New interpretation of shape coexistence in ^{99}Zr

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Levels in ^{99}Zr populated by β decay of on-line mass separated ^{99}Y have been studied by γ-ray spectroscopic methods, including γ-γ coincidences recorded with an array of ten Ge detectors and level-lifetime measurements. The formerly reported strongly collective character of the 53-keV transition turns out to be questionable. This implies a revision of the experimental evidence for shape coexistence in ^{99}Zr. Transition rates and γ-ray branching ratios make a new level at 679 keV with $t_{1/2}=9$ ns another candidate for a deformed state. [S0556-2813(97)00711-5]

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I. INTRODUCTION

The region of neutron-rich Sr, Y, and Zr isotopes with $N=58$–60 is the stage of a dramatic transition of nuclear ground-state shape. The extreme lowering of the 2^+ energy, from 1223 keV in the spherical nucleus ^{98}Zr to 212 keV in the strongly deformed nucleus ^{100}Zr, is the most obvious observable. Shape coexistence has been reported for both nuclei. In the spherical nucleus ^{98}Zr, the 0_3^+ state at 1436 keV was interpreted as deformed, based on the $\rho^2(E0,0_3^+\rightarrow 0_2^+)$ value of 0.08 implying a large mixture of states with very different mean-square radii [1]. In the deformed nucleus ^{100}Zr, the very low-lying 0_2^+ level at 331 keV is assumed to be spherical or slightly deformed. Detailed calculations of transition rates accounting for level mixing have been performed by the TRISTAN group [2]. Thus, shape coexistence is expected to occur for the intermediate odd-neutron nucleus ^{99}Zr. A level scheme of ^{99}Zr, based on γ-γ coincidences measured at the OSTIS separator at the ILL-Grenoble, was presented in Ref. [1]. The low-lying levels in ^{99}Zr are spherical, but the 614- and 724-keV levels have been proposed to be deformed states associated with the [541]3/2 and [422]3/2 Nilsson orbitals, respectively. Several authors reported large enhancement of the E2 component in the 53-keV transition from the 667-keV level to the 614-keV level, suggesting the beginning of a band [3]. Recently, prompt-fission experiments were performed in this region. The [422]3/2 band in ^{97}Sr [4], the odd-neutron isotone of ^{99}Zr, could be extended to $I=11/2$ [5] but no band structure was observed in ^{99}Zr [6]. In order to clarify the nature of the states proposed as being deformed in Ref. [1], we have reinvestigated the decay of ^{99}Y to ^{99}Zr with high statistics and high-quality γ-γ coincidence data.

II. EXPERIMENT

The ^{99}Y parent activity ($T_{1/2}$=1.5 s [3]) of ^{99}Zr was obtained as a fission product of ^{238}U bombarded by 25 MeV protons at the K-130 cyclotron of the Jyväskylä University. The isobaric $A=99$ activities were separated by the ion-guide technique at the IGISOL mass-separator facility [7,8], allowing the production of ^{99}Y as a beam. The mass-separated beam was implanted on a movable tape viewed by various detection systems. In one experiment, γ-γ coincidences were recorded with the DORIS array consisting of nine medium size Ge detectors and a LEPS detector. In another experiment, β-γ-time and γ-γ-time coincidences were recorded with a thin plastic scintillator for the β particles and two Ge detectors for the γ rays, the detectors being close to the implanted source. The separator beam was pulsed in order to gain half-life information from growth and decay curves of γ-rays and to allow identification of the parent activity. While γ-γ coincidence data were exploited in a standard way to construct level schemes, X-γ coincidences were used for the determination of conversion coefficients by the fluorescence method and β-γ-time spectra for the determination of level half-lives. Details of the experiments and their analysis are reported in recent publications on the decays of ^{99}Nb [9] and ^{99}Zr [10].

III. RESULTS

The data do not lead to new information about a possible band structure based on the 724-keV level. Nevertheless, part of the level scheme in relation with the 614-keV level has to be revised. Energy spectra with gates on the 53 and 614 keV transitions show new transitions placed above the 614-keV level, see Fig. 1. As explained in this section, our new results lead to the some changes in the previous level scheme [1]—the addition of two new levels, at 678.6 and 867.6 keV, and the removal of the level at 755.0 keV. All levels below 667.8 keV are shown in Fig. 2, but only the two new levels above 667.8 keV are shown. The 87-keV transition, reported in [1] is not seen anymore. It could have been due to scattering of the 140.5-keV isomeric transition in ^{99}Tc [3] between the two detectors, giving a false indication of a 87.2-keV γ ray in coincidence with the 53.3-keV γ ray. Thus, the existence of the 755-keV level, based only on this transition, is not supported. This level was postulated to belong to the band whose lowest members are the 614- and 667-keV levels. It is also worth noticing that, in spite of the low detection limit

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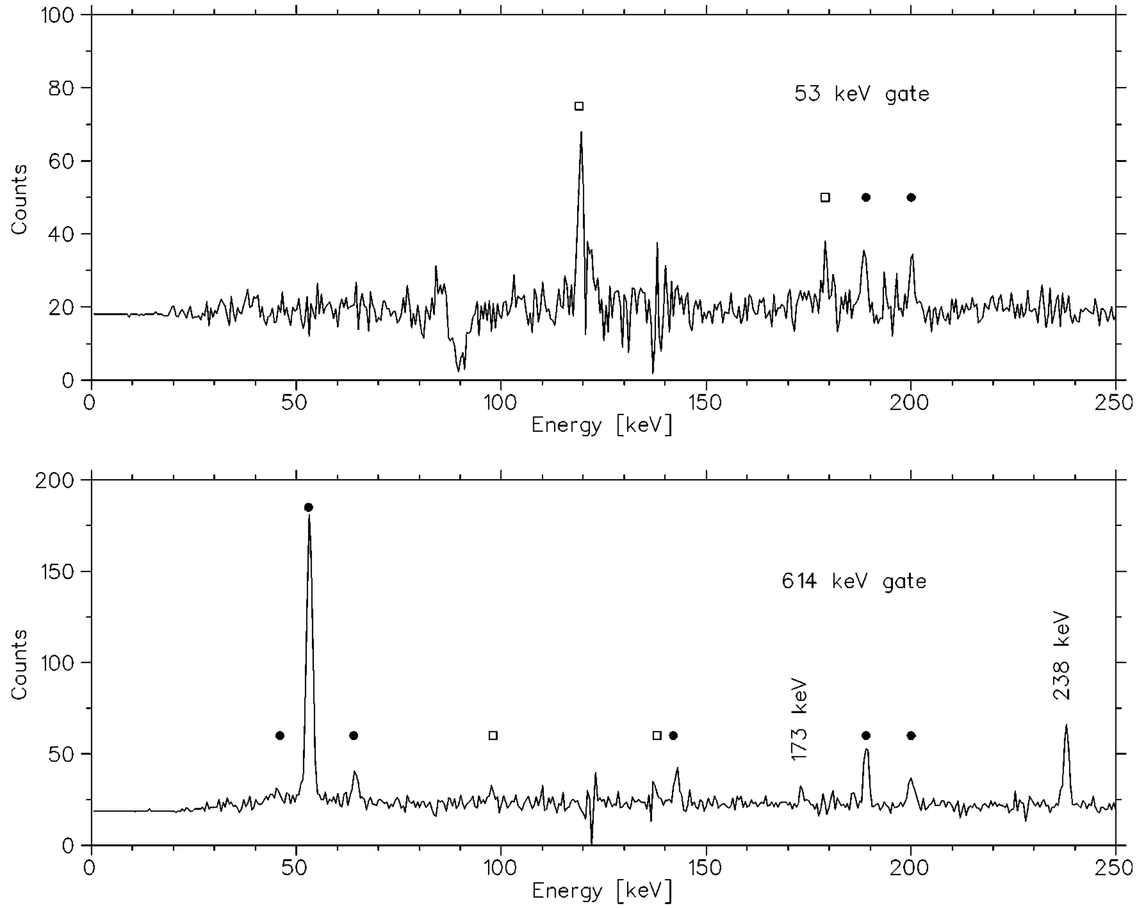

FIG. 1. Gates on the 53- (top) and 614-keV (bottom) transitions. Transitions discussed in the text are marked with a closed circle. Open squares refer to additional transitions. In the 53-keV gate, the 119-keV and 179-keV γ rays are due to overlap of the gate with transitions of 52 keV (^{98}Y [13,14]) and 56 keV (^{99}Nb [3]), respectively. In the 614-keV gate, the small peaks at 98 and 138 keV are random coincidences of γ rays in ^{99}Mo [3]. The 173- and 238-keV lines are listed in Table I.

(about 0.05 intensity units), no transitions from the 614-keV level to the 122- ($I^\pi=3/2^+$) and 252-keV ($I^\pi=7/2^+$) levels [3] are observed. Thus, the only decay mode of the 614-keV level is to the $I^\pi=1/2^+$ ground state. This favors a low spin for the 614-keV level, in agreement with $I^\pi=3/2^-$ proposed in Ref. [1]. The total amount of γ and e^- population of the 614-keV level is estimated, using data listed in Table I, to be about 11 relative intensity units. This implies a weak direct β feeding.

The coincidence data establish two new levels with strong decay branches to the 614- and 667-keV levels, see Fig. 2. A connection between the new level at 679 keV and the 667-keV level is required by the coincidences of the 189-keV transition (868→679) with the lines depopulating the 667-keV level, from which an estimate of the total intensity of the 11-keV linking transition is obtained. By the same method, comparison of the 53- and 614-keV peak intensities in the 200-, 1629- and 1733- keV gates yields the total intensity of the 53-keV transition. It is the first time that transitions above the 667-keV level can be used for this purpose. The deduced conversion coefficient $\alpha(53)=1.1(4)$ implies a lower conversion than the average of previous reports, $\alpha_K=2.0(6)$ [3]. Nevertheless, the present fluorescence measurement, comparing the intensities of the K-x-rays and of the 53-keV transition in the 614 keV gate, still gives $\alpha_K=2.4(6)$, in agreement with Ref. [3]. This discrepancy is removed after noticing the changes of the relative intensities of the 189-keV (868→679) and 200-keV (868→667) peaks in the 53- and 614-keV gates, see Fig. 1. This gives a measure of the total intensity of the new 64-keV transition (679→614) which turns out to be strongly converted with $\alpha=8(5)$, see Table II. Thus, extra K-x-rays from conversion of the 64-keV transition are responsible for the former reports of a higher conversion of the 53-keV transition. The new $\alpha(53)$ value does not rule out an $M1$ transition with a sizable $E2$ admixture. Nevertheless, a fairly pure $M1$ is a more probable alternative and even the $E1$ multipolarity cannot be excluded. Finally, the 46- and 143-keV transitions form a cascade which, in agreement with energy sums and weak coincidences, is parallel to the 189-keV transition. An appealing placement is to put the 46-keV transition at the bottom of the cascade, since the intermediate level would have an energy of 724.3 keV, perfectly matching the well established 724.3-keV level (not shown in Fig. 2). The γ-γ coincidences, when setting gates on the transitions feeding the 724-keV level, are too weak to provide evidence for a 46-keV transition from the 724-keV level. The detection limit corresponds to about 20% of the 724-keV γ-ray intensity ($I_\gamma=37$ [1]), yet much above the expected intensity to be observed. Therefore, the order of the 46- and 143-keV transitions cannot be determined.

The analysis of the slope of the time distribution between

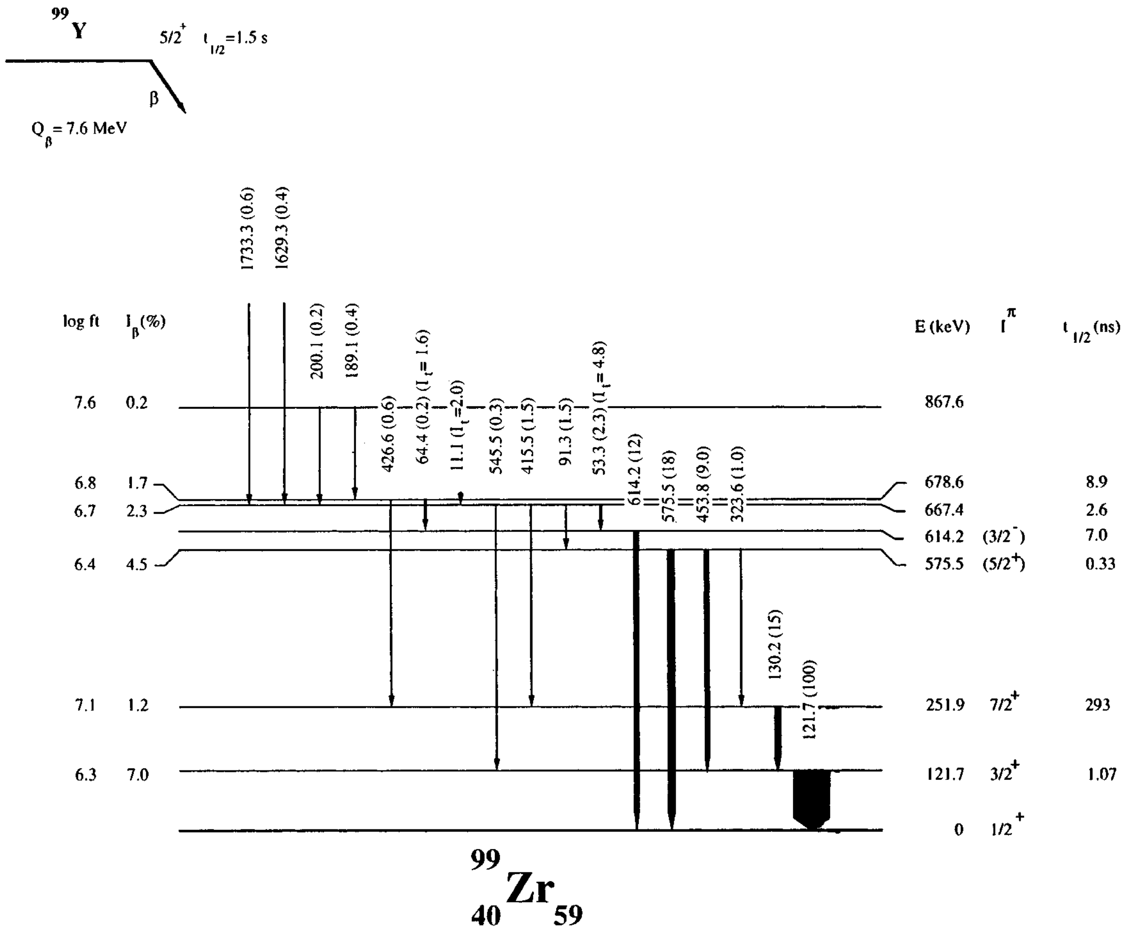

FIG. 2. Partial level scheme of ^{99}Zr. Only the transitions and levels discussed in this work and levels fed in their decay are shown. Not all transitions feeding the 614-keV level are shown, but they are listed in Table I. Transitions depopulating the 614-keV level and the levels below are from [1] and their half-lives from [3,11].

β particles and the 614-keV γ-ray, see Fig. 3, yields $t_{1/2} = 9.7(6)$ ns, in reasonable agreement with the half-life of 8.7(5) ns reported for the 667-keV level [3]. However, this halflife cannot originate from the 667-keV level. The 53- and 91-keV transitions, which directly depopulate this level, have smaller centroid shifts than the 614-keV transition, see the inset in Fig. 3. These shifts correspond to an average $t_{1/2}$ of 4.8(8) ns only, whereas the centroid shift for the 614-keV transition yields a half-life of 10.0(5) ns, in agreement with the above mentioned slope analysis. The time spectrum for the 426.6-keV transition, see Fig. 3, shows that the lifetime has to be attributed to the new 679-keV level. Half-lives of 8.3(15) and 9.5(12) ns are deduced from the slope and the centroid shift, respectively. It can be excluded that this result comes from interference of the 428.5-keV transition from a 9-ns level in ^{98}Y [13] since mass contamination was very weak during this timing measurement. Moreover, there are no delayed transitions close in energy. The time centroid for the neighboring gate, see the inset in Fig. 3, indicates only a short lifetime, assigned to the 429.3-keV transition in ^{99}Nb [10]. In conclusion, we adopt a half-life of 8.9(12) ns for the 679-keV level. The statistically more accurate value deduced from the 614-keV transition is not the result of a single lifetime. First, the centroid shifts of the 53- and 91-keV transitions cannot be accounted solely by the delayed feeding of the 667-keV level via the 11-keV transition, but require $t_{1/2}$=2.6(14) ns for the 667-keV level. Second, according to the intensities listed in Table I, about 50% of the feeding to the 614-keV level by-passes the 667- and 679-keV levels. The absence of a clearly visible prompt component in the time spectrum for the 614-keV transition implies that the 614-keV level itself has a measurable lifetime, a fact that was overlooked in previous reports. Expressing the centroid shift for the 614-keV transition as being due to the lifetimes of the levels at 679-, 667-, and 614-keV, a half-life of 7.0(9) ns is derived. A fit of the slope of the time spectrum with two components, whose half-lives have to be very close to each other, does not significantly improve the fit with a single component.

Nevertheless, a more direct measurement of the 614-keV level lifetime can be obtained from γ-γ-time coincidences recorded with a coaxial Ge detector and the LEPS detector. Although the setup was not optimized for timing, Fig. 4 clearly displays a shift of the time distributions for the 53–614 γ-ray coincidences with respect to the prompt 56–594-keV γ-ray cascade in ^{99}Nb [15]. After small corrections of the centroid shift for the walks in both detectors, a value of $t_{1/2}$=6.9(12) ns is obtained, in excellent agreement with the estimate using the centroid shifts of the β-γ-time spectra.

TABLE I. List of transitions in the decay of ^{99}Y to ^{99}Zr, relevant for the present discussion. Previously reported transitions [1] are listed only if directly related to the levels of interest. Intensities are calculated from the number of coincidence counts with the normalization $I_\gamma(122)=100$.

Energy [keV]		Intensity	Placed from	Placed to	Coincidences
11.1 (2)		[a]	679	667	
45.7 (4)		0.3 (2) [b]	(724	679)	(64), (143), (427)
53.3 (2)	[c]	2.3 (4) [d]	667	614	189, 200, 614,[c] (1629), 1733
64.4 (3)		0.18 (5) [e]	679	614	614 [c]
91.3 (4)	[c]	1.5 (5)	667	576	122,[c] (189), (200), (324 [c]), 454,[c] 576,[c] (1629), (1733)
142.6 (2)		0.20 (6) [b]	(868	724)	(46), (427), 614 [c]
173.3 (3)		0.05 (2)	(788	614)	(614)
189.1 (2)	[f]	0.36 (7)	868	679	53,[c] 91,[c] (130,[c]) (427), 614,[c]
200.1 (2)		0.22 (5)	868	667	(53 [c]), (91 [c]), 614 [c]
237.9 (2)	[c]	0.40 (8)	852	614	614 [c]
391.5 (4)		0.11 (5)	1005	614	(614) [c]
415.5 (3)	[c]	1.5 (4)	667	252	(200), (1733)
426.6 (2)	[f]	0.56 (11)	679	252	122,[c] 130,[c] (189)
614.2 (2)	[c]	12.1 (12) [c]	614	0	53,[c] 64, (143), (173), 189, 200, 238,[c] 392, 831, (1220), (1629), (1733), 1786 [c]
830.8 (4)		0.27 (8)	(1444	614)	(614) [c]
1220.2 (4)		0.37 (11)	1834	614	(614) [c]
1629.3 (5)		0.40 (10)	2296	667	(53 [c]), (91 [c]), (614 [c])
1733.3 (5)		0.61 (15)	2400	667	53,[c] 91,[c] 122,[c] 130,[c] (454,[c]) (576,[c]) 614 [c]
1786.3 (4)	[c]	2.7 (7)	2400	614	614 [c]
1833.4 (8)		0.17 (9)	2448	614	(614) [c]
1869.6 (7)		0.25 (9)	(2484	614)	(614) [c]

[a]Total intensity I_t=2.0 (8) is determined by coincidences of 189-keV line with γ-rays from the 667-keV level.
[b]Intensity calculated assuming the transition directly feeds the 679-keV level.
[c]Transition reported in Ref. [1].
[d]Total intensity I_t=4.8 (6) is determined by coincidences of γ rays feeding the 667-keV level with the 614-keV line.
[e]Total intensity I_t=1.6 (7) is determined by comparison of coincidences of 189 keV and γ rays feeding the 667-keV level with the lines from the 667-keV level and the 614-keV line, respectively.
[f]Transition reported in Ref. [1] but was not placed.

IV. DISCUSSION

First consequences of this work are that the existence of a strongly enhanced $E2$ component in the 53-keV transition is questionable, although not yet ruled out, and that the 87-keV transition proposed to continue the band is not confirmed. The upper conversion coefficient value at 1 standard deviation of $\alpha(53)=1.5$, and the estimate of the 667-keV level half-life yield a reasonable enhancement of about 180 for the $E2$ component in the 53-keV transition. Nevertheless, the fact that higher-spin members of the band are not observed, neither in the prompt-fission experiment of Ref. [6] nor in this work, casts doubts about the interpretation presented in Ref. [1]. The new result of a measurable half-life for the 614-keV level is not inconsistent with its interpretation as the [541]3/2 Nilsson orbital. If we adopt a half-life of 7 ns, the rate of the $E1$ transition to the $1/2^+$ ground state is 2.0×10^{-7} single-particle units. This large hindrance is not exceptional in this region for transitions between deformed and spherical states, see Table III. The other main new result of this work is the level at 679 keV, with $t_{1/2}$=8.9 ns and exhibiting an unusual decay-branching pattern. This level has strong decay branchings by very low-energy transitions of 11 and 64 keV. The 64-keV transition has a partial γ-half-life of 200 ns, which excludes $M2$ and higher multipolarities. It could be an alternative to the 53-keV γ ray as a transition with a fast $E2$ component. The enhancement would be 190 for a $M1$ + 50% $E2$ transition and the corresponding conversion coefficient of about 3 would be consistent with the experimental one. In this interpretation, the very strong decay branching out of the band by the 11-keV transition is, however, difficult to understand. High-energy transitions from the 679-keV level are strongly hindered. The only other decay is to the $7/2^+$ state by the 427-keV transition. While a level with branchings such as those of the 679-keV level is

TABLE II. Total conversion coefficients for the 53- and 64-keV transitions. Theoretical coefficients are calculated from [12].

Energy [keV]	α exp.	α (theoretical) $E1$	$M1$	$E2$	$M2$
53	1.1 (4)	0.71	1.23	10.8	22.8
64	8 (5)	0.41	0.71	5.38	10.9

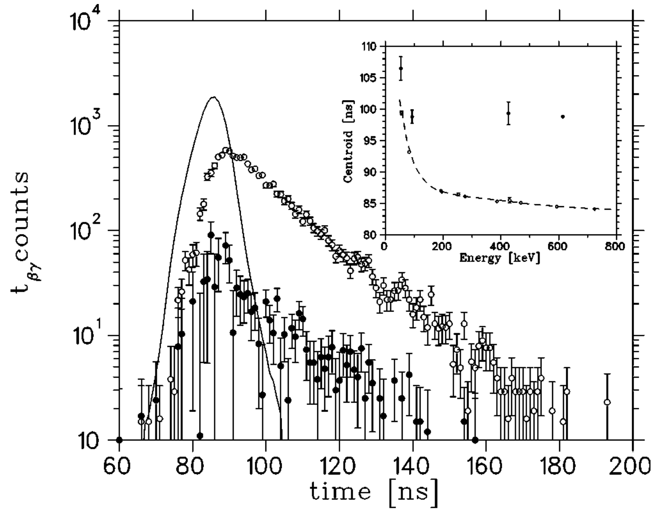

FIG. 3. Time distributions from β-γ coincidences for the 614-keV (open circles) and 427-keV (closed circles) transitions. The solid line is the time spectrum for the 469-keV transition in ^{99}Nb (divided by 20) representative of a prompt distribution. The inset displays the centroids for the lines of 53, 91, 427, and 614 keV (closed circles) discussed in the text. The prompt curve (dashed line) is defined by the lines at 56, 82, 387, 469 keV (^{99}Nb) [15], 192, 194, 276, 724 keV (^{99}Zr) [1], and the 253-keV line (^{99}Mo) [16].

hard to find in ^{97}Sr [4], a similarity can be found with the level at 495 keV ($t_{1/2}$=8.0 μs) in the odd-odd nucleus ^{98}Y, a nearer isotone of ^{99}Zr. This level is the head of a band with $K=2$ or 3, known from experiments at the recoil separator JOSEF of which only preliminary reports are available [17]. The value $K=3$ results from the dipole character of the 121-keV transition to the 4^- level, obtained from its conversion coefficient. However, $K=3$ leads to a moment of inertia larger than the rigid rotor value, and $K=2$ has been favored

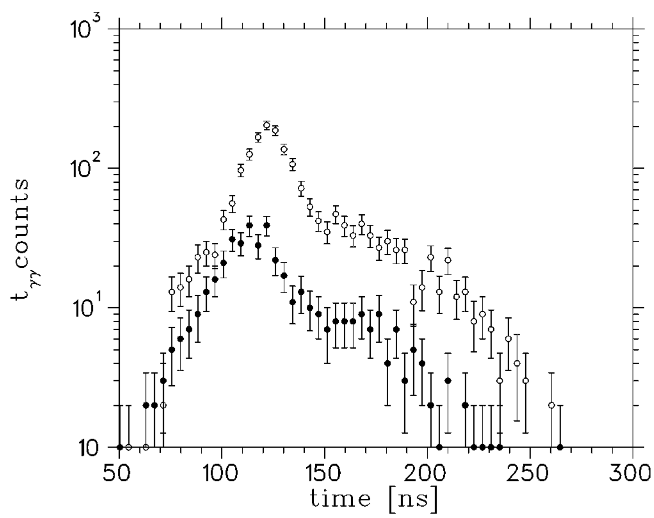

FIG. 4. Time distributions from γ-γ coincidences for the 53-keV (stop)–614-keV (start) cascade (closed circles) and the prompt 56-keV–594-keV cascade (open circles) in ^{99}Nb [15] as a reference. Note that the direction of the physical time is from the right to the left.

TABLE III. Transition rates in Weisskopf units for the decays of the 495- and 679-keV levels in ^{98}Y [17] and ^{99}Zr (this work), respectively, and of the 614-keV level.

Nucleus	Transition [keV]	Transition to	Partial $t_{1/2\gamma}$	$E1$	$M1$	$E2$
^{98}Y	121	(4^-)	9.3 μs	1.9×10^{-8}	1.4×10^{-6}	0.09
	50	(3^-)	242 μs	1.1×10^{-8}	0.7×10^{-6}	0.28
^{99}Zr	427	$7/2^+$	65 ns	6.5×10^{-8}	4.5×10^{-6}	0.02
	64	$(3/2^-)$	203 ns	5.9×10^{-6}	4.2×10^{-4}	94
	614	$1/2^+$	7.0 ns	2.0×10^{-7}	1.4×10^{-5}	0.03

by the authors under the assumption that possibly a very low-energy transition could have been overlooked. The decay of this level is characterized by very large hindrances of transitions to lowest-lying 1^- and 2^- states, which cannot be observed, whereas about 96% of the decay intensity is to the 4^- level at 374 keV and the other 4% is to a 3^- level at 446 keV. This looks similar to the decay of the 679-keV level in ^{99}Zr, where the 427-keV transition to the $7/2^+$ level at 252 keV is observed in this work, but not the branchings to the $1/2^+$ and $3/2^+$ levels. In both cases, large and similar hindrances are deduced, see Table III.

Calculations of the spherical low-lying levels have been made in the frame of the interacting boson-fermion model for ^{97}Sr [4], ^{98}Y [18], and very recently for ^{99}Zr [19]. The odd-neutron levels are the $s_{1/2}$ ground state and the $g_{7/2}$ second excited state. The first excited state is more complex [20]. Spherical levels in ^{98}Y can be regarded as simply coupling a $p_{1/2}$ proton to these neutron levels. In particular, the 4^- and 3^- levels are built on the $\pi p_{1/2}\otimes \nu g_{7/2}$ configuration and do correspond to the $g_{7/2}$ level at 252 keV in ^{99}Zr. Therefore, it is tempting to propose that the 679-keV level is a deformed state and that its structure is the same as the neutron component in the 495-keV level in ^{98}Y. It is impossible to determine the spin and parity of the 679-keV level in ^{99}Zr. Considerations about the 495-keV level in ^{98}Y in order to extract the neutron component are too tentative, due to the uncertain $K=2$ or 3 value and the unknown parity.

V. CONCLUSION

Shape coexistence in ^{99}Zr is expected from systematics and, mainly based on the presence of a strongly enhanced $E2$ component in the 53-keV transition from the 667-keV level, it was proposed that the 614- and 667-keV levels were the lowest levels of a deformed band [1]. In contrast, this work has shown that the previously reported 8.7 ns half-life originates from a combination of the lifetimes of the 614-keV level and of the new level at 679 keV. Nevertheless, a sizeable half-life of about 2.6 ns is still deduced for the 667-keV level, whereas the amplitude of the $E2$ component in the 53-keV transition has to be lowered with respect to previous measurements and could even vanish. Thus, the new results do not rule out the interpretation in Ref. [1], but this is not anymore the only one possible. A better candidate for a deformed state is the new level at 679 keV. Whether it is a bandhead or the $I^\pi=5/2^-$ level of the band built on the [541]3/2 Nilsson orbital at 614 keV, cannot be established.

More detailed experiments using β decay will be unfortunately extremely difficult to perform. However, our interpretation of the 679-keV level could be tested by prompt-fission experiments searching for band structure built on it. The half-life of the level will facilitate this kind of study by allowing suppression of the prompt events.

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