Collective Structure in ^{94}Zr and Subshell Effects in Shape Coexistence

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Based on results from a measurement of weak decay branches observed following the β^- decay of ^{94}Y and on lifetime data from a study of ^{94}Zr by inelastic neutron scattering, collective structure is deduced in the closed-subshell nucleus ^{94}Zr. These results establish shape coexistence in ^{94}Zr. The role of subshells for nuclear collectivity is suggested to be important.

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Nuclei exhibit a wide range of collectivity associated with their finite many-fermion character. This collectivity is dominated by quadrupole deformations, either static (spheroidal or ellipsoidal shapes) or dynamic (quadrupole shape vibrations). The finiteness of nuclei results in (energy) shell structure. Conventionally, nuclear collectivity has been regarded as weak or absent at closed shells and strong far away from closed shells. A few exceptions have been found in the form of shape coexistence (or shape isomerism) [1], i.e., eigenstates of the nucleus with different quadrupole moments. In general, evidence for such structures is indirect, e.g., from the observation of excited rotational energy patterns in nuclei with spherical ground states, where there is a lack of direct evidence of quadrupole deformation, most notably via the observation of enhanced electric quadrupole ($E2$) transition rates. In the current work, we present an example that, more clearly than in any previous case of shape coexistence in the $A = 90$–100 mass region, illustrates how such structures *may have been overlooked* and establishes, for the first time, shape coexistence in the closed-subshell zirconium nuclei through determination of $E2$ transition strengths.

The zirconium isotopes span a range of masses from a mid-open-shell deformed region ($^{80}Zr_{40}$), through a closed neutron shell ($^{90}Zr_{50}$), to a closed neutron subshell ($^{96}Zr_{56}$), and then to a sudden reappearance of deformation ($^{100}Zr_{60}$), which persists to a mid-open-shell region ($^{108}Zr_{68}$). This behavior is unprecedented anywhere on the nuclear mass surface. Of special interest is how collectivity appears and disappears in these isotopes. A key clue is the evidence for the occurrence of shape coexistence in ^{98}Zr [1,2]. Earlier hints of shape coexistence in the zirconium isotopes exist [3,4]; however, these suggestions depended on the *indirect* evidence from rotational band energy patterns [1–3] and electric monopole transition strengths [4–7]. Further, this evidence for shape coexistence was limited to ^{98}Zr [2,5] and (likely) ^{100}Zr [6]. In the present Letter, we combine comprehensive data from an earlier ^{94}Zr$(n, n'\gamma)$ study [8], with new measurements of level lifetimes [9], and a new detailed study of ^{94}Zr from the β^- decay of ^{94}Y to form a consistent picture of shape coexistence in ^{94}Zr based on the *direct* evidence provided by $B(E2)$ values.

The relevant levels for the present discussion are shown in Fig. 1, and their properties are summarized in Table I. The detailed $(n, n'\gamma)$ study [8] was published previously; however, the lifetimes of the 1671-keV 2^+ and the 2330-keV 4^+ levels were recently remeasured and significantly revised [9].

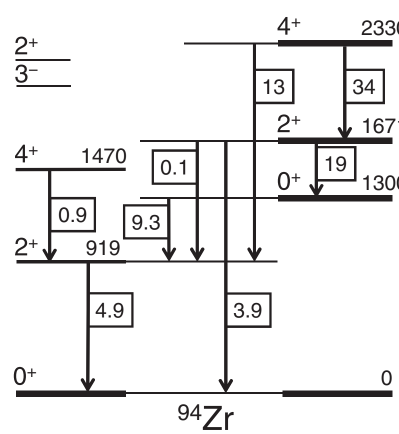

FIG. 1. Levels of ^{94}Zr below 2350 keV, with the band based on the 1300-keV 0^+ state emphasized. The $B(E2)$ values in W.u. are given in boxes.

TABLE I. Excitation energies of levels (E_x), γ-ray energies (E_γ), initial (J_i^π) and final (J_f^π) spins, relative γ-ray intensities (I_γ), level lifetimes, and reduced $E2$ transition probabilities of selected states in ^{94}Zr. The quoted errors in intensities and lifetimes from the present work are statistical only.

E_x[a] (keV)	E_γ[a] (keV)	$J_i^\pi \rightarrow J_f^\pi$	I_γ (%)	τ (ps)	$B(E2)$ (W.u.)
918.82	918.8	$2_1^+ \rightarrow 0_1^+$	100	9.9(21)[b]	4.9(11)
1300.39	381.6	$0_2^+ \rightarrow 2_1^+$	100	420(16)[c]	9.3(4)
1469.70	550.8	$4_1^+ \rightarrow 2_1^+$	100	721(19)[c]	0.880(23)
1671.45	371.1(2)[d]	$2_2^+ \rightarrow 0_2^+$	0.150(6)	$0.368^{+0.027}_{-0.023}$[e]	19(2)
	752.5[f]	$2_2^+ \rightarrow 2_1^+$	41.9(10)		$0.06^{+0.13}_{-0.06}$
	1671.4	$2_2^+ \rightarrow 0_1^+$	57.9(10)		3.9(3)
2329.97	658.5	$4_2^+ \rightarrow 2_2^+$	5.5(3)	$0.42^{+0.20}_{-0.11}$[g]	34^{+10}_{-17}
	1411.1	$4_2^+ \rightarrow 2_1^+$	94.5(3)		13^{+4}_{-7}

[a]From Ref. [8].
[b]From Ref. [10].
[c]From Ref. [11].
[d]From the current work.
[e]From the current work [9]. The lifetime given represents the average of the values determined with the Zr metal and ZrO_2 scattering samples at $E_n = 2.0$ MeV.
[f]$\delta = 0.02(2)$ from Ref. [8].
[g]From the current work [9]. The lifetime was determined with the Zr metallic scattering sample at $E_n = 2.5$ MeV.

These measurements at the University of Kentucky Accelerator Laboratory provide a detailed characterization of the low-energy excited states (excitation energies, spins and parities, decay transition intensities and multipolarities, and level lifetimes); however, it is the lifetime data that are essential to the present Letter, as they are in a lifetime regime associated with a range of level spins and excitation energies that are very difficult to access by other means.

For levels with short lifetimes, the Doppler-shifted γ-ray energy, $E_\gamma(\theta)$, measured at a detector angle of θ with respect to the incident neutrons, can be related to E_0, the energy of the γ ray emitted by a nucleus at rest, by the expression

$$E_\gamma(\theta) = E_0\left[1 + F(\tau)\frac{v_{\mathrm{c.m.}}}{c}\cos\theta\right],$$

where $v_{\mathrm{c.m.}}$ is the velocity of the center of mass in the inelastic neutron scattering collision with the nucleus and c is the speed of light. $F(\tau)$ is the experimental attenuation factor determined from the measured Doppler shift and is compared with calculated attenuation factors to determine the lifetime [12,13]. Data from which the lifetimes of the 1671-keV 2_2^+ and 2330-keV 4_2^+ states in ^{94}Zr were determined are shown in Fig. 2.

As the lifetimes for the 1671- and 2330-keV levels (see Table I) differ from those published previously [8], some comment on these discrepancies is in order. A detailed description of the problems with the measurements of Elhami *et al.* [8] will be published elsewhere, along with the revised level lifetimes [9]; however, we feel confident that these discrepancies can be attributed to uncertainties in the composition of the enriched $^{94}ZrO_2$ scattering sample, which was not completely in the most stable monoclinic crystalline phase, as was assumed in the Doppler-shift attenuation method (DSAM) analysis [12]. This conclusion is supported by subsequent analyses of this material by x-ray powder diffraction and scanning electron microscopy, which revealed a large amorphous component in the enriched sample [9].

In our new DSAM measurements of the lifetimes of levels in ^{94}Zr, samples of metallic zirconium and zirconium (IV) oxide [ZrO_2] of natural isotopic composition were used in $(n, n'\gamma)$ angular distribution measurements at neutron energies of 2.0 and 2.5 MeV and at additional energies of 2.3 and 2.8 MeV for the metal. These energies were chosen to minimize γ-ray feeding of the levels of interest and to limit the complexity of the spectrum (which can experience contributions from each of the five stable isotopes of zirconium in the natural material). An advantage of this approach is that using the natural material provides an "internal calibration" as levels with well-known lifetimes are present in the other Zr isotopes. The best example of such an internal calibration

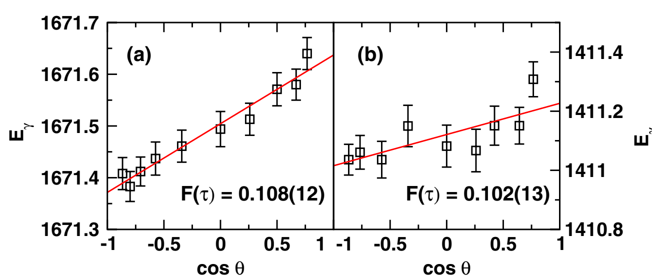

FIG. 2 (color online). (a) Measured energies as a function of $\cos\theta$ for the 1671-keV γ ray from the 1671-keV level in ^{94}Zr at $E_n = 2.0$ MeV for metallic Zr and (b) similar data for the 1411-keV γ ray from the 2330-keV level measured at $E_n = 2.5$ MeV. Linear fits to the data, which yield $F(\tau)$, the attenuation factor, are shown.

is the 2186-keV 2^+ level of ^{90}Zr, which has a well-determined lifetime of 127 ± 4 fs [14] and is in the middle of the DSAM lifetime range. The values of the lifetimes that we obtained for this level were 127^{+10}_{-8} fs with the metallic Zr sample and 122^{+10}_{-9} fs with the oxide sample.

A study of the radioactive decay of ^{94}Y provided the second crucial input to the present report: namely, the identification of a very weak decay branch between the 1671- and 1300-keV levels and quantification of its intensity with high precision. The significance of this observation is that, due to the energy dependence of quadrupole transitions (the $E2$ transition rate is proportional to E_γ^5), high-energy transitions dominate over low-energy transitions from a given excited state. This effect usually obscures the existence of low-energy transitions, even those with large reduced transition strengths. We point to this failure to identify weak, structurally significant decay branches as a widespread problem in nuclear spectroscopy, and it is likely that many excited collective structures have gone unnoticed.

The radioactive sources of ^{94}Y ($T_{1/2} = 18.7$ min, $J^\pi = 2^-$, $Q_\beta = 4.918$ MeV [14,15]) used in the present work were produced by 500-MeV proton-induced fission of a ^{238}UCx target at the TRIUMF Isotope Separator and Accelerator radioactive beam facility. Following mass separation of the fission products, measured yields in the $A = 94$ mass chain at the beginning of the experiment were 2×10^7 s^{-1} (^{94}Rb) and 6×10^7 s^{-1} (^{94}Sr). The $A = 94$ activities were deposited on a moving tape collector at the center of the 8π spectrometer, an array of 20 Compton-suppressed high-purity germanium detectors, and a plastic scintillation detector was close to the deposited activity to detect β^- particles. Typically, counting cycles were 10 min for collection of the radioactivity, followed by 20 min for cooling and 60 min of data collection; i.e., an allowance was made for the decay of the shorter-lived species into ^{94}Y. The data were sorted off-line to create a random-background-subtracted γ-γ coincidence matrix that contained 2×10^8 events.

Figure 3 shows the key result from the present work, i.e., that the excited 2^+ state at 1671 keV in ^{94}Zr decays by a 371-keV transition to the excited 0^+ state at 1300 keV. We determine the decay branch of this transition to be $0.150 \pm 0.006\%$ and deduce the $B(E2; 2_2^+ \rightarrow 0_2^+)$ to be 19 ± 2 W.u. It is important to note that such sensitivity with associated high counting statistics (needed for reasonable precision in the determination of the branching ratio and partial lifetime for the transition) has few precedents. This low-intensity γ ray was not reported in the earlier $(n, n'\gamma)$ study by Elhami *et al.* [8]; however, a reexamination of γ-ray singles data from that study reveals a γ ray at this energy with a branching of $0.12 \pm 0.04\%$. While this value is in agreement with the ^{94}Y decay data, it exhibits greater uncertainty and lacks a coincidence-based placement.

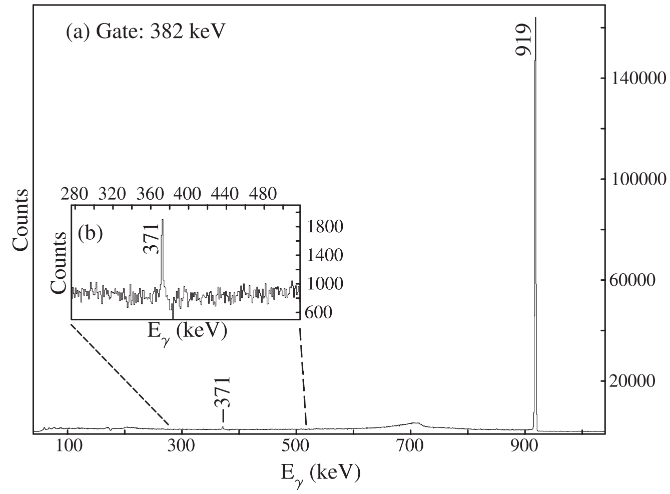

FIG. 3. (a) Portion of the γ-ray spectrum gated on the 382-keV ($0_2^+ \rightarrow 2_1^+$) γ ray in ^{94}Zr following the β^- decay of ^{94}Y. (b) Confirmation for the placement of the deexciting 371-keV $2_2^+ \rightarrow 0_2^+ \gamma$ ray is evident.

It is expected that the deformed band (shown in Fig. 1) continues to higher spins. Evidence for the 6^+ member of the band at 3142 keV has been obtained from the study of γ rays from fission fragments following heavy-ion fusion reactions [16,17]: in addition to an 812-keV γ ray to the 2330-keV 4^+ state, it decays to the 1470-keV 4^+ state and to a 5^- state at 2606 keV. However, in neither of these studies was a firm spin-parity assignment possible. A search of our high-statistics γ-γ coincidence data from ^{94}Y β^- decay for an indication of this level yields a weak peak at 812 keV in the gate on the 1411-keV $4_2^+ \rightarrow 2_1^+$ transition, but no additional information is available, so the 3142-keV level remains a tentative band member.

The interpretation of the 1671-keV 2_2^+ state as a member of the coexisting structure appears unequivocal from the data presented here. It exhibits a much larger quadrupole deformation parameter, β, of 0.18, determined from the $B(E2)$'s [10], than the first excited state, which has $\beta = 0.09$. Moreover, this interpretation is consistent with the large, positive g factor of the 2_2^+ state [18], suggesting proton dominance in this state, and with the strong population of the 1300- and 1671-keV states in the ^{94}Mo(^{14}C, ^{16}O) and ^{94}Mo(^{6}Li, ^{8}B) reactions [19,20] that indicate an underlying proton-pair excitation across a $Z = 40$ subshell gap. Moreover, recent large-valence-space shell model calculations indicate that the first excited 0^+ state of ^{94}Zr corresponds to a two-particle two-hole proton excitation from the $p_{1/2}$ and $p_{3/2}$ orbitals to the $g_{9/2}$ orbital [21].

The present results establish an excited collective structure in ^{94}Zr. This recognition is an important step toward developing a systematic view of shape coexistence in the zirconium isotopes. The possible occurrence of shape coexistence in ^{98}Zr has been argued to involve rotationally induced deformation [22], and the mass region centered on these neutron-rich zirconium isotopes has also been suggested as an example of a nuclear quantum phase transition [23] (which depends on a "critical point" in the nucleon number). Such ideas imply that nuclei in this region are "soft." The present work supports an opposing view: namely, that deformation is present at the lowest spin (i.e., it is not rotationally induced) and that shape coexistence occurs widely in this region (i.e., there is no critical point in the nucleon number). Thus, we conclude that changes across the region result from changed ordering, by excitation energy, of spherical and deformed states.

As noted previously and presented in reviews [1,3], shape coexistence is well established for nuclei at singly and doubly closed shells but it has not been established for subshells. The current situation for subshells near ^{98}Zr is summarized in Fig. 27 of Ref. [1], but these arguments rely on indirect evidence from rotational band energy patterns and $E0$ transition strengths. The conclusions reached here are based on *electric quadrupole transition probabilities* that confirm the existence of spherical and deformed states in ^{94}Zr, a nucleus where the preponderance of data would lead one to infer that it should not occur.

This new result for ^{94}Zr and the previous data for ^{98}Zr have wider implications for this mass region. A question, which has been discussed previously [24,25], immediately arises: Do such structures occur in ^{96}Zr? There is an indication of such a structure from the ^{100}Mo(d, ^{6}Li)^{96}Zr reaction [26] (and see Ref. [1]), but critical transition rate data for ^{96}Zr are not available. Other nuclei in this mass region that are suggested to display shape coexistence include ^{96}Sr [1,27], ^{97}Sr [1,2], ^{98}Sr [6], and ^{99}Zr [1,2], but $E2$ transition rate data are lacking in all cases.

From the new results presented here, we conclude that shape coexistence can be expected to occur far more widely than previously believed because subshell structure will occur more widely than shell structure. The criterion for the occurrence of shape coexistence is the existence of sufficiently large energy gaps between subshells; closely spaced subshells lose their individuality due to pairing correlations and behave as a single, larger subshell. This perspective, introduced in Ref. [1] and now supported by evidence of quadrupole collectivity in ^{94}Zr, i.e., at the $Z = 40$ subshell, offers a broad prospect for the experimental investigation of excited 0^+ states and their associated collectivity in nuclei. Indeed, it is now clear that our view of the effects of subshell structure on nuclear collectivity requires reassessment.

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[1] K. Heyde and J. L. Wood, Rev. Mod. Phys. **83**, 1467 (2011).
[2] C. Y. Wu, H. Hua, D. Cline, A. B. Hayes, R. Teng, R. M. Clark, P. Fallon, A. Goergen, A. O. Macchiavelli, and K. Vetter, Phys. Rev. C **70**, 064312 (2004).
[3] J. L. Wood, K. Heyde, W. Nazarewicz, M. Huyse, and P. van Duppen, Phys. Rep. **215**, 101 (1992).
[4] J. L. Wood, E. F. Zganjar, C. De Coster, and K. Heyde, Nucl. Phys. **A651**, 323 (1999).
[5] H. Mach, M. Moszyński, R. L. Gill, F. K. Wohn, J. A. Winger, J. C. Hill, G. Molnár, and K. Sistemich, Phys. Lett. B **230**, 21 (1989).
[6] C. Y. Wu, H. Hua, and D. Cline, Phys. Rev. C **68**, 034322 (2003).
[7] G. Lhersonneau *et al.*, Phys. Rev. C **49**, 1379 (1994).
[8] E. Elhami *et al.*, Phys. Rev. C **78**, 064303 (2008); (to be published).
[9] E. E. Peters *et al.* (to be published).
[10] S. Raman, C. W. Nestor, Jr., and P. Tikkanen, At. Data Nucl. Data Tables **78**, 1 (2001).
[11] H. Mach, E. K. Warburton, W. Krips, R. L. Gill, and M. Moszyński, Phys. Rev. C **42**, 568 (1990).
[12] T. Belgya, G. Molnár, and S. W. Yates, Nucl. Phys. **A607**, 43 (1996).
[13] P. E. Garrett, N. Warr, and S. W. Yates, J. Res. Natl. Inst. Stand. Technol. **105**, 141 (2000).
[14] D. Abriola and A. A. Sonzogni, Nucl. Data Sheets **107**, 2423 (2006).
[15] G. Audi, A. H. Wapstra, C. Thibault, J. Blachot, and O. Bersillon, Nucl. Phys. **A729**, 3 (2003).
[16] N. Fotiades *et al.*, Phys. Rev. C **65**, 044303 (2002).
[17] D. Pantelica *et al.*, Phys. Rev. C **72**, 024304 (2005).
[18] V. Werner *et al.*, Phys. Rev. C **78**, 031301(R) (2008).
[19] W. Mayer, D. Pereira, K. E. Rehm, H. J. Scheerer, H. J. Körner, G. Korschinek, W. Mayer, and P. Sperr, Phys. Rev. C **26**, 500 (1982).
[20] R. S. Tickle, W. S. Gray, and R. D. Bent, Nucl. Phys. **A376**, 309 (1982).
[21] K. Sieja, F. Nowacki, K. Langanke, and G. Martínez-Pinedo, Phys. Rev. C **79**, 064310 (2009).
[22] W. Urban *et al.*, Nucl. Phys. **A689**, 605 (2001).
[23] S. Naimi *et al.*, Phys. Rev. Lett. **105**, 032502 (2010).
[24] G. Molnár, S. W. Yates, and R. A. Meyer, Phys. Rev. C **33**, 1843 (1986).
[25] R. A. Meyer, E. A. Henry, L. G. Mann, and K. Heyde, Phys. Lett. B **177**, 271 (1986); Hyperfine Interact. **22**, 385 (1985).
[26] A. Saha, G. D. Jones, L. W. Put, and R. H. Siemssen, Phys. Lett. **82B**, 208 (1979).
[27] K. Becker, G. Jung, K.-H. Kobras, H. Wollnik, and B. Pfeiffer, Z. Phys. A **319**, 193 (1984).