

# Neutron-Proton Pairing Competition in $N = Z$ Nuclei: Metastable State Decays in the Proton Dripline Nuclei $^{82}_{41}\text{Nb}$ and $^{86}_{43}\text{Tc}$

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The low-lying structures of the self-conjugate ( $N = Z$ ) nuclei  $^{82}_{41}\text{Nb}$  and  $^{86}_{43}\text{Tc}$  have been investigated using isomeric-decay spectroscopy following the projectile fragmentation of a  $^{107}\text{Ag}$  beam. These represent the heaviest odd-odd  $N = Z$  nuclei in which internal decays have been identified to date. The resulting level schemes shed light on the shape evolution along the  $N = Z$  line between the doubly-magic systems  $^{58}_{28}\text{Ni}$  and  $^{100}_{50}\text{Sn}$  and support a preference for  $T = 1$  states in  $T_z = 0$  odd-odd nuclei at low excitation energies associated with a  $T = 1$  neutron-proton pairing gap. Comparison with Projected Shell Model calculations suggests that the decay in  $^{82}\text{Nb}$  may be interpreted as an isospin-changing  $K$  isomer.

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Pairing correlations are a fundamental aspect in describing many areas of physical structure, for example BCS pairs in superconducting metals [1]. The nucleus is a unique system in this respect as three pairing modes are present; neutron-neutron ( $nn$ ), proton-proton ( $pp$ ) and neutron-proton ( $np$ ). The  $np$  pairing mode represents a two-fluid system in which the components can be coupled (or "paired") together in two ways, (i) with the intrinsic spins,  $s$ , parallel ( $T = 0$ ), or (ii) anti-parallel ( $T = 1$ ), where  $T$  is the isospin quantum number [2].

Structurally related states appear in nuclei of the same mass number,  $A$ , (but different numbers of protons,  $Z$ , and neutrons,  $N$ ) when a  $np$  pair is exchanged with a

$nn$  or  $pp$  pair. The appearance of these isospin multiplets is a manifestation of the charge independence of the strong nuclear force, a cornerstone of our current understanding of nuclear structure. States with the same isospin quantum number form analogous spectral patterns in these isobaric chains, with individual nuclear species characterised by their isospin projection quantum number  $T_z = \frac{N-Z}{2}$ . In most nuclei the states with the lowest energy have isospin values  $T = T_z$ . However this pattern can be modified in nuclei with equal numbers of protons and neutrons. It is only in  $N \approx Z$  nuclei where the proton and neutron states at the Fermi surfaces have a sufficient spatial overlap that protons and

neutrons in equivalent orbitals can couple (i.e.,  $np$  pairs) to form both  $T=1$  and  $T=0$  states of similar energies [3]. There has been longstanding interest in the structure of medium mass even-even  $N = Z$  nuclei with the aim of determining the magnitude of the  $T=1$ ,  $np$  pairing strength through the observation of ‘delayed alignments’ at higher spins in such systems [4, 5]. Self-conjugate nuclei represent a unique laboratory in which the direct competition between the  $T = 1$  and  $T = 0$  neutron-proton pairing mode can be investigated.

The ground state  $\beta$ -decay half lives of all odd-odd  $N = Z$  nuclei up to  ${}^{98}_{49}\text{In}$  have been reported [6, 7]. All those between  ${}^{34}_{17}\text{Cl}$  and  ${}^{98}_{49}\text{In}$ , except  ${}^{58}_{29}\text{Cu}$ , are consistent with superallowed Fermi  $\beta$ -decay, indicating  $T=1$  ground states with spin/parity,  $I^\pi = 0^+$ . Excited states in odd-odd  $N = Z$ ,  $fpg$  shell nuclei up to  ${}^{78}_{39}\text{Y}$  have also been identified using heavy-ion fusion-evaporation reactions and charged-particle detectors [8–15]. In each case excited states have been identified and assigned as Isobaric Analogue States of the  $T=1$ ,  $I^\pi = 2^+$  and  $4^+$  states in the  $T_z = 1$  isobar providing evidence for a  $T = 1$ ,  $np$  pairing condensate. Odd-spin states at low excitation energy have also been observed and interpreted as  $T=0$  states.

Deformation plays a key role in the structure of self-conjugate nuclei due to the coinciding low level densities in both the proton and neutron nuclear potential. Stable ground-state deformation has been shown to exist along the  $N = Z$  line with the maximum at  ${}^{76}_{38}\text{Sr}$  [16]. A swift change from this deformed nuclear potential to a near-spherical shape is predicted to lie just above the  $Z = 40$  shell gap. The  $Z = 41$  nuclei are suggested to represent this boundary [17] and therefore it is expected that heavier systems will have softer, less-deformed shapes.

With the exception of  ${}^{78}\text{Y}$  ( ${}^{76,77}\text{Y}$  are particle bound), the odd-odd  $N = Z$  nuclei with  $A \geq 70$  also lie on the proton dripline [18–20] i.e. they are the lightest particle-bound isotopes of their respective element. The low proton (compared to neutron) separation energies of these nuclei make the production cross-sections of these nuclei in fusion-evaporation reactions extremely low ( $\sim \mu\text{b}$ ) compared to the total fusion cross-section ( $\sim 1 \text{ b}$ ). Projectile fragmentation provides an alternative mechanism to populate such nuclei, where the existence of isomers allows the identification of excited states [21].

Here we report on new results for the self-conjugate proton drip-line nuclei,  ${}^{82}_{41}\text{Nb}$  and  ${}^{86}_{43}\text{Tc}$ . These are the heaviest odd-odd  $N = Z$  nuclei in which  $\gamma$ -ray transitions have been observed. Their structure is also of nuclear astrophysical interest as they lie on the predicted rapid proton capture nucleosynthesis path ( $rp$ -process) at high temperature and density [22]. The isomer in  ${}^{86}\text{Tc}$  has been reported previously, and two transitions tentatively assigned [21]. In the same work evidence for a short-lived isomer was reported in  ${}^{82}\text{Nb}$  but no discrete  $\gamma$  rays observed. Some preliminary analyses of the current work have been reported in conference proceedings [23, 24].

The current experiment was performed at the

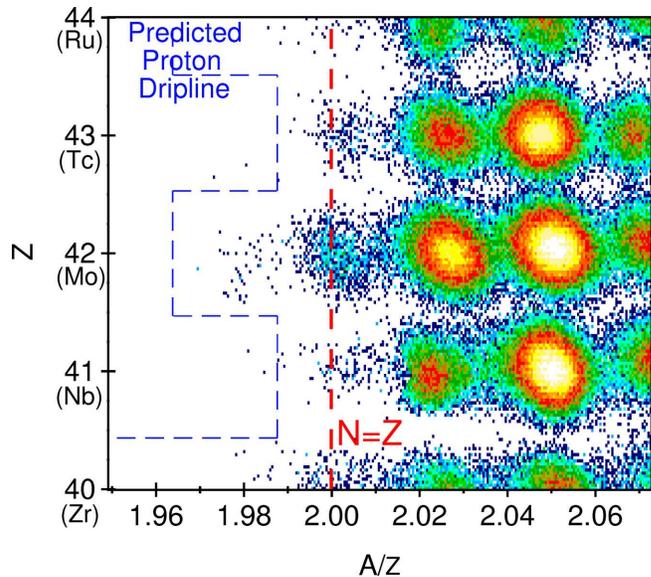


FIG. 1: Particle identification plot for combined data of the current work and the location of the predicted proton dripline as calculated from ref. [25].

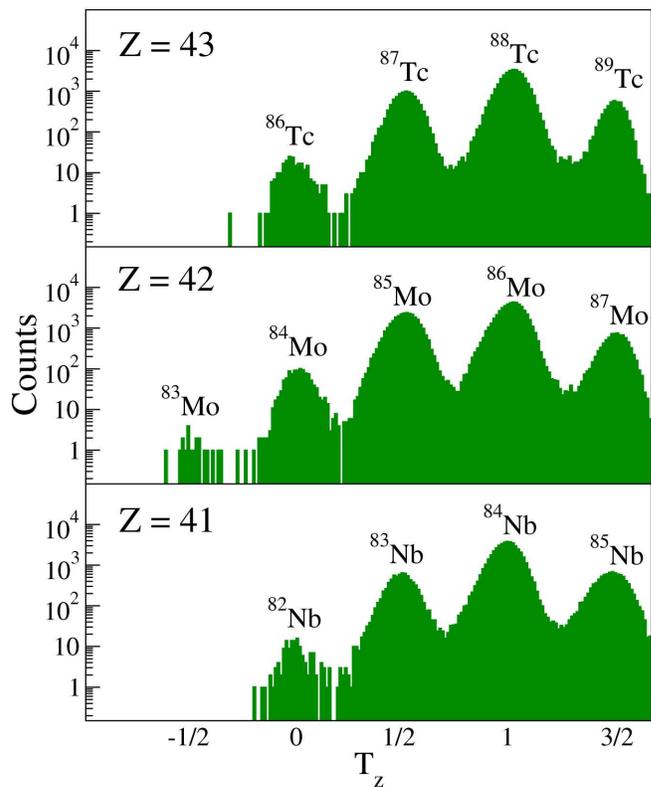


FIG. 2: Projections of the 2D spectrum in Fig. 1 for Technetium, Molybdenum and Niobium isotopes from the combined data of the current work.

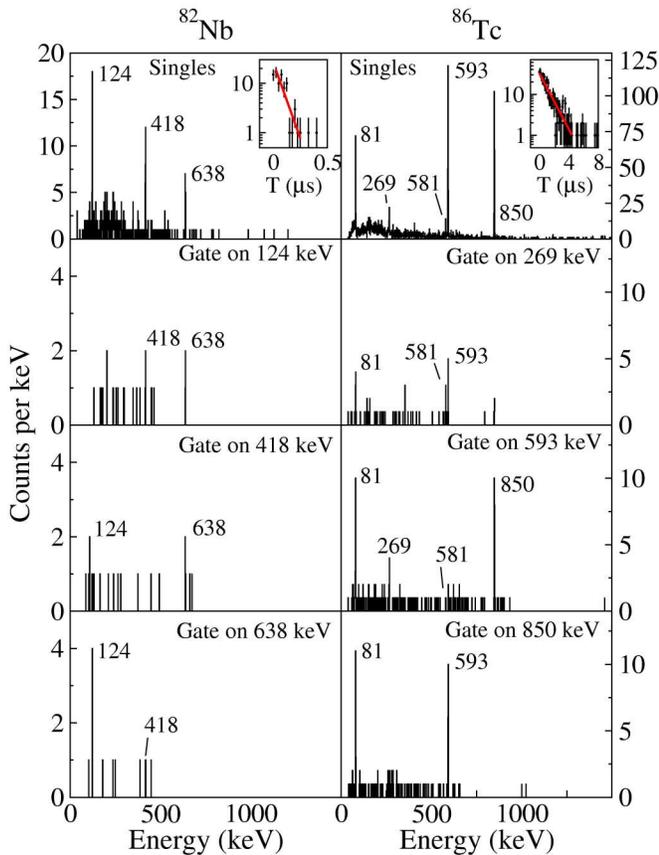


FIG. 3: Singles and coincident energy spectra of delayed  $\gamma$ -ray events associated with  $^{82}\text{Nb}$  (**Left**) and  $^{86}\text{Tc}$  (**Right**). The upper panels show singles data for  $^{82}\text{Nb}$  gated between 150  $\rightarrow$  500 ns and  $^{86}\text{Tc}$  between 150 ns  $\rightarrow$  5  $\mu\text{s}$  after implantation. The fitted half lives are 133(25) ns and 1.59(20)  $\mu\text{s}$  respectively.

Gesellschaft für Schwerionenforschung (GSI) where a  $^{107}\text{Ag}$  primary beam was accelerated to 750 MeV per nucleon by the SIS-18 synchrotron and made to impinge on a 4 g/cm<sup>2</sup> Be target. The typical primary beam current was  $2 \times 10^9$  ions per beam spill over an extraction time of 5 s. The reaction products were transported to the focal plane of the FFragment Separator (FRS) [26] and identified using values of  $A/q$  and  $Z$  calculated from measurements of magnetic rigidity, time-of-flight, position and energy loss. The fully-stripped nature (i.e.  $Z = q$ ) of the ions enabled an unambiguous particle identification [26, 27]. The ions were stopped in a 7 mm thick perspex block at the centre of the Stopped RISING gamma-ray array [27, 28]. In this configuration the array consisted of 105 Ge crystals grouped into 15, 7 element clusters with a measured photo-peak efficiency of  $\sim 15\%$  at 662 keV. Gamma rays emitted in the decay of isomeric states were detected in the array and correlated with the arrival time of the associated ion.

During the experiment, the FRS was tuned to maximise the transmission of  $^{82}\text{Nb}$  ( $\sim 5$  hrs primary beam

time) and  $^{86}\text{Tc}$  ( $\sim 90$  hrs) in two separate settings. In the  $^{86}\text{Tc}$  setting,  $^{82}\text{Nb}$  ions were also transmitted to the focal plane and in the final analysis the data from both settings were combined. Figures 1 and 2 show example particle identification plots from the current work. The  $T_z = 0$  nuclides  $^{82}\text{Nb}$  and  $^{86}\text{Tc}$ , lie on the proton dripline, but the even- $Z$ ,  $T_z = -\frac{1}{2}$   $^{83}\text{Mo}$  is also particle bound, consistent with previous findings [19, 20]. Figure 3 shows the delayed  $\gamma$ -ray spectra associated with isomeric decays in  $^{82}\text{Nb}$  and  $^{86}\text{Tc}$ . These spectra were produced from the implantation of  $\sim 4500$  and  $\sim 7700$  ions of  $^{82}\text{Nb}$  and  $^{86}\text{Tc}$  respectively.

Three discrete  $\gamma$  ray transitions (124, 418 and 638 keV) are associated with the decay of a  $T_{1/2} = 133(25)$  ns isomer in  $^{82}\text{Nb}$  and are demonstrated to be in mutual coincidence (see Fig. 3). The half-life measurement was made by performing a least-squares fit to the summed time spectra associated with the 418 and 638 keV transitions (see inset of Fig. 3). The two higher energy transitions are notably similar in energy to the  $2^+ \rightarrow 0^+$  (407 keV) and  $4^+ \rightarrow 2^+$  (634 keV) in the  $T_z = +1$  isobar,  $^{82}\text{Zr}$  [29] suggesting these are decays from  $T = 1$  isobaric analogue states in  $^{82}\text{Nb}$ . On this basis they are assigned as the first two transitions of the  $^{82}\text{Nb}$ ,  $T = 1$  ground-state band.

The  $\gamma$ -ray intensity balance around the ( $4^+$ ) state has been used to infer the internal conversion coefficient,  $\alpha_{tot}$ , of the 124 keV transition to be 0.3(3). This is consistent with M1, E1 or E2 multipolarity (see Table I [30]), but does not allow a clear discrimination between the possibilities. The deduced value for the isomeric ratio [31] depends on the value of the internal conversion coefficient of the direct decay. Although the statistical uncertainties are significant, an E2 multipolarity for the 124 keV transition would result in an unphysically large isomeric ratio greater than 100%. E1 or M1 assignments yield more physically realistic values less than 100%. Using these arguments, plausible spin/parity assignments are restricted to  $I^\pi = 5^-$  and  $5^+$ .

Five  $\gamma$  ray transitions are identified following the decay of a  $T_{1/2} = 1.59(20)$   $\mu\text{s}$  isomer in  $^{86}\text{Tc}$  (Fig. 3). These data show the 81, 593 and 850 keV  $\gamma$  rays to be in mutual coincidence. The latter two are assumed to be decays from the  $T = 1$  isobaric analogue states of the  $T_z = +1$  isobar,  $^{86}\text{Mo}$  ( $2^+ \rightarrow 0^+ = 567$  keV and  $4^+ \rightarrow 2^+ = 761$  keV [32]). The 269 and 581 keV  $\gamma$ -ray energies sum to 850 keV indicating a competing decay branch to the ( $4^+$ )  $\rightarrow$  ( $2^+$ ) transition. Although the ordering cannot be unambiguously determined here, the cascade can be confirmed by the 269 and 850 keV coincidence gates. (We note that some counts at 850 keV are observed in the 269 keV gate due to components of the Compton background associated with the 593 keV transition).

Using intensity balance arguments the internal conversion coefficient for the 81 keV  $\gamma$  ray is inferred to be  $\alpha_{tot} = 3.5(8)$ . A comparison with calculated values (Table I) indicates this transition to be a stretched E2, leading to a spin/parity assignment of ( $6^+$ ) for the isomeric state. While such a conversion coefficient could in prin-

TABLE I: Weisskopf single-particle half life estimates (neglecting internal conversion) and total conversion coefficients for transitions in  $^{82}\text{Nb}$  and  $^{86}\text{Tc}$  [30].

	$^{82}\text{Nb}$ , 124 keV		$^{86}\text{Tc}$ , 81 keV	
	$T_{1/2}$ (s)	$\alpha_{tot}$	$T_{1/2}$ (s)	$\alpha_{tot}$
E1	$1.87 \times 10^{-13}$	0.065	$6.50 \times 10^{-13}$	0.245
M1	$1.46 \times 10^{-12}$	0.131	$3.41 \times 10^{-12}$	0.514
E2	$8.97 \times 10^{-7}$	0.534	$7.08 \times 10^{-6}$	2.690
M2	$5.64 \times 10^{-5}$	1.120	$4.59 \times 10^{-4}$	6.420

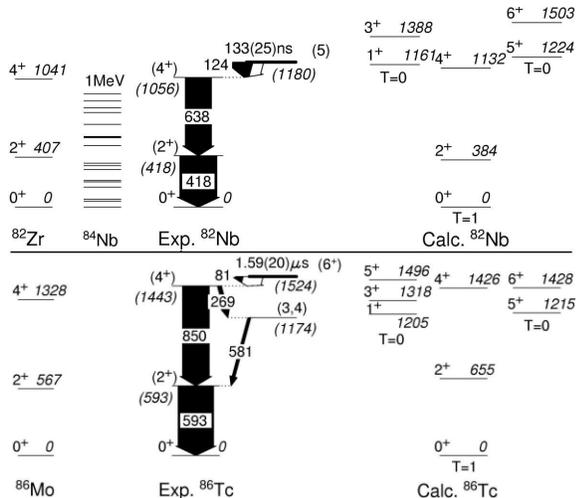


FIG. 4: The experimental and theoretical (Projected Shell Model) level schemes of  $^{82}\text{Nb}$  and  $^{86}\text{Tc}$ . Partial level schemes of  $^{82}\text{Zr}$ ,  $^{84}\text{Nb}$  and  $^{86}\text{Mo}$  are also shown for comparison [32–34].

Multiple arise from a highly mixed E1/M2 transition such an assignment is unlikely on the basis of the expected partial half-lives for such competing multipoles (see Table I). The measured half life is also consistent with a single-particle 81 keV E2 transition rate.

Level schemes for  $^{82}\text{Nb}$  and  $^{86}\text{Tc}$  are presented in Fig. 4. The low level-density observed following the isomeric decay is consistent with the reduced level density reported in other odd-odd  $N = Z$  systems. Only one excited state is observed below 1 MeV in  $^{82}\text{Nb}$  compared to 17 excited states reported in  $^{84}\text{Nb}$  [34]. We suggest this is further evidence for a  $T = 1$   $np$  pair gap specific to  $N = Z$  nuclei [11, 12].

The Projected Shell Model (PSM) [35] including  $np$  interactions [36] reproduces the observed ground state structures in the even-even  $N = Z$  nuclei with  $A = 68 \rightarrow 88$ . PSM calculations have now been performed for  $^{82}\text{Nb}$  and  $^{86}\text{Tc}$ . The results for the positive parity states are shown in Fig. 4. For both nuclei, the calculation shows a low-lying  $I^\pi = 5^+$  2-quasiparticle state with a Nilsson configuration of  $\nu[422]5/2^+ \times \pi[422]5/2^+$ . In  $^{82}\text{Nb}$  this  $I^\pi = 5^+$  configuration is predicted to lie just above the  $T = 1$ ,  $4^+$  state. The population of a  $T = 0$  state with this configuration is consistent with the low-lying band

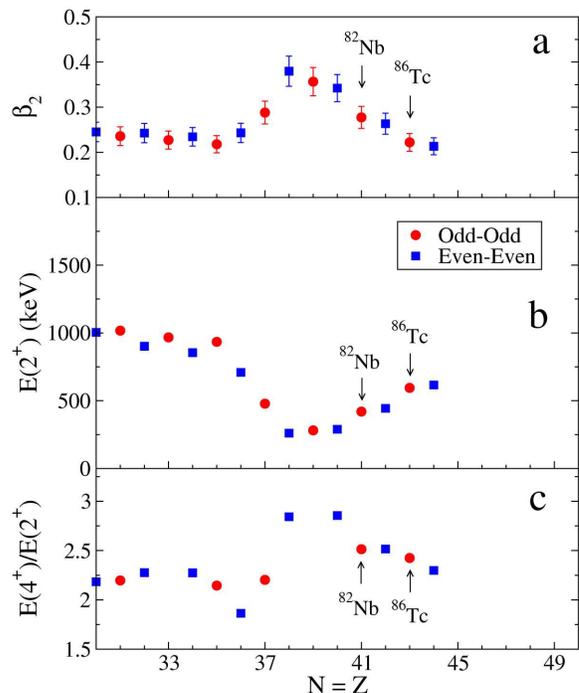


FIG. 5: Structural evolution of the  $N = Z$  nuclei across the  $fp$  shell; **a**: Deformation calculated using the empirical relationship described in Ref. [39], **b**: Excitation energy of the first  $I^\pi = 2^+$  state, and **c**:  $E(4^+)/E(2^+)$  ratio.

structures in the  $T_z = +\frac{1}{2}$  neighbours  $^{81}\text{Zr}$  [37] and  $^{83}\text{Nb}$  [17, 38]. The calculations also predict two low-lying  $K = 4^-$  states at approximately 1.3 MeV in  $^{82}\text{Nb}$  and negative parity states of mixed  $K = 5, 6$  at  $\approx 1.2$  MeV in  $^{86}\text{Tc}$ . A  $I^\pi = K^\pi = 6^+$  state can be formed in  $^{86}\text{Tc}$  by a coupling of the  $[422]5/2^+$  and  $[413]7/2^+$  Nilsson orbitals but appears at a significantly higher excitation energy in the calculation.

Figure 5 shows energy systematics for  $N = Z$  nuclei from  $A = 60 \rightarrow 88$  including the current data. Using an empirical relationship between the energy of the first  $2^+$  energy and quadrupole deformation ( $\beta_2$ ) [39] an estimate of the ground-state deformation can be made for both even-even ( $T = 0$ ) and odd-odd ( $T = 1$ ) structures.

One mechanism by which nuclear half lives can be prolonged in axially deformed nuclei is that of  $K$  hindrance [40]. The quantum number  $K$ , is the projection of the total angular momentum along the nuclear axis of symmetry. If there is a significant mismatch between the  $K$  values of initial and final states in electromagnetic decay, such that  $\Delta K \geq \lambda$  where  $\lambda$  is the multipole order of the decaying transition, then the transition is expected to be ‘ $K$  hindered’. A  $124$  keV M1 decay from an  $I^\pi = K^\pi = 5^+$  to a  $I^\pi = 4^+$ ,  $K = 0$  state has  $\Delta K = 5$ ,  $\nu = 4$ , where  $\nu = \Delta K - \lambda$ . The reduced hindrance for  $K$ -isomeric decays is defined by  $f_\nu = (T_{1/2}/T_{1/2}^W)^{1/\nu}$

[40]. An assumption of a pure M1 isomeric decay in  $^{82}\text{Nb}$  gives  $f_\nu \approx 18$ , a substantial value intermediate between the accepted values of  $f_\nu \sim 100$  for the best case axially-symmetric  $K$  isomers and  $\sim 1$  for unhindered decays.

It is noteworthy that the isomeric state in  $^{86}\text{Tc}$  lies in the vicinity of the predicted proton separation energy of 1393 (409) keV [25] thus allowing the possibility of direct proton emission from the isomer competing with internal electromagnetic decay. Such a competing (proton) decay branch would speed up the total mean lifetime of the isomeric state and could explain the apparent absence of any  $K$ -hindrance in this nucleus. This possibility cannot be confirmed in the current experiment. (The proton separation energy for  $^{82}\text{Nb}$  is calculated to be 1775 (341) keV [25]).

In even-even nuclei, the ratio of excitation energies of the yrast  $4^+$  and  $2^+$  states ( $R = \frac{E(4^+)}{E(2^+)}$ ) is used as a signature for nuclear shapes with an idealised quadrupole vibrational nucleus having  $R = 2.0$  and a perfect axially symmetric rotor having  $R = 3.33$ . Figure 5 shows such a plot for  $N = Z$  nuclei between  $^{60}_{30}\text{Zn}$  and  $^{88}_{44}\text{Ru}$ . The value of  $R = 2.53$  for  $^{82}\text{Nb}$  is consistent with some axial asymmetry and/or  $\gamma$ -softness, which could explain the inferred reduced hindrance for an M1 transition. We note that any proposed  $K$ -hindrance is not apparent for the

(likely) E2 isomeric decay in  $^{86}\text{Tc}$ . This could be qualitatively understood by a reduction in the deformation and associated increase in  $\gamma$ -softness compared to  $^{82}\text{Nb}$  or by competition from an unobserved proton decay branch.

In summary, we have identified low-lying structures in  $^{82}\text{Nb}$  and  $^{86}\text{Tc}$ , the heaviest odd-odd  $N = Z$  systems for which internal decays have been measured to date. The data suggest a dominance of the  $T = 1$  pairing interaction over its  $T = 0$  counterpart throughout the  $fpg$  shell. Plausible evidence for an isospin changing  $K$  isomer along the  $N = Z$  line is also presented.

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