ^{102}Nb, and ^{104}Nb. Transitions confirmed by coincidence relationships are marked by dots.

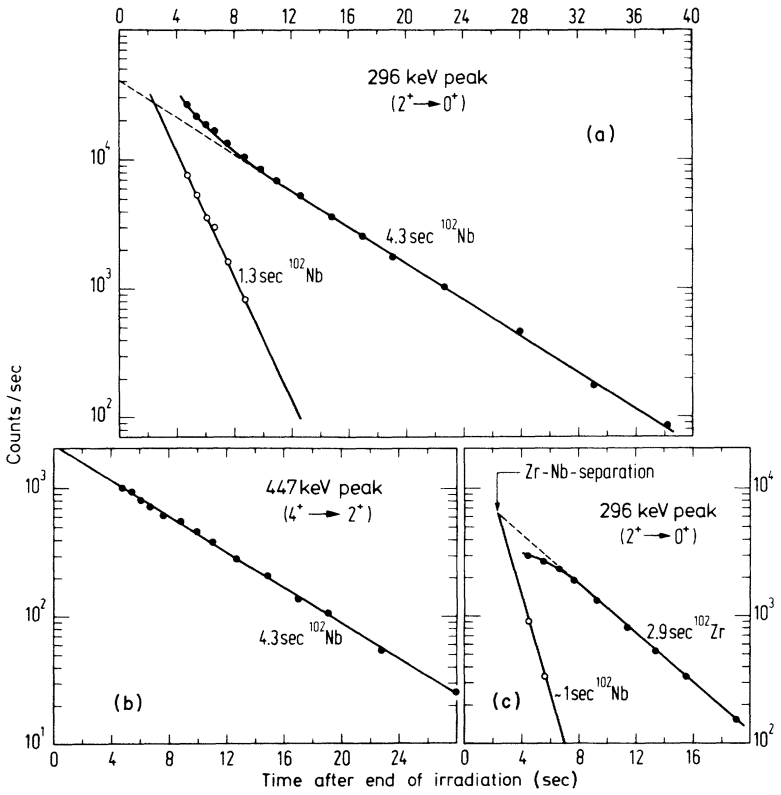

FIG. 6. Decay and growth-and-decay curves of the (a), (c) 296- and (b) 447-keV γ-ray peaks of ^{102}Nb in niobium (a), (b) and zirconium (c) samples (Ref. 1).

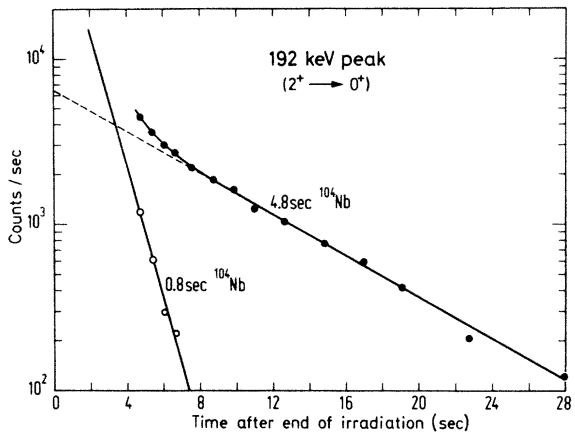

FIG. 7. Decay curve of the 192-keV γ-ray peak of ^{104}Nb in niobium samples.

uous half-life determination. Since it was not possible to study the decay of ^{104}Zr into ^{104}Nb, assignments of the two components to low- or high-spin isomers are not made.

Cheifetz *et al.*[12,13] assigned an energy of 171.7 keV to the $2^+ \rightarrow 0^+$ transition in ^{106}Mo. With ^{239}Pu or ^{249}Cf as fissionable material, our niobium samples showed a weak γ-ray peak at this energy which decayed with a half-life of about 1 sec. Hence, this half-life is assigned to ^{106}Nb.

IV. CONCLUSIONS

The present work shows that the well-known isomerism in odd-odd niobium isotopes extends to at least mass number 104. The formation of both low- and high-spin isomers in fission offers the possibility of studying excited states in the even-even molybdenum daughters over a large energy and spin range. These nuclei are located in a region of transitional nuclei which are soft towards β and γ deformations. For ^{98}Mo, e.g., it has been shown by Gneuss and Greiner[15] that the spectrum of low-lying states can be explained with a potential energy surface having two minima separated only by a relatively flat barrier. One minimum, corresponding to the ground state, occurs for the spherical shape; the other one, responsible for the low-lying excited 0^+ state, is situated in the γ plane at a deformation of $\beta \approx 0.45$. Sheline, Ragnarsson, and Nilsson[16] have suggested that an inversion of the two minima occurs in heavier molybdenum isotopes: the deformed shape, now, represents the ground state, and the spherical shape an excited 0^+ state. In this context, detailed information on the level structure of ^{104}Mo and ^{106}Mo should be of great interest.

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