The shape of the $T_z = +1$ nucleus $^{94}$Pd and the role of proton-neutron interactions on the structure of its excited states

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1. Introduction

The $N = Z = 50$ $^{100}$Sn is the heaviest self-conjugate doubly-magic nucleus that is stable with respect to particle emission. Nuclear structure of hole states in the region “south-west” of the shell closure between the $N = 50$, $Z = 40$ and the $N = Z$ lines is dominated by the $0_{9/2}^+$ intruder orbital from the $N = 4$ harmonic oscillator shell. It is well separated from the $N = 3$ $pf$-shell orbitals, both energetically and by its parity, contributing only with even-parity even-hole excitations into the intruder orbital. This makes the region “south-west” of $^{100}$Sn the subject of increased focus for both experimental and theoretical investigation [1,2]. In particular, $0_{9/2}^+$ is the first valence high-spin orbit, where seniority breaking is discussed extensively in the literature. [3–9]. In addition, the strong spatial overlap of proton- and neutron-hole wave functions causes strong proton-neutron ($pn$) interaction gives rise to unique structural features such as spin-gap isomers and seniority induced symmetries. Remnants of the seniority level scheme in the open $πν(0_{9/2}^+)$ orbitals have also been addressed in reference [10].

Experimental work directly related to this topic includes the yrast spectroscopy of $^{92}$Pd [11] and the decay of the $I^\pi = 16^+$ spin trap isomer and yrast sequence in $^{96}$Cd [12–14]. The strength of the $pn$ interaction in the $πν(0_{9/2}^+)$ orbitals manifests itself best in the strongly-binding $T = 0$ ($0_{9/2}^+$), $I^\pi = 9^+$ isoscalar two-body matrix element (TBME), which is comparable with the $T = 1$ isovector pairing, as introduced in early works [15–18] using empirical interactions employing the $πν(1_{12}^+,0_{9/2}^+)$ model space. They are reviewed in Ref. [1] together with calculations in the full $πν(1_{12}^+,0_{9/2}^+)$ model space using empirical [19] and realistic [20] interactions. Subsequently, Large Scale Shell Model (LSSM) calculations were presented for the upper $πν(0_{9/2}^+)$ shell using the Nowacki-Sieja interaction in Ref. [12].

Following the discovery of excited states in $^{92}$Pd [11], a series of multi-step shell-model and Interacting Boson Model (IBM) studies investigated the role of $πν(0_{9/2}^+)$ proton-neutron pairs with maximum aligned spins of $9^+$ in the $N = Z$ nuclei $^{96}$Cd, $^{94}$Ag and $^{92}$Pd with particular interest on the dependence of the controlling $9^+$-TBME [11,21–23,5]. The content of the various $pn$-pairs within the nuclear wave functions in the three nuclei with increasing spin was discussed. However, overlap of the aligned $9^+$ $pn$-boson wave functions with the exact shell-model diagonalization could only be established for low- and high-spin states, and little overlap was found for intermediate spin [5]. These conclusions are subject to modifications when excitations in the full $πν(1_{12}^+,0_{9/2}^+)$ and $πν(0_{9/2}^+)$ space are considered in the LSSM calculations as presented in this work.

In Ref. [24], predictions in these model spaces were compared with a pure $0_{9/2}^+$ approach for $B(E2)$ values and spectroscopic quadrupole moments in $^{92}$Pd and $^{96}$Cd. In the low-spin range ($J \leq 6$), the three approaches are equivalent for excitation energy and $B(E2)$ values, but exhibit large differences in the (presently experimentally inaccessible) spectroscopic quadrupole moments. Moreover, the lower-$Z$ nuclei in the $0_{9/2}^+$ orbital exhibit signs of significant quadrupole deformation [25]. This is expected to evolve for higher spins and for nuclei closer to the $N = Z = 50$ doubly-magic closure due to model space exhaustion, resulting in a gradual reduction in collectivity.

The $T_c = +1$ nucleus $^{94}$Pd, with its 2 neutron and 4 proton holes in $0_{9/2}^+$ orbital below $^{100}$Sn, is situated at a crucial point of this evolution. It is the neighbour of the even-even $N = Z$ systems $^{92}$Pd and $^{96}$Cd, and represents the $T = 1$ isospin partner for states in the odd-odd $N = Z$ system $^{94}$Ag. In particular, the detailed structure of the $8^+$ seniority remnant state in $^{94}$Pd will reveal the interplay between the isovector and isoscalar coupling of the $pn$ pairs. Moreover, the structure of $^{94}$Pd in terms of seniority-mixed states may provide a first indication of emerging collectivity when neutrons are removed from the doubly-magic system $^{100}$Sn. The emergence of deformation is also supported by the prediction that favoured $pn$ transition $T = 0$ pairs arrange themselves in a spin-aligned configuration to form shears blades in the Anti-Magnetic Rotational (AMR) behaviour for the yrast band of $^{92}$Pd [26]. A recent theoretical publication using the EXVAM (Excited VAMPiR) approach [27] notes the relation of $T = 0$ $pn$-pairing component to the emergence of prolate deformation and shape coexistence in $^{94}$Pd.

The experimental information on excited states in $^{94}$Pd is presently available up to spin-parity $I^\pi = (20^+)$ and originates from experiments in which decays of the isomeric states with spin-parity $I^\pi = 14^+$ and $(19^+)$ were studied [28–30], and from high-spin $β$-decay studies of $^{94}$Ag [31,32]. Only states fed by delayed transitions are known and no prompt $γ$-ray radiation from states in $^{94}$Pd has so far been observed. This letter presents results on electromagnetic transition rates between yrast states in $^{94}$Pd. This allows a direct comparison between the predictions of various approaches of shell-model interactions and valence spaces. Special interest is put on $pn$ interaction treatment for this $T_c = +1$ nucleus intermediate between the $N = Z$ line and the $N = 50$ closed neutron shell.

2. Experimental details

The decay of the isomeric, yrast $I^\pi = 14^+$ state in $^{94}$Pd [30] was studied through its production via the projectile fragmentation of a $^{124}$Xe primary beam at 982 MeV/u from the SIS18 synchrotron at GSI Helmholtzzentrum für Schwerionenforschung accelerator facility, Darmstadt, Germany. The secondary cocktail beam, resulting from reactions between the primary beam and a 4 g/cm² thick Be target, was separated in terms of mass-to-charge ratio ($A/Q$) and atomic number ($Z$) in the FReagment Separator (FRS) [33]. The fragmentation products were identified on an event-by-event basis using the standard $Bγ - Eγ - Bp$ and $ToF - Bp - ΔE$ identification methods [34]. The ions reaching...
the final focal plane of the FRS were implanted in the Advanced Implantation Detector Array (AIDA) [35] in the center of the DEcAY SPEctrosopy (DESPEC) setup [36]. The $\gamma$ rays emitted in the deexcitation of the $^{14}$+ isomeric state ($T_{1/2} = 515(1)$ ns) in $^{94}$Pd were registered using 6 triple-cluster High Purity Germanium (HPGe) detectors (GALILEO) [37,38] and 36 LaBr$_3$(Ce) detectors, constituting the Fast TIMing Array (FATIMA) [39,40]. Each detector subsystem was equipped with an independent data acquisition system. The synchronization of the different subsystems was achieved using White Rabbit (WR) time stamp [41], which is driven by a 125 MHz clock with time accuracy of up to $\sim$ 1 ns. A preliminary analysis of these data on excited states transition rates in $^{96}$Pd has been reported by the collaboration [42].

3. Data analysis and results

To extract nuclear excited-state mean lifetimes, the energy and timing data recorded by the FATIMA array were used to construct $E_{\gamma} - E_{\gamma} - \Delta T_{\gamma 1}$ coincidence cubes, where a delayed coincidence with implanted $^{94}$Pd ions was applied. The delayed time distribution was obtained under the condition that the feeding transition provides the start signal and the decay - the stop signal. The $\gamma$-ray spectrum obtained as total projection of the matrix is shown in Fig. 1(a) along with a resulting coincidence spectrum with the 1092-keV $\gamma$ ray in Fig. 1(b). A time alignment was performed for all FATIMA detectors using coincidences between the 344- and 779-keV transitions from $^{152}$Eu source data. The centroid of the delayed time distribution [43-45]

$$C(D) = \frac{\int_{-\infty}^{\infty} D(t)dt}{\int_{-\infty}^{\infty} D(t)dt},$$  (1)

where $D(t)$ is the measured time distribution, was calculated for each detector pair. The centroid of the anti-delayed time distribution was obtained in an analogous way, where in contrast to the delayed distribution the feeding transition provides the stop signal and the decay - the start signal. The generalized centroid difference ($\Delta C$) was obtained by subtracting the two centroids. In the Generalized Centroid Difference (GCD) method [44,45] $\Delta C$ is directly related to the mean lifetime $\tau$ according to the expression:

$$\Delta C(E_{\gamma}) = PRD(E_{\gamma}) + 2\tau,$$  (2)

where $PRD(E_{\gamma}) = PRD(E_{\text{feeder}}) - PRD(E_{\text{decay}})$ is the Prompt Response Difference and the symmetry condition with respect to feeder-decay inversion [43] is:

$$\Delta C(E_{\gamma})_{\text{decay}} = -\Delta C(\Delta E_{\gamma})_{\text{feder}},$$

$$PRD(E_{\gamma})_{\text{decay}} = -PRD(E_{\gamma})_{\text{feder}}.$$  (3)

Here $\Delta E_{\gamma} = E_{\text{feder}} - E_{\text{decay}}$ is the energy difference between the feeding and decaying $\gamma$ rays. The PRD is energy dependent and was calibrated using various coincident transitions from $^{152}$Eu source data. The values were adjusted to the 344-keV reference energy and fitted using the equation [44]:

$$PRD(E_{\gamma}) = \frac{a}{\sqrt{E_{\gamma} + b}} + cE_{\gamma} + d,$$  (4)

where $a$, $b$, $c$, $d$ are the parameters for the fit presented in Fig. 1(c). The fit residuals in Fig. 1(d) allow the systematic error of the PRD to be evaluated.

This analysis method is sufficiently accurate to measure excited-state half-lives in the range from of tens of picoseconds to nanoseconds, therefore a careful background treatment is essential. To minimize the influence of the Compton background beneath the full-energy peaks (FEP), the experimental centroid difference $\Delta C_{\text{exp}}$ was corrected using [46]:

$$\Delta C_{\text{FEP}} = \Delta C_{\text{exp}} + \frac{t_{\text{corr}}(\text{decay}) + t_{\text{corr}}(\text{feder})}{2}$$  (5)
4. Discussion

The experimental results presented in this work are discussed within the shell-model framework. In Fig. 3 the experimentally-established level energies together with the known γ rays are shown in comparison to the two most advanced shell-model calculations in the full diagonalization of the nuclear Hamiltonian. The first one uses the JUN45 interaction [47] in the full πv(f1/2p3/2) model space, while the second one is a LSSM calculation employing the GDS interaction [12] with πv(gs) as the model space. Both calculations reproduce the experimental yrast level energies very well up to the highest known spins.

In order to access the structure of involved states, and the associated nuclear deformation using the πv(gs) valence space and effective GDS Hamiltonian, the potential energy surface (PES) of 92,94,96Pd were obtained from Discrete Nonorthogonal Shell Model (DNO-SM) calculations in the same way as introduced in Ref. [48,49]. As shown in Fig. 4, 94Pd exhibits a non-spherical shallow minimum at moderate prolate deformation. The ground-state wave function contains dominant contributions around β ~ 0.1 – 0.2 to high spins for the yrast and yarate states with no other coexisting minimum found in the PES. This is at variance with the claim made in Ref. [27]. The predicted (β, γ) distributions in the wave functions evolve from a spherical regime in 96Pd towards a more axially-deformed prolate shape in the N = Z system 92Pd (see Fig. 4), with T = 1+ 94Pd being the transitional nucleus between these two extremes. This trend is particularly noticeable in the I = 0+ ground states. For the I = 8+ state in 96Pd the shape remains spherical, whereas the deformation pattern in the two other nuclei shifts towards sphericity and maintains as such up to higher spins, in particular for the 14+ state in 94Pd. It should be noted that in 92Pd, where the development of an axial prolate shape in the ground state is the most pronounced, there is no indication of other shape-coexisting minima within the configuration space.

The experimentally-obtained half-lives for the 6+ and 8+ states in 94Pd from the current work were used to determine reduced E2 transition strengths. The deduced B(E2) values, together with the value for the decay of the I = 14+ isomeric state reported in Ref. [50], are summarized and compared to the two aforementioned shell-model approaches (JUN45, GDS) in Table 1 and Fig. 5. The values from Ref. [27] are provided in the table for a cross comparison. Effective charges of e1 = 1.5e and e2 = 1.1e according to Ref. [47] were used for the JUN45 interaction [47] as determined by the least-squares fit to the experimental data. This well-known phenomenologically-tuned realistic interaction, which has reproduced many nuclear properties from the N = 3 harmonic oscillator shell and the region of 100Sn, is based on the Bonn-C potential. The calculated B(E2) values are over-estimated when compared to the experimental data (see Fig. 5) and do not allow a simultaneous reproduction of the 8+ and the 14+ states in 94Pd within the experimental uncertainties for any charge state combination. This is most probably a consequence of the strong mixing of the upper fp shell with the f7/2 orbital, characteristic for this interaction and required for lighter nuclei to substitute the missing f5/2 orbital in the corresponding model space.

The agreement between the experimental results and the most challenging LSSM calculation, which employs the GDS interaction [12] with effective charges of e1 = 1.1e and e2 = 0.84e, extracted from 100Sn and 96Cd [51], is excellent. The involvement of core excitations (up to SpSh) in the πv(gs) model space, exhibits an almost exact reproduction of high-spin states (see Fig. 3) as well as of the reduced transition rates (see Fig. 5).

On the other hand, the AMR calculations shown in Fig. 5 (denoted by solid line) reproduce the transition rates measured in the current work very well. Ref. [26] demonstrates a good reproduction of the energy levels in 94Pd using the AMR coupling scheme. For 94Pd, the calculation is based on a similar 4 quasiparticle configuration as for the ground state of 96Pd, where the sheets closing behaviour takes over beyond I = 8+. This may indicate that the T = 1 proton-proton and neutron-neutron pairs in the f5/2, f7/2 orbitals rearrange themselves to form two oppositely aligned T = 0 proton blades, the closing mechanism of which takes over in generating the higher-spin states of 94Pd and continues until the sheets blades are maximally aligned at I = 16+. This supports the
dominance of the isoscalar ($T = 0$) phase beyond $I^f = 8^+$ states. It is worth noting that the 4 quasiparticle AMR configuration for the spin states $I^f < 8^+$ (denoted by dotted line in Fig. 5) is expected to mix with those of 2 quasiparticle one.

With the aim of examining further the interplay of the isoscalar ($T = 0$) versus isovector ($T = 1$) components of the $pn$ shell-model interaction on the structure of $^{94}$Pd, the excited-state lifetimes were analysed within the single-$0g_{9/2}$ model. Although shell-model results presented in the current work indicate that a multi-orbital space including core shell $N, Z = 50$ excitations are needed to describe $^{94}$Pd quantitatively, the restriction to this rather simple model is justified by the spherical or slightly-deformed nature of Pd nuclei evidenced by these results as well as by the prominent role played by the $0g_{9/2}$ orbital in the low-lying states of nuclei around $N, Z = 50$ [12,11,21–23]. Indeed, the wavefunction overlap of all $^{94}$Pd states with the ($0g_{9/2})^{-4} \otimes (v0g_{9/2})^{-2}$ configuration, as calculated within the LSSM approach, exceeds 95%. Therefore, based on this overwhelming dominance, the calculations in the single-$0g_{9/2}$ model were performed by using the two-body effective interaction derived within the framework of the many-body perturbation theory starting from the high-precision CD-Bonn $NN$ potential [52]. Details of the calculation are described in Ref. [53]. In addition, two subsets of interactions were obtained by separately removing the $T = 0$ and $T = 1$ $pn$ matrix elements.

Similar to the $N = Z$ case of $^{92}$Pd discussed in [53], the energies of the yrast levels of $^{94}$Pd are reasonably-well reproduced when using the full interaction. A spectrum with the same structure is obtained only if the pure $T = 0$ $pn$ component is considered, while the inclusion of only the $T = 1$ component leads to excited states compressed in a smaller energy interval, although the effect is smaller than the $N = Z$ system $^{92}$Pd. These findings are in line with those reported in Ref. [11,1], indicating that the evolution from the seniority to vibrational-type spectrum from $^{96}$Pd to $^{92}$Pd, with $^{90}$Pd exhibiting an intermediate character is related to the $T = 0$ $pn$ interaction.

This is the first time when such an analysis has been performed for $B(E2)$ transition strengths. Considering the significant restrictions of using a single-$j$ space, this analysis is not intended to reproduce the experimental values, but only to investigate the relevance of the isovector with respect the isoscalar component of the $pn$ interaction. The same effective charges of $e_p = 1.5e$ and $e_n = 1.1e$ as for the JUN45 calculation, were used. The $B(E2)$ corresponding to the full interaction and to the pure $T = 0/1$ component are reported in Table 1, while in Fig. 5 the results of the full interaction are compared with the experimental and other shell-model values. The overall behaviour is similar to that predicted by the JUN45 as well as the LSSM (GDS) approaches. However, the precision of the LSSM calculation when compared to the experimental values provides the best match, indicating again the relevance of core excitations. Furthermore, as shown in Table 1, the exclusion of the $T = 1pn$ component from the single-$j$ interaction does not change significantly the calculated $B(E2)$ values with respect to the full interaction for the decay of the yrast $I_g = 6^+$, $8^+$ and $14^+$ states. In contrast, when considering a pure $T = 1$ force, considerably longer predicted half-lives are obtained. This finding demonstrates the effect of the different structure of the wave functions resulting from the $T = 1$ force, which is in general unable to produce a sufficient fragmentation of the basis states arising from the $0g_{9/2})^{-4} \otimes (v0g_{9/2})^{-2}$ configuration.

5. Conclusion

The half-life and transitions rates for decays of intermediate-spin states in $^{90}$Pd have been established using the FATIMA array. A range of restricted-basis model spaces and interactions were used to reproduce level energies and experimentally deduce $B(E2)$ values. The LSSM approach with a $xv(gds)$ model space provides the best agreement with the experimental results. This model indicates no evidence of deformation for Pd isotopes, which is predicted at the $N = Z$ line following a parallel potential energy surface analysis. Based on this conclusion, the $T = 0$ contribution of the $pn$ interaction is manifest as the dominant one in the transition strengths of the $8^+$ seniority remnant state and the $14^+$ isomeric state.

### Table 1

<table>
<thead>
<tr>
<th>Quantity [ms/e^2 fm^4]</th>
<th>$I^f = 14^+$</th>
<th>$I^f = 12^+$</th>
<th>$I^f = 10^+$</th>
<th>$I^f = 8^+$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T = 0$ Pimeter</td>
<td>515(1)</td>
<td>0.755(100)</td>
<td>≤0.04</td>
<td></td>
</tr>
<tr>
<td>$T = 0$ BGEvRH(E2)</td>
<td>52.1(1)</td>
<td>205(15)</td>
<td>113</td>
<td></td>
</tr>
<tr>
<td>$B_{K(E2)}(E2)$</td>
<td>101</td>
<td>252</td>
<td>453</td>
<td></td>
</tr>
<tr>
<td>$B_{K(E2)}(E2)$</td>
<td>49</td>
<td>192</td>
<td>548</td>
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<td>$B_{K(E2)}(E2)$</td>
<td>112</td>
<td>144</td>
<td>398</td>
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</tr>
<tr>
<td>$B_{K(E2)}(E2)$</td>
<td>82</td>
<td>191</td>
<td>398</td>
<td></td>
</tr>
<tr>
<td>$B_{K(E2)}(E2)$</td>
<td>9</td>
<td>11</td>
<td>5</td>
<td></td>
</tr>
<tr>
<td>$B_{K(E2)}(E2)$</td>
<td>56</td>
<td>165</td>
<td>336</td>
<td></td>
</tr>
</tbody>
</table>

![Fig. 4](Image) Potential energy surface (PES) plots for $^{96,92}$Pd nuclei for the ground state as well as for the first $I^f = 8^+$ states. Additionally, the PES for the $I^f = 14^+$ state in $^{94}$Pd is shown indicating a shallow prolate minimum (see text for details).

![Fig. 5](Image) Experimental and shell-model calculated $B(E2)/2$ values, using different effective interactions and model spaces, for states in $^{94}$Pd (see text for details).
Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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