Shape coexistence and isomeric states in neutron-rich ¹¹²Tc and ¹¹³Tc

A. M. Bruce,¹ S. Lalkovski,^{1,*} A. M. Denis Bacelar,¹ M. Górska,² S. Pietri,^{2,3} Zs. Podolyák,³ Y. Shi,⁴ P. M. Walker,³ F. R. Xu,⁴

P. Bednarczyk,² L. Cáceres,² E. Casarejos,⁵ I. J. Cullen,³ P. Doornenbal,^{2,6} G. F. Farrelly,³ A. B. Garnsworthy,³ H. Geissel,² W. Gelletly,³ J. Gerl,² J. Grębosz,^{2,7} C. Hinke,⁸ G. Ilie,⁶ G. Jaworski,^{9,10} I. Kojouharov,² N. Kurz,² S. Myalski,⁷ M. Palacz,⁹

W. Prokopowicz,² P. H. Regan,³ H. Schaffner,² S. Steer,³ S. Tashenov,² and H. J. Wollersheim²

¹School of Computing, Engineering and Mathematics, University of Brighton, Brighton BN2 4GJ, United Kingdom

²Gesellschaft für Schwerionenforschung mbH, Planckstrasse 1, D-64291 Darmstadt, Germany

³Department of Physics, University of Surrey, Guildford GU2 7XH, United Kingdom

⁴Department of Technical Physics, Peking University, Beijing 100871, China

⁵Facultad de Física, Universidad de Vigo, E-36310 Vigo, Spain

⁶Institut für Kernphysik, Universität zu Köln, Zülpicher Straße 77, D-50937 Köln, Germany

⁷Niewodniczański Institute of Nuclear Physics, Polish Academy of Science, ulica Radzikowskiego 152, Krakow 31-342, Poland

⁸Physik-Department E12, Technische Universität München, D-85748 Garching, Germany

⁹Heavy Ion Laboratory, Warsaw University, ulica Pasteura 5A, 02-093 Warszawa, Poland

¹⁰Faculty of Physics, Warsaw University of Technology, Koszykowa 75, 00-662 Warszawa, Poland

(Received 9 September 2010; published 14 October 2010)

Isomeric states in ¹¹²Tc and ¹¹³Tc, with half-lives of 150(17) ns and 500(100) ns, respectively, have been observed following the relativistic fission of ²³⁸U. The fission fragments have been separated in a fragment separator and identified by means of energy-loss and time-of-flight techniques. In both nuclei, the ground-state configuration is calculated to have an oblate shape and the isomerism is proposed to arise due to transitions from a triaxial excited state to a low-lying oblate state.

DOI: 10.1103/PhysRevC.82.044312

PACS number(s): 21.10.Tg, 23.20.Lv, 23.35.+g, 27.60.+j

I. INTRODUCTION

The shapes of neutron-rich nuclei with $A \approx 110$ have been the subject of intensive discussion in the last decade since large scale Finite-Range Liquid Drop Model (FRLDM) calculations [1] showed that the neutron-rich nuclei in all of the isotopic chains with Z = 40-48 undergo a prolate-tooblate shape transition when approaching N = 80. This was reinforced by Skalski et al. [2], who used both the liquid drop and the FRLDM for the macroscopic energy and both BCS and the particle number projection method for the pairing energy. ^{112,113}Tc have Z = 43 and N = 69,70, respectively, and, for the neighboring Z = 42 (Mo) nuclei, calculations [2] indicate a ground-state shape which is prolate for $N \leq 66$ and oblate for $N \ge 68$. For Z = 44 (Ru), the prolate-oblate shape change in the ground state is predicted to occur between N = 64 and 66. Moreover, relativistic mean-field calculations [3] predict an oblate ground state at N = 64, ¹⁰⁶Mo and, more recently, cranked and configuration-constrained shellmodel calculations have predicted the appearance of oblate multiquasiparticle states in N = 66 isotones with prolate ground states [4]. Clearly, this is predicted to be a region of rapidly changing shapes.

Although the nuclei which are predicted [1] to have oblate ground states in neutron-rich Zr, Nb, and Mo are not yet accessible experimentally, those in neutron-rich Tc, Ru, Rh, Pd, and Ag isotopes can be reached by "conventional" studies of fission products. Among the first experimental articles

reporting on such nuclei was a study of the neutron-rich Pd isotopes [5], produced in induced fission reactions, in which the oblate shape of the ¹¹¹Pd ground state predicted by the FRLDM [1] was ruled out. Indeed, more recent experimental work has led to the prolate interpretation being extended to the heavier Pd isotopes with Hua et al. [6], suggesting that a prolate-to-oblate shape transition occurs only at ¹¹⁶Pd. In the ruthenium isotopic chain this transition was assumed to take place at ¹¹¹Ru [6] but, more recently, ¹¹³Ru has been interpreted to have a significant degree of triaxiality [7]. The development of techniques for fission fragment γ -ray spectroscopy has also made it possible to cross the (50,82) neutron mid shell in the Tc isotopic chain, reaching ¹¹¹Tc (N = 68) [8]. In this work the authors conclude, mainly on the basis of systematics, that the ground state corresponds to the $5/2^{+}[422]$ proton excitation in a prolate-deformed potential. Intriguingly, however, they do state that "there are excitations seen in ¹¹¹Tc which suggest that the deformation above N = 68 may be oblate." Thus the question of where the prolate-oblate transition occurs in the technetium isotopic chain remains open. In this context, the structures of ¹¹²Tc and ¹¹³Tc, which are the subject of this paper, are of particular interest.

II. EXPERIMENTAL DETAILS

The nuclei of interest were studied as part of the RISING "stopped beam" campaign [9] at Gesellschaft für Schwerionenforschung (GSI), Darmstadt. They were produced in the relativistic fission of a ²³⁸U beam accelerated in the GSI LINAC to 11.4 A MeV and then to 750 A MeV in the GSI SIS18 synchrotron. The typical beam intensity was

^{*}Current address: Department of Physics, University of Sofia, 1164 Sofia, Bulgaria.

10⁹ particles per second for a 1-s beam spill, with a repetition period of 10–15 s. The beam impinged on a ⁹Be target with a thickness of 1.0 g/cm². Fully stripped fission fragments were separated in the GSI FRagment Separator (FRS) [10], using the $(B\rho)_1 - \Delta E - (B\rho)_2$ technique, and identified by means of time-of-flight and energy-loss techniques. The Time of Flight (ToF) is measured between two scintillation detectors at the intermediate and final focal planes of the FRS and the energy loss is measured in an eight-anode ionization chamber (MUSIC). These detectors were calibrated using the primary ²³⁸U beam. The data set presented in this paper was collected with the FRS tuned to ¹¹⁰Nb.

At the end of the separator, the fragments were slowed by an Al degrader with a thickness of 3.7 g/cm² and implanted in a 5-mm-thick copper stopper. γ rays emitted by the implanted nuclei were measured in the RISING γ -ray array, comprising 15 cluster detectors, which has an efficiency of ~15% for the 662-keV transition in ¹³⁷Cs [11]. The signals from each Ge detector have been processed via XIA Digital Gamma Finder modules for energy and time analysis. The time stamp of the modules was 25 ns. A detailed description of the RISING performance in its stopped beam configuration is given in [11,12]. The acquisition system was triggered by the arrival of a fragment and remained "open" for a time window of 100 μ s. Fragments which did not implant triggered a scintillation detector behind the copper stopper which provided a veto.

III. EXPERIMENTAL ANALYSIS AND RESULTS

The atomic number of the fragment (*Z*) was calculated from the energy loss in the MUSIC detector, corrected for particle velocity and trajectory. The mass-to-charge ratio (A/q) was calculated from the magnetic rigidity of the particles and the ToF. Figure 1 shows the relevant particle identification plot where the locations of ¹⁰⁶Nb and ¹¹³Tc ions are indicated. ¹⁰⁶Nb contains a known isomer with a half-life of 845(35) ns [13] which served to confirm the particle identification. In addition to the veto detector mentioned above, other





FIG. 2. γ -ray energy spectra obtained in coincidence with ¹¹²Tc ions (a) and ¹¹³Tc ions (b) and with a time gate of width 1.25 μ s starting 300 ns after implantation (to reduce the bremsstrahlung background).

cleaning conditions imposed in the off-line analysis included the exclusion of events where the fragment changed its charge state during transmission through the separator and events where the fragment interacts with the degrader. Following identification of the fragments, two-dimensional spectra of the energies of γ rays measured in the RISING array and their emission time (relative to fragment deposition) were constructed for each isotope identified.

¹¹²Tc. Figure 2(a) shows two γ rays which are associated with ¹¹²Tc fragments. The relative intensities of the two transitions are $I_{\gamma}^{92} = 100 \pm 22\%$ and $I_{\gamma}^{258} = 91 \pm 9\%$. Analysis of the background fluctuations in the spectrum gives an upper limit of 10% of I_{ν}^{92} for a 166-keV γ ray which corresponds to the energy difference between the 92- and 258-keV transitions. There is no evidence of any transition at 340 keV, which is the sum of 258 and 92. Figure 3 shows coincidence spectra gated on (a) the 92-keV transition and (b) the 258-keV transition. These spectra have not had any background subtracted but, although the statistics are low, they do provide good evidence that the 92- and 258-keV transitions are in mutual coincidence. Figure 4(a) shows the time distribution of the 258-keV transition and a fit to this decay curve gives a half-life of 150(17) ns. The 92-keV transition is in a region with much background and a fit to its decay curve gives a half-life of 162(100) ns. Therefore it is concluded that there is one isomeric state in ¹¹²Tc which has a



FIG. 3. γ -ray energy spectra obtained in coincidence with ¹¹²Tc ions and (a) the 92-keV transition, (b) the 258-keV transition.



FIG. 4. Background-subtracted time distributions of (a) the 258-keV transition in 112 Tc and (b) the 114-keV transition in 113 Tc, along with associated fits.

half-life of the weighted average value of 150(17) ns. This is consistent with the recent findings of Folden *et al.* [14], who observed only the 258-keV transition and proposed that there is an isomeric state with a half-life of less than 500 ns.

¹¹³*Tc.* Figure 2(b) shows a 114-keV γ ray which is associated with ¹¹³Tc fragments. The transition is very weak but its time distribution is shown in Fig. 4(b). The associated fit indicates a half-life of the parent level of 500(100) ns. Examination of the coincidence spectrum associated with the 114-keV transition shows no evidence of coincident γ rays.

IV. INTERPRETATION AND DISCUSSION

 $^{112}Tc.$ Figure 5 shows the results of Potential-Energy Surface (PES) calculations [4] carried out separately for the positive- and negative-parity states in ¹¹²Tc. The calculations indicate a negative-parity ground state [shown in Fig. 5(a)] with an oblate deformation of $\beta_2 = 0.23$ and $\beta_4 = -0.05$ formed from the $7/2^{-}[523]$ neutron $\otimes 5/2^{+}[422]$ proton coupling. This shape is in good agreement with that calculated by Möller *et al.* [1] (oblate with $\epsilon_2 = 0.25$ and $\epsilon_4 = +0.09$) and by Skalski et al. [2] for neighboring even-even Mo (Z = 42) and Ru (Z = 44) isotopes (oblate with $\beta_2 = 0.25$ and $\beta_4 = -0.09$). The Gallagher-Moszkowski rule indicates that the $K^{\pi} = 6^{-}$ coupling should be the lowest with the $K^{\pi} = 1^{-}$ coupling expected at an excitation energy of about 200 keV [15]. Figures 5(b) and 5(c) indicate the shapes for the two possible positive-parity states. In Fig. 5(b) the shape of the $+\nu, +\pi$ configuration with lowest energy is shown as $\beta_2 = 0.24$, $\beta_4 = -0.03$, and $\gamma = 44^{\circ}$ and in Fig. 5(c), the shape of the $-\nu$, $-\pi$ configuration with lowest energy is shown as $\beta_2 = 0.29$, $\beta_4 = -0.01$, and $\gamma = 31^\circ$. Both of these



FIG. 5. Potential-Energy Surface (PES) calculations for ¹¹²Tc for a negative-parity neutron and a positive-parity proton $(-\nu, +\pi)$ (a), $+\nu$, $+\pi$ (b), and $-\nu$, $-\pi$ (c). Isolines are placed at intervals of 200 keV and the energies of the minima in (b) and (c) are 411 and 474 keV relative to the $-\nu$, $+\pi$ ground state. The $+\nu$, $-\pi$ configuration is also calculated to be triaxial but at an even higher excitation energy.

configurations are calculated to have J = 3 and excitation energies ~450 keV.

Figure 6 shows the energy spectrum for these states and includes "in-band" oblate states calculated assuming a moment-of-inertia one-half of that of a rigid body. Comparison of the measured half-life of the observed isomeric state [150(17) ns] with calculations of Weisskopf single-particle estimates indicates that a transition with $\lambda > 2$ is unphysical. It is therefore unlikely that the observed transitions correspond to the decay from either of the $J^{\pi} = 3^+$ triaxial states directly to the ground state. On this basis, it is proposed that the 258-keV transition is from one of the triaxial $J^{\pi} = 3^+$ states to a $J^{\pi} = 2^{-}$ state built on the $J^{\pi} = 1^{-}$ state from the unfavored $7/2^{-}[523]$ neutron $\otimes 5/2^{+}[422]$ proton coupling. The measured half-life indicates that the E1 transition is hindered by a factor of $\sim 10^7$ compared to the Weisskopf single-particle estimate, corrected for electron conversion [16]. E1 transitions are known, however [17], to be hindered, typically by factors $\sim 10^5$, and a transition from a triaxial $J^{\pi} = 3^+$ state to an oblate $J^{\pi} = 2^-$ state would be expected to exhibit additional hindrance due to the shape change. In



FIG. 6. The calculated energy spectrum for 112 Tc. The in-band oblate states assume a moment of inertia one-half of that of a rigid body. The triaxial states correspond to the configurations in Figs. 5(b) and 5(c).



FIG. 7. PES calculations for the lowest-energy positive-parity (a) and negative-parity (b) states in 113 Tc. Iso-lines are placed at intervals of 200 keV.

this scenario, which is not inconsistent with the observed γ -ray intensities, the 92-keV transition then corresponds to the in-band transition from the $J^{\pi} = 2^{-}$ state to the $J^{\pi} = 1^{-}$ state. The *M*5 decay from the $J^{\pi} = 1^{-}$ to the $J^{\pi} = 6^{-}$ ground state would not be expected to be observed since the Weisskopf single-particle half-life estimate, corrected for electron conversion [16], for a level decaying by such a transition is ~120 000 years and the level might be expected to preferentially decay by β emission.

¹¹³*Tc*. The results of PES calculations carried out separately for the positive- and negative-parity states in ¹¹³Tc are shown in Fig. 7. Figure 7(a) shows the results for the positiveparity configuration, indicating a shallow oblate minimum at $\beta_2 = 0.24$, $\beta_2 = -0.05$, $\gamma = 60^{\circ}$ corresponding to the $5/2^+[422]$ Nilsson orbit. The results for the negative-parity configuration in Fig. 7(b) show a deep triaxial minimum at $\beta_2 = 0.29$, $\beta_2 = -0.02$, and $\gamma = 29.8^{\circ}$ with $J^{\pi} = 5/2^{-}$. This state is calculated to lie above the positive-parity minimum and therefore the ground state of ¹¹³Tc is assigned as $J^{\pi} = 5/2^+$. Under the assumption that there is no unobserved low-energy transition, it is proposed that the 114-keV transition connects the two different configurations. The Weisskopf single-particle half-life estimate, adjusted for electron conversion [16], for a 114-keV *E*1 transition is 1.8×10^{-4} ns, which, when compared with the measured value of 500(100) ns indicates a hindrance of 2.8×10^6 . Given the typical *E*1 hindrance [17] discussed in the preceding section, and the additional hindrance expected due to the shape change, it is again proposed that the isomerism is due to an *E*1 transition between states of triaxial and oblate shape.

V. CONCLUSIONS

Isomeric states with half-lives of 150(17) and 500(100) ns, respectively, have been observed in ¹¹²Tc and ¹¹³Tc. Comparison with the results of PES calculations suggest that both nuclei have oblate ground states. The isomerism in both nuclei is interpreted as being associated with a shape change from triaxial excited states to low-lying oblate structures. The location of these isomeric levels may prove extremely useful as experimental tags in future in-beam measurements.

ACKNOWLEDGMENTS

This work is supported by the STFC(UK), AWE plc, the Bulgarian National Science Fund (Grant No. DMU02/1-06.01.2010), the Polish Ministry of Science and Higher Education (Grants No. 1-P03B-030-30 and No. 620/E-77/SPB/GSI/P-03/DWM105/2004-2007), the Spanish Ministerio de Educación y Ciencia (Grant No. FPA2005-00696), the German Federal Ministry of Education and Research (Grant No. 06KY205I), and EURONS (EU Contract No. 506065).

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