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Structure of beta-decaying states in the deformed, neutron-rich nucleus ^{104}Nb RECEIVED
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Excited structures in ^{104}Mo were populated by β decays of the ground and isomeric states in the neutron-rich nucleus ^{104}Nb . The beams were produced by the CARIBU facility at Argonne National Laboratory, re-accelerated by the ATLAS accelerator and implanted on a moving-tape system in the middle of the GAMMASPHERE array. Separate decay schemes for the two β -decaying states in ^{104}Nb were constructed for the first time. The structure of the isomers are discussed in the framework of the deformed Nilsson model and systematics of known quasiparticle structures in neighboring nuclei.

1. Introduction

Neutron-rich nuclei in the region near $A \sim 100$ and $Z \sim 40$ are known to exhibit an evolution from spherical to prolate-deformed shape near $N = 60$, as well as shape coexistence (see for example [1, 2] and references therein). The directly measured quadrupole moments and mean charge radii for the odd-Z Y ($Z = 39$) and Nb ($Z = 41$) nuclei with $N = 60$ and 62 [3, 4] indicate stable prolate-deformed shapes, which coupled with the measured magnetic moments, allowed for the assignment of the Nilsson orbitals associated with the ground state for these nuclei. The structure of neighboring odd-odd nuclei is not well known, partially because of the increased density of low-lying states and the additional effect of the spin-dependent residual proton-neutron interactions. There are also conflicting predictions by various theoretical models. For example, the FRDM2016 framework [5] suggests strongly prolate-deformed shapes for the odd-odd Y and Nb nuclei with $N = 61$ –67, while the Quasiparticle Random-Phase Approximation (QRPA) with Skyrme energy-density functional [6] predicts prolate- and oblate-deformed shapes for the same Y and Nb nuclei, respectively.

Spectroscopic information on the odd-odd ^{104}Nb ($Z = 41$, $N = 63$) nucleus is summarized in the latest Evaluated Nuclear Structure Data File (ENSDF) evaluation [7] and in several more recent publications [8–10]. Two β -decaying states, one of low spin and longer half-life [$I^\pi = (1^+)$, $T_{1/2} = 4.9(3)$ s] and the other of a shorter half-life [$T_{1/2} = 0.94(4)$ s], but without a spin assignment, are established with the former proposed to be the ground state [7]. The decay properties of the two β -decaying states in ^{104}Nb are of interest to the nuclear structure and nuclear astrophysics [11, 12] communities, as well as for nuclear applications, such as decay heat from nuclear reactors [13] and antineutrino spectra reconstructions [14].

In the present work, we report on $\beta - \gamma - \gamma$ coincidence studies of ^{104}Nb using the recently developed decay station at Argonne National Laboratory and the GAMMASPHERE spectrometer [15]. The new results allowed for the first construction of separate decay schemes of the two β -decaying states and these results resolved ambiguities that existed from previous studies. Their structures are discussed using the deformed Nilsson model along with systematics of known quasiparticle structures in neighboring nuclei.

2. Previous studies

The first $\beta - \gamma - \gamma$ coincidence studies of ^{104}Nb were reported by Ahrens *et al* [16], where two β -decaying states with half-lives of 0.8(2) s and 4.8(4) s were identified. The latter work of Kern *et al* [17] confirmed the existence of two β -decaying states and reported a half-life of 0.99(7) s for the shorter-lived one. A decay scheme comprising of 24 excited levels was also proposed in [17], however, it was not specified which states in the ^{104}Mo daughter nucleus were fed by the shorter-lived or the longer-lived states, or possibly from both. Although it was noted that the population of the $I^\pi = 6^+$ level of the ground-state band in ^{104}Mo from the shorter-lived state and of the $I^\pi = 0^+$ level at 887 keV from the longer-lived one, constitute relatively high and low spins to the two β -decaying states, respectively. Additional half-life information on the two β -decaying states in ^{104}Nb was reported by Mehren *et al* [18], where β -delayed neutron emissions from both states were measured. Recently, the half-life of the shorter-lived β -decaying state was measured as 0.97(10) s by Dombos *et al* [8] and its decay was studied using the Total Absorption Gamma-ray Spectroscopy (TAGS) technique [9]. Early results from the CARIBU facility at Argonne National Laboratory reported a half-life of 0.97(1) s for the short-lived state as well [10].

The energy separation between the two β -decaying states in ^{104}Nb was reported to be 215(120) keV by Graefenstedt *et al* [19] using β -decay end-point energy measurements, where they associated the isomer with the shorter-lived state. Recently, Penning trap mass measurements by Orford [20] and Hukkanen [21] revealed that the energy separation between the long- and short-lived β -decaying states in ^{104}Nb was much smaller, being only 9.8 keV, but they have not provided information about their ordering.

Excited structures in ^{104}Nb and its ^{104}Mo daughter were also studied in spontaneous fission of ^{252}Cf in [22, 23] and [24–27], respectively. Additional information on the level structures of ^{104}Mo was obtained in spontaneous fission of ^{248}Cm [28] and in α -induced fission of ^{238}U [29].

3. Experiment details

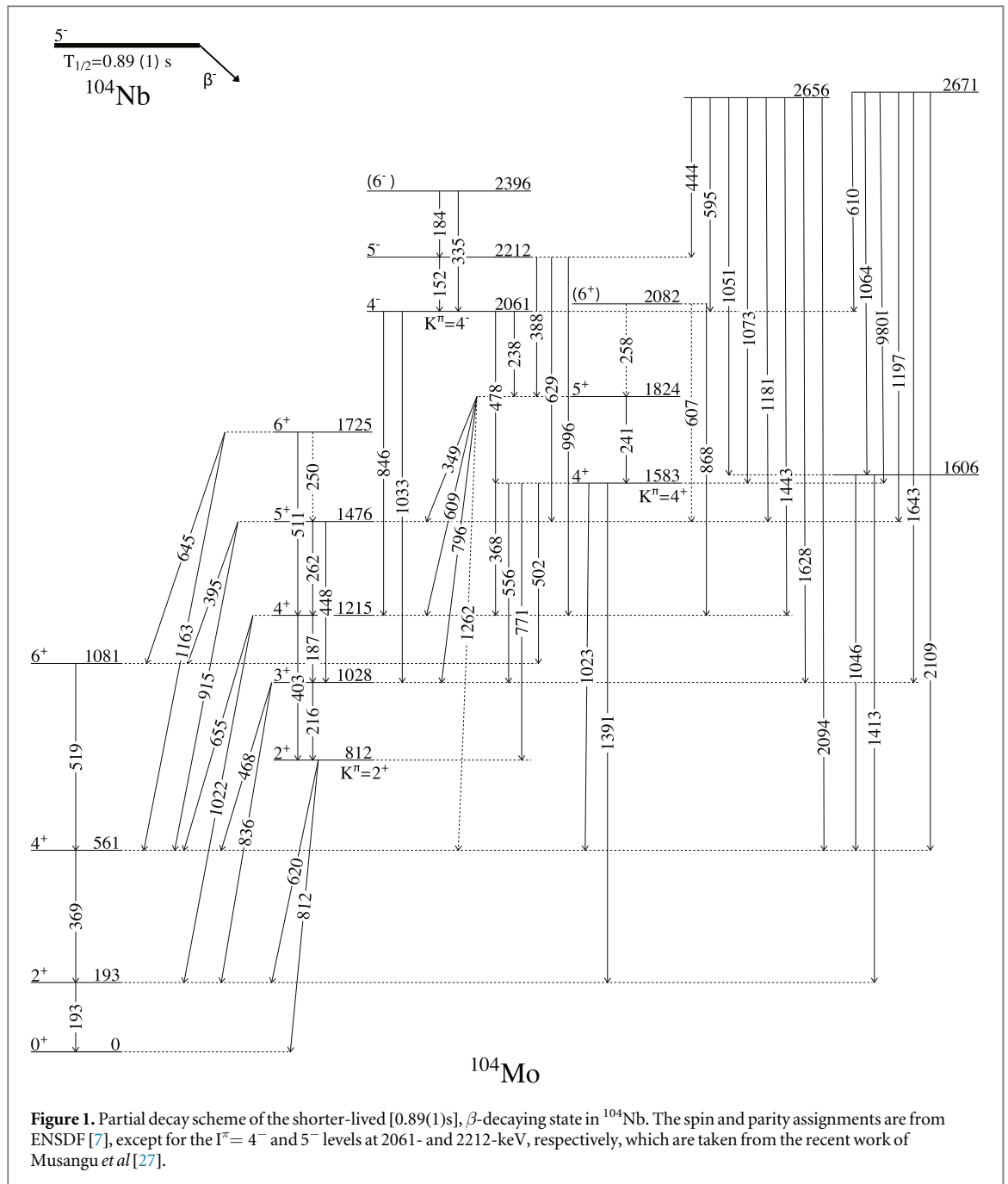
Beta-decay spectroscopy studies were conducted at the Californium Rare Isotope Breeder Upgrade (CARIBU) facility [30], located at the Argonne Tandem Linear Accelerator System (ATLAS) facility at Argonne National Laboratory. Low-energy CARIBU beams of the two β -decaying states in ^{104}Nb , produced in the spontaneous fission of ^{252}Cf with a typical intensity of $\sim 10^4$ ions/s, were re-accelerated by ATLAS and implanted onto a 0.5-inch wide Mylar moving tape. The implantation point was surrounded by an array of six plastic scintillator detectors, known as Hexagonal ARray for Triggering (HEART) [15], which was used to detect β particles. The system was located in the center of the GAMMASPHERE spectrometer, comprising of 36 Compton-suppressed HPGe detectors for this experiment. Energy and efficiency calibrations of the array was carried out using ^{56}Co , ^{152}Eu , ^{182}Ta and ^{243}Am sources. Given the previously known half-lives of the two β -decaying states in ^{104}Nb [7], moving-tape cycles of 10 s implantation time and 20 s decay time (20 s cycle), as well as 10 s implantation time and 40 s decay time (40 s cycle), were used in the present studies. The collected data were sorted offline into various one- and two-dimensional histograms using the GEBsort code [31]. The data analysis was performed with the ROOT [32] and the Radware [33] software packages. Several matrices were generated, including β -particle-gated E_γ - E_γ coincidence matrices that were used to construct the decay schemes, as well as a β -particle-gated E_γ -time histogram that was used to deduce the half-lives of the observed β -decaying states.

4. Experimental results

The present work confirmed the existence of two β -decaying states in ^{104}Nb and for the first time established separate decay schemes for the shorter- and the longer-lived states, as shown in figures 1–4.

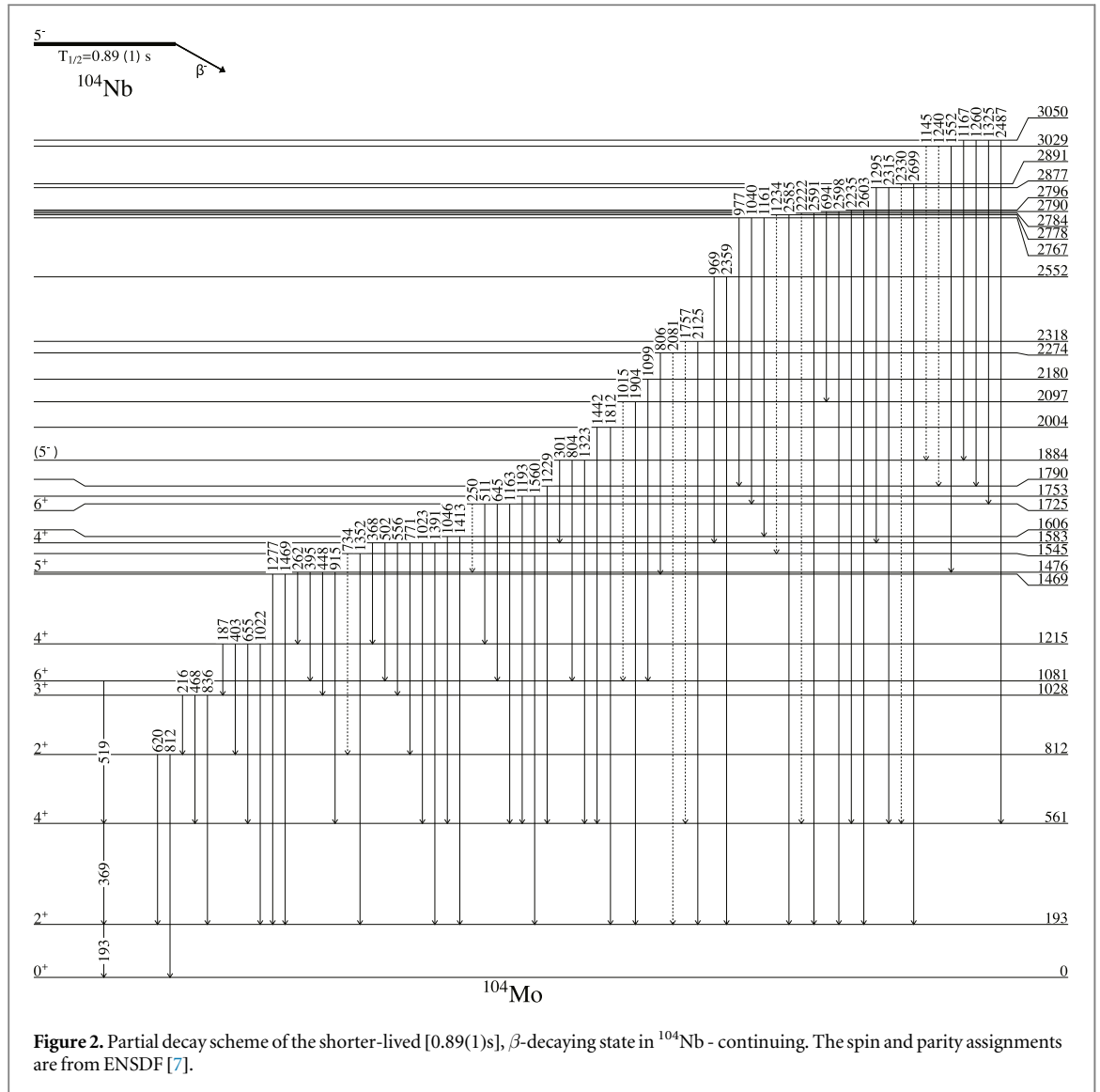
The half-lives of the two β -decaying states were obtained from background-subtracted time spectra produced by gating on individual γ rays in the E_γ -time histogram. Figure 5(a) shows a time spectrum produced by summing gates on the 478- and 771-keV γ rays that depopulate the 2061- and 1583-keV levels in ^{104}Mo , respectively. A least-squares fit using a single-exponential decay and a constant background resulted in a half-life of 0.89(1) s for the short-lived, β -decaying state in ^{104}Nb , which is consistent, but more precise, with the previously reported values [7, 8, 10]. Figure 5(b) shows a time spectrum generated by summing gates on the 193-, 369- and 812-keV γ -rays, depopulating the lowest-lying states in the daughter nucleus ^{104}Mo . The spectrum has a complex shape and it was fitted with two-exponential decays where the half-life of shorter component was fixed to 0.89(1) s. The half-life of the long-lived state was obtained as 6.1(1) s, which is somewhat longer compared to the previously measured values of 4.8(4) s [16] and 5.0(4) s [18], as well as the ENSDF recommended value of 4.9(3) s [7].

The strongest decay branch [$I_\beta = 36.0(8)\%$] of the shorter-lived, β -decaying state in ^{104}Nb was found to populate the $K^\pi = 4^-$ state at 2061 keV in the daughter nucleus ^{104}Mo . The spin assignment of the latter was



unambiguously established by the angular correlation data analysis of Musangu *et al* [27]. A γ -ray coincidence spectrum produced by gating on the 478-keV γ ray, directly depopulating the 2061 keV level, is shown in figure 6. The observed direct β -decay feedings to the $I^\pi = 5^-$ and (6^-) members of the $K^\pi = 4^-$ band (note the 152-keV and the weaker 184-keV and 335-keV γ rays in figure 6), coupled with the allowed-unhindered nature ($\log ft = 5.08$) of the β^- transition to the $K^\pi = 4^-$ band head at 2061 keV, unambiguously establishes $K^\pi = I^\pi = 5^-$ for the shorter-lived β -decaying state in ^{104}Nb . It is worth noting that we have not observed any direct β -decay feedings from the shorter-lived state in ^{104}Nb to levels of the $K^\pi = 0^+$ ground-state band of ^{104}Mo , presumably because of their $\Delta K=5$ forbidden nature. In contrast, the recent work of Gombas *et al* [9] reported a direct β -decay transition from the shorter-lived β -decaying state in ^{104}Nb to the $I^\pi = 4^+$ member of the ground-state band of ^{104}Mo . Using $I_\beta = 2.9(6)\%$ for this branch [9], one can deduce a $\log ft = 6.4$ value, whereas $\log ft \sim 16$ can be expected for such a $\Delta K=5$, K-forbidden branch [34]. A direct β -decay feeding of $I_\beta = 5.6(13)\%$ from the shorter-lived, β -decaying state in ^{104}Nb to the $K^\pi = 0^+$ ground state of ^{104}Mo was also reported in [9] which is also remarkable since one can obtain $\log ft = 6.6$ that would imply unprecedented (and most likely spurious) strength for such a 5-fold forbidden β -decay transition.

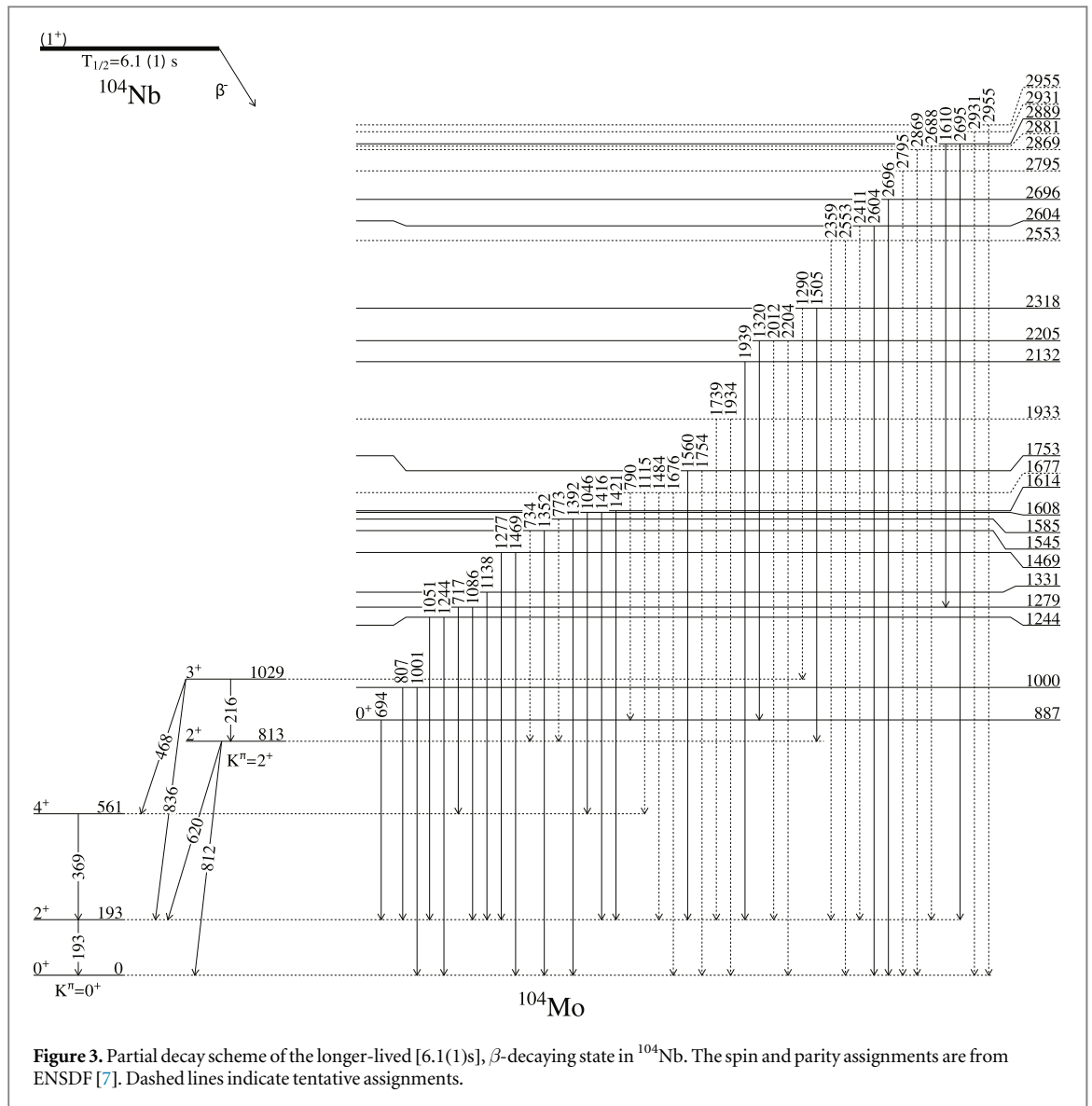
Figure 7 shows a β -gated, γ -ray singles spectrum produced with a time gate between 12 s and 20 s from the 20 s tape-cycle data. It contains only γ rays that belong to the decay of the longer-lived, β -decaying state in ^{104}Nb .



The γ rays associated with decays of the daughter (^{104}Mo) and grand daughter (^{104}Tc) nuclei were removed by subtracting, after appropriate normalization, a singles spectrum produced with a time gate between 30 s and 40 s from the 40 s tape-cycle data. We found that the decay of the longer-lived state in ^{104}Nb is very different compared to that of the shorter-lived one because of: (a) the direct β -decay population of the $I^\pi = 2^+$ and 0^+ levels at 813- and 887-keV, respectively, as evident by the presence of strong 620-, 812- and 694-keV γ rays, depopulating these levels in figure 7, and (b) the absence of any decay paths via the $I^\pi = 4^-$ and/or 4^+ states at 2061keV and 1583keV in ^{104}Mo [7, 27], respectively, which are strongly populated in the β -decay of the shorter-lived state. This implies that the 6.1 s state in ^{104}Nb most-likely has $I^\pi = (1^+)$, in agreement with the proposed assignment in ENSDF [7]. Another feature of the decay scheme is that the β -decay branches are spread over many highly-excited levels in ^{104}Mo which predominantly decay via a single γ ray to the $I^\pi = 0^+$ ground state or via two γ rays in parallel, one to the $I^\pi = 0^+$ ground state and the other to the first-excited $I^\pi = 2^+$ state in ^{104}Mo . This decay pattern is also consistent with the low-spin nature of the 6.1 s state in ^{104}Nb . It is worth noting that despite the assigned $I^\pi = (1^+)$ to the longer-lived β -decaying state, we have not observed a significant direct β -decay branch to the $I^\pi = 0^+$ ground state of ^{104}Mo , but we are able to established a limit of $I_\beta < 5\%$.

5. Discussion

The current ENSDF evaluation for ^{104}Nb [7] associates the long-lived activity with the ground state, while the shorter-lived one is assigned as an isomer with an excitation energy of 220(120) keV. The $I^\pi = (1^+)$ assignment is tentatively proposed for the former, while no spin and parity were established for the isomer [7]. Potential Energy Surface (PES) and Projected Shell Model (PSM) calculations by Luo *et al* [23] indicate that the ground state is best associated with the strongly-deformed, prolate $K^\pi = 1^+, \pi 3/2[301] \otimes \nu 5/2[532]$ configuration. The

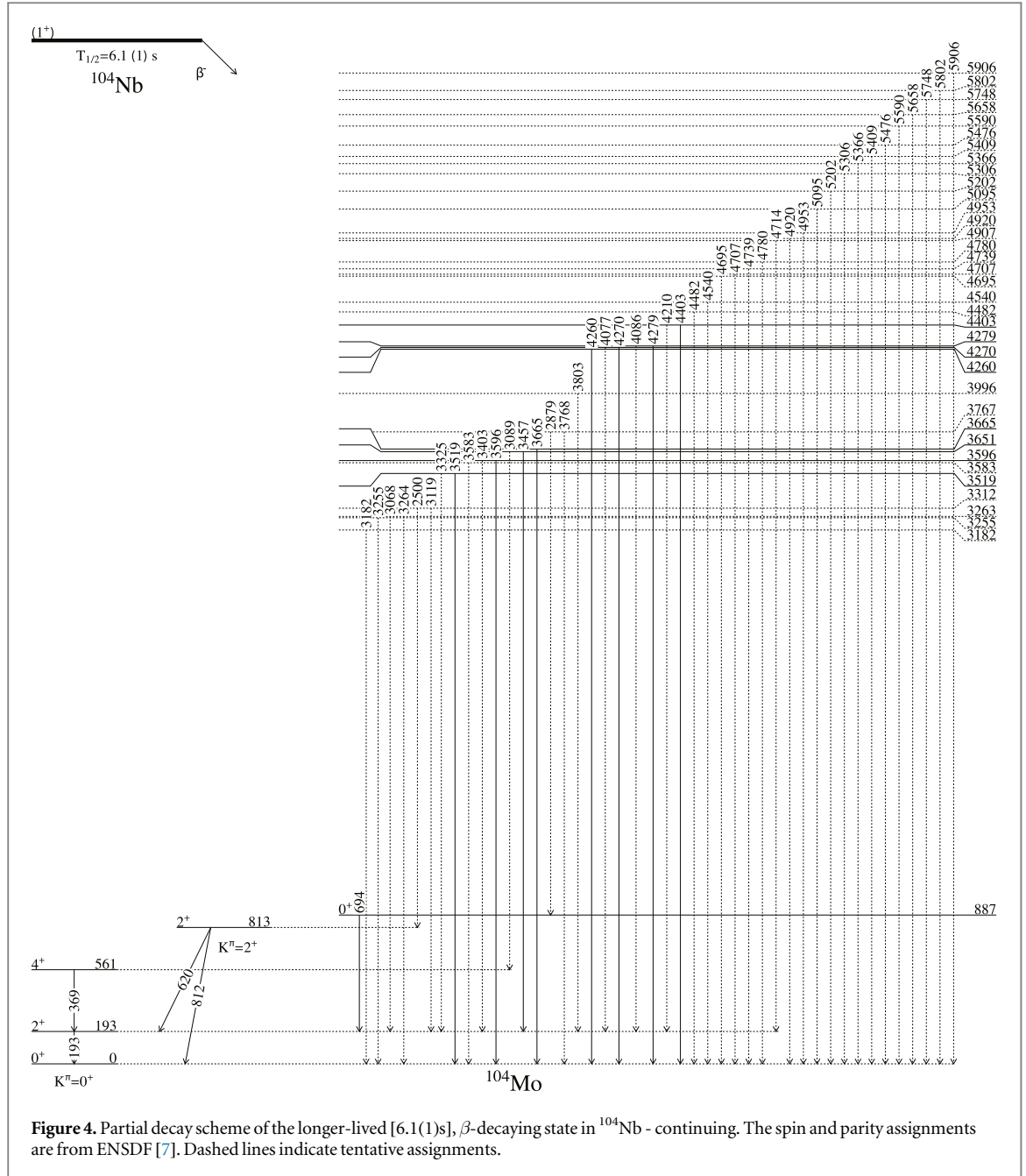


same assignment, guided by results from QRPA calculations, was also favored by Gombas *et al* [9]. It should be noted, however, that the proposed $K^\pi = 1^+, \pi 3/2[301] \otimes \nu 5/2[532]$ configuration for the ^{104}Nb ground state would break the Gallagher-Moszkowski rule [35], since the triplet $K^\pi = 4^+, \pi 3/2[301] \otimes \nu 5/2[532]$ state would be expected to be lowest in energy.

In the present work, we propose an alternative interpretation of the structure of the two β -decaying states in ^{104}Nb . Systematics of experimentally-observed one-quasiparticle states in neighboring ^{103}Nb ($Z = 41$) and ^{105}Mo ($N = 63$) nuclei are shown in figure 8 (left). The directly measured spin and moments (both quadrupole and magnetic) associate the ground state of ^{103}Nb with the $\pi 5/2[422]$ Nilsson proton orbital [4], while the excited states at 164 keV and 248 keV are assigned the $\pi 5/2[303]$ and $\pi 3/2[301]$ orbitals, respectively [36]. On the neutron side, the measured magnetic moment [37] and the properties of the observed ground-state rotational band [38] of ^{105}Mo ($N = 63$) suggest that the $\nu 5/2[523]$ Nilsson orbital is the closest to the neutron Fermi surface at $N = 63$. The $\nu 3/2[411]$ orbital is assigned to the excited 247-keV level, the $\nu 5/2[413]$ one to the 310-keV level and the $\nu 1/2[411]$ to the 332-keV level [38], as shown in figure 8 (left). This ordering is also supported by the fact that the $\nu 3/2[411]$ orbital is associated with the ^{103}Mo ($N = 61$) ground state [37], while the $\nu 5/2[413]$ orbital is assigned to the ^{107}Mo ($N = 65$) ground state with the $\nu 1/2[411]$ one being 65 keV higher [39] (note that the ordering of the $\nu 5/2[413]$ and $\nu 1/2[411]$ orbitals in ^{107}Mo was reversed in [40]).

As discussed in [41], the excitation energy of a given two-quasiparticle state in deformed odd-odd nuclei can be expressed as:

$$E_{KI}^{\pi\nu} = E_{qp}^{\pi} + E_{qp}^{\nu} + a[I(I+1) - K^2] + V^{\pi\nu} \quad (1)$$



where $E_{qp}^{\pi(\nu)}$ is the quasiparticle energy for the odd proton (neutron), $a = \hbar^2/2\mathcal{J}$ is the rotational constant, \mathcal{J} is the moment of inertia and $V^{\pi\nu} = \frac{\Delta E_{GM}^{\pi\nu}}{2} + (-1)^I [B_N^{\pi\nu} + E_a^{\pi\nu}] \delta_{K,0}$ is the energy shift due to residual proton-neutron interactions, where $\Delta E_{GM}^{\pi\nu}$ is the Gallagher-Moszkowski splitting energy, $B_N^{\pi\nu}$ is the Newby shift and $E_a^{\pi\nu}$ is the rotation-particle coupling term, which contributes only when $\Omega(\pi) = \Omega(\nu) = 1/2$. Following the Gallagher-Moszkowski rule [35], the sign of $\Delta E_{GM}^{\pi\nu}$ is positive when the proton and neutron spins are coupled anti-parallel and negative for a parallel coupling, while the sign of $B_N^{\pi\nu}$ can be either positive or negative.

By combining the observed proton and neutron states in ^{103}Nb and ^{105}Mo and by applying equation (1) (for simplicity $a = 17.3$ keV, corresponding to 70% of the rigid-body value of the moment-of-inertia was used for all states), the predicted lowest energy two-quasiparticle states in ^{104}Nb are shown in figure 8 (right). Since $\Delta E_{GM}^{\pi\nu}$ and $B_N^{\pi\nu}$ values for the involved Nilsson orbitals are not experimentally known in the $A \sim 100$ mass region, we assumed $|\Delta E_{GM}^{\pi\nu}| = 190$ keV and $B_N^{\pi\nu} = 50$ keV for all configurations. Similar values are not uncommon in the better studied rare-earth region [41].

As can be seen from figure 8 (right), the $K^\pi = 5^-, \pi 5/2[422] \otimes \nu 5/2[532]$ configuration is predicted to be the ground state of ^{104}Nb , which can be associated with the shorter-lived, $K^\pi = 5^-$ β -decaying state from the present work. Consequently, the fast β -decay branch with $\log ft = 5.08$ to the $K^\pi = 4^-$ state at 2061 keV in ^{104}Mo indicates that this is an allowed, spin-flip transition, which requires the following selection rules for the Nilsson orbital parameters: $\Delta N = 0$, $\Delta n_z = 0$, $\Delta \Lambda = 0$, $\Delta \Omega = \pm 1$ [43, 44]. The cascade to crossover branching ratio for the

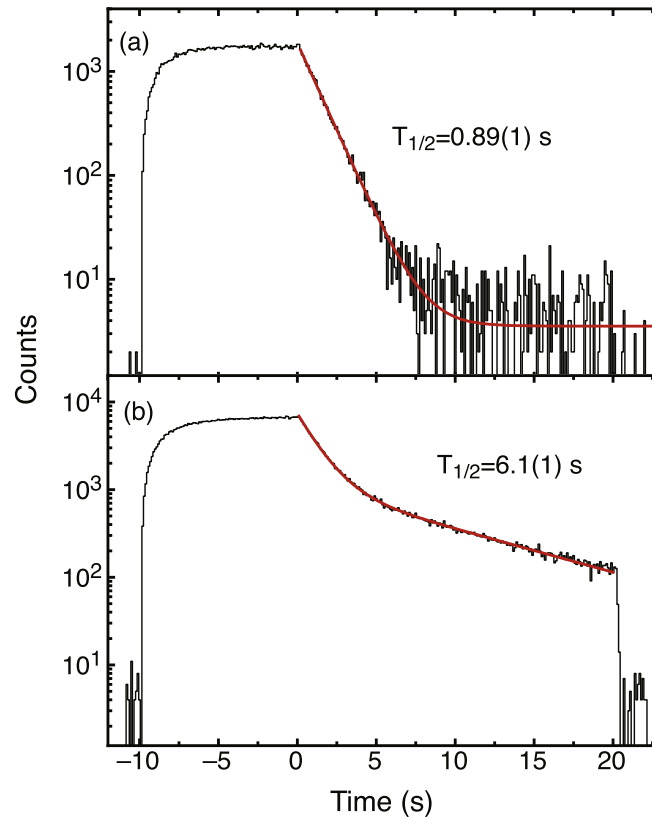


Figure 5. Background-subtracted time spectra produced by gating on: (a) sum of the 478- and 771-keV γ rays that follow the decay of the shorter-lived, β -decaying state in ^{104}Nb . The solid line represents a least-squares fit using a single-exponential decay and a constant background; (b) sum of the 193-, 369- and 812-keV γ rays that follow the decay of the two β -decaying states in ^{104}Nb . The solid line represents a least-squares fit using two-exponential decays with the half-life of the shorter component fixed to 0.89(1) s.

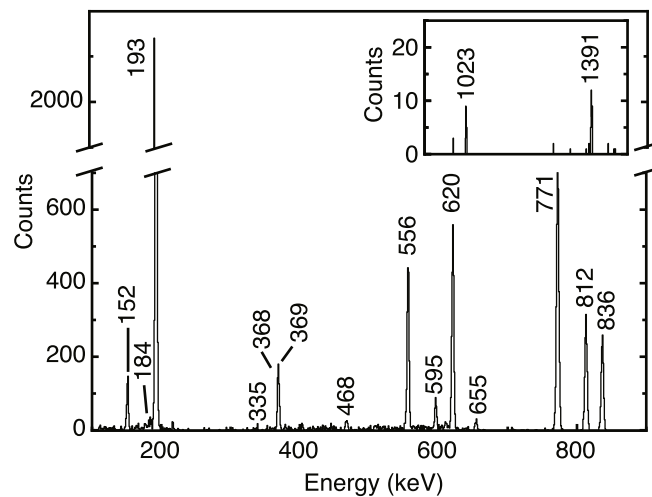
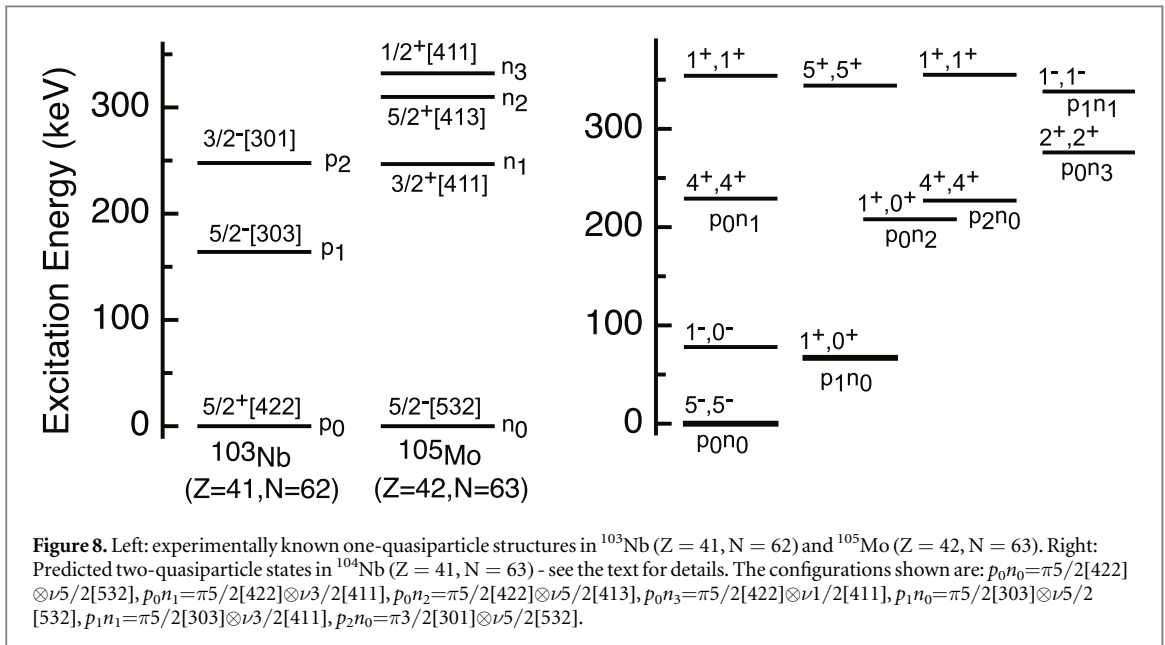
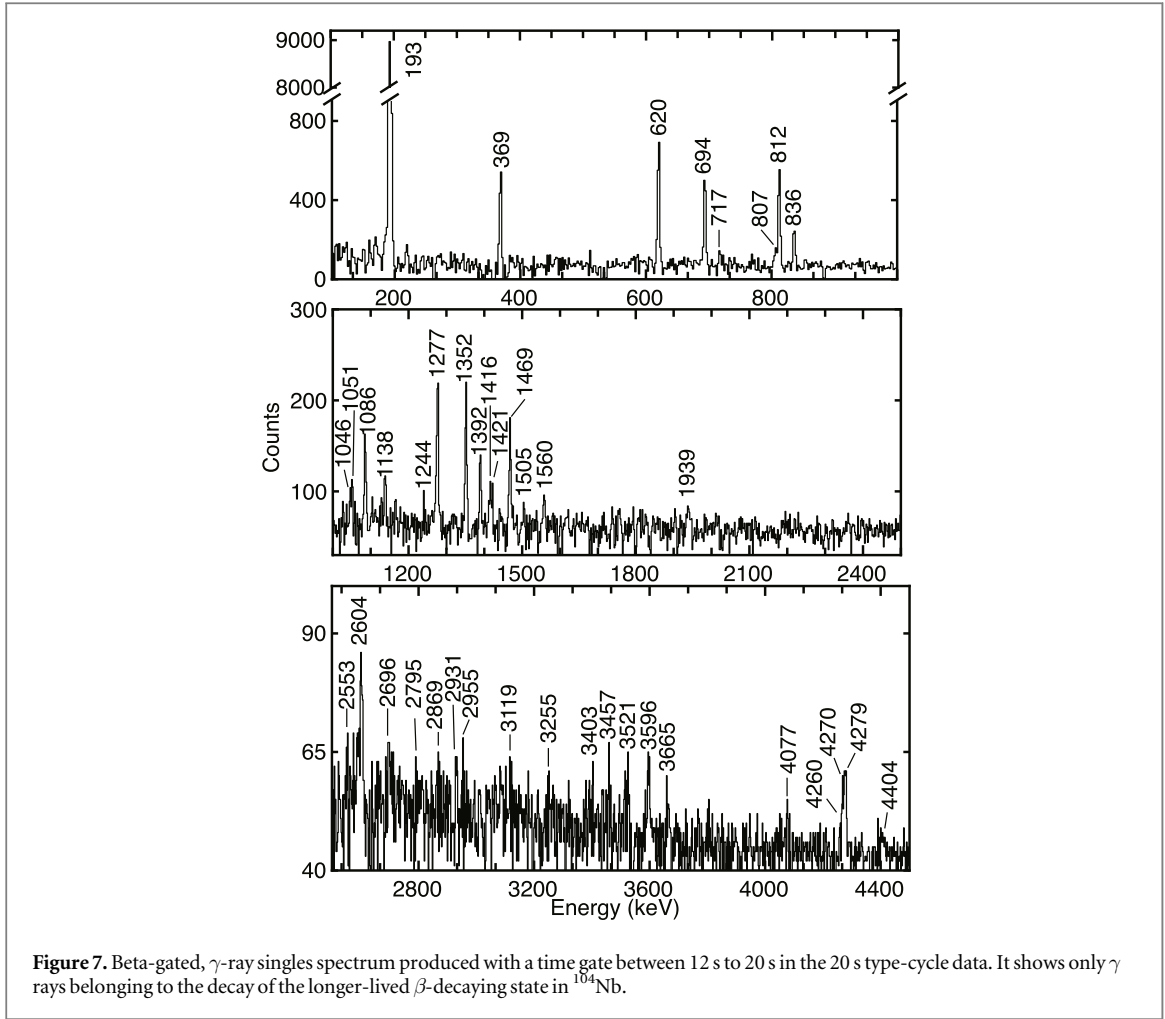
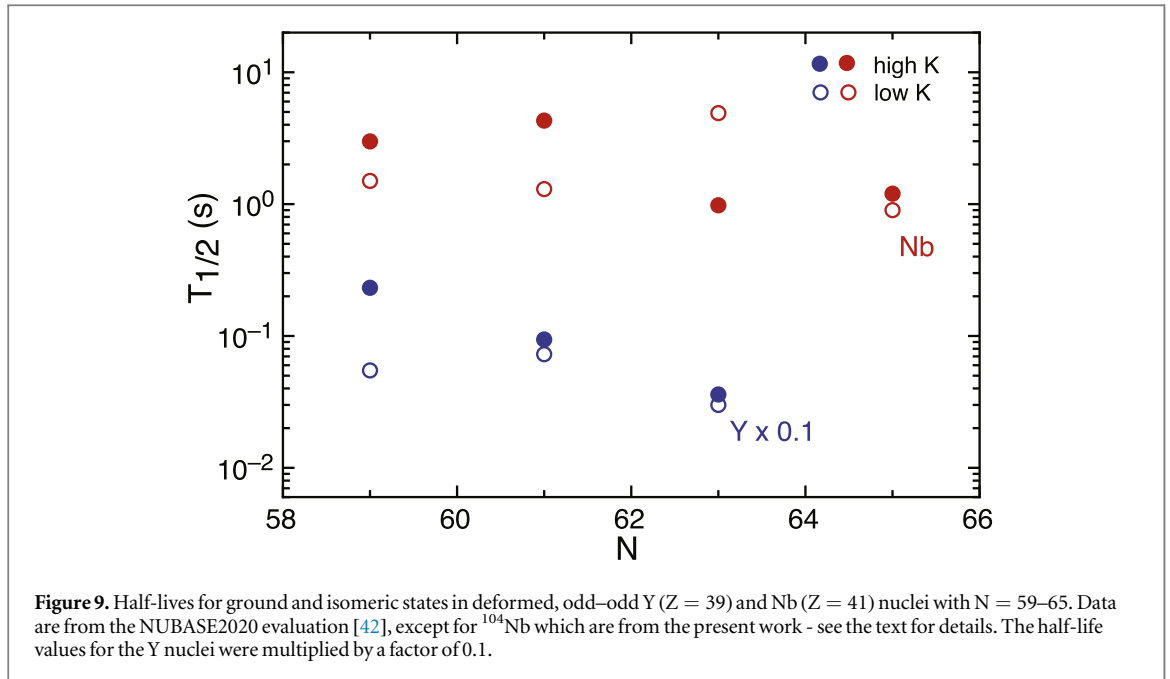


Figure 6. Beta-gated, γ - γ coincidence spectrum produced by gating on the 478-keV γ ray, depopulating the $K^\pi = 4^-$ level at 2061-keV in the ^{104}Mo daughter nucleus.

$I^\pi = (6^-)$ level of the $K^\pi = 4^-$ band was measured, which allowed for the $|g_K - g_R| = 0.28(5)$ value to be determined, where g_K and g_R are the intrinsic and rotational g factors, respectively. This is in agreement with the Nilsson model predicted value of $|g_K - g_R| \approx 0.39$ for the two-quasineutron $\nu^2(3/2[422], 5/2[532])$ configuration. The competing $K^\pi = 4^-$ two-quasiproton $\pi^2(3/2[301], 5/2[422])$ and two-quasineutron $\nu^2(3/2[411], 5/2[532])$ configurations are predicted to have $|g_K - g_R|$ values of 1.37 and 0.81, respectively. Thus, the allowed, spin-flip transition to the $K^\pi = 4^-$ level at 2061 keV in ^{104}Mo implies the $\nu 3/2[422] \rightarrow \pi 5/2[422]$ Nilsson orbital change, in agreement with the proposed $K^\pi = 5^-, \pi 5/2[422] \otimes \nu 5/2[532]$ configuration to the ground state of ^{104}Nb .



Our calculations predict that the 9.8-keV isomer in ^{104}Nb most-likely originates from the $I^\pi, K^\pi = 1^+, 0^+ \pi 5/2[303] \otimes \nu 5/2[532]$ configuration. The competing $I^\pi, K^\pi = 1^-, 0^- \pi 5/2[422] \otimes \nu 5/2[532]$ configuration is unlikely to be involved, since it could decay via an allowed, spin-flip $\nu 3/2[422] \rightarrow \pi 5/2[422]$ transition to a $K^\pi = 1^-, \nu 3/2[422] \otimes \nu 5/2[532]$ state in ^{104}Mo , expected approximately 200 keV higher than the experimentally observed $K^\pi = 4^-$ state at 2061 keV that has the same two-quasineutron configuration. However, no such a decay was experimentally observed in the present work. The proposed configuration for the isomer can explain



the observed hindered decay ($\log ft > 7.3$) to the $I^\pi, K^\pi = 0^+, 0^+$ ground state in ^{104}Mo . In accordance with the β -decay selection rules for deformed nuclei [43, 44], such a transition involves the $\nu 5/2[532] \rightarrow \pi 5/2[303]$ Nilsson orbital changes and, therefore, it is $\Delta N = 2$ and $\Delta n_z = 3$ forbidden. It is worth noting that a similar hindered transition ($\nu 5/2[642] \rightarrow \pi 5/2[413]$) is responsible for the long-lived nature [$\log ft = 9.83(4)$] of the $I^\pi, K^\pi = 0^+, 0^+$ ground state of ^{156}Eu [45].

Figure 9 shows systematics of known half-lives for ground and isomeric states in several deformed, odd-odd Y and Nb nuclei in the $A \sim 100$ region. A general feature is that the half-lives of the high-K β -decaying states are always longer compared to those for the low-K ones, owing to the reduced effective Q_β value for the former [34]. However, as can be seen from figure 9, this is not the case for ^{104}Nb , where the low-K state is longer lived. This anomaly can be explained with the underlining nuclear structure of the two β -decaying states in ^{104}Nb , namely, the dominant fast, spin-flip decay of the $K^\pi = 5^-$ ground state and the $\Delta N = 2$ and $\Delta n_z = 3$ forbidden decay of the $I^\pi = (1^+)$ isomer.

6. Conclusion

Excited states in ^{104}Mo were populated in β^- decay of the neutron-rich nucleus ^{104}Nb , produced by the CARIBU facility at Argonne National Laboratory. The present data confirm the existence of two β -decaying states in ^{104}Nb and half-life values of 0.89(1) s and 6.1(1) s were measured by tagging on specific γ rays associated with their decays. For the first time separate decay schemes were established for the two β -decaying states and their ordering was revised with respect to previous studies and evaluated nuclear data. The structure of ^{104}Nb is discussed using the deformed Nilsson model and systematics of known quasiparticle structures in neighboring nuclei. The ground state of ^{104}Nb is associated with the shorter-lived [$T_{1/2} = 0.89(1)$ s] state and assigned the $K^\pi = 5^-, \pi 5/2[422] \otimes \nu 5/2[532]$ Nilsson configuration. The 9.8-keV isomer is proposed to originate from the $K^\pi = 0^+, \pi 5/2[503] \otimes \nu 5/2[532]$ configuration and it is associated with the longer-lived [$T_{1/2} = 6.1(1)$ s] state. The longer β -decay half-life of the low-spin isomer is in contrast to systematics observed in neighboring deformed, odd-odd Y and Nb nuclei. It is most-likely associated with the $\Delta N = 2$ and $\Delta n_z = 3$ forbidden nature of the decays to low-lying states in the daughter nucleus ^{104}Mo .

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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