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Letter

First observation of excited states in the ^{111}Tc nucleus —A new region of deformation at $40 \leq Z \leq 46$, $N \geq 68$?

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Abstract. The ^{111}Tc nucleus, populated in the spontaneous fission of ^{248}Cm, was studied by means of prompt γ-ray spectroscopy using the EUROGAM2 array. Excited states in ^{111}Tc were observed for the first time. Systematics of energy levels in odd-A Tc isotopes, obtained in our study of ^{107}Tc and ^{109}Tc provide a reliable spin and parity assignment $I = 5/2^+$ to the head of the new band in ^{111}Tc, interpreted as the $\pi 5/2^+[422]$ orbital originating from the proton $g_{9/2}$ shell. This level is most likely the ground state. Therefore, the $(9/2^+, 7/2^+)$ spin-parity assignment to the ground state of ^{111}Tc, reported previously, is unlikely. Properties of the yrast band in ^{111}Tc suggest prolate deformation of this band. There are hints that the deformation of ^{111}Tc is larger than that of ^{109}Tc, possibly due to admixtures of oblate-deformed configurations, which lower their excitation energy with increasing neutron number.

PACS. 21.10.-k Properties of nuclei; nuclear energy levels – 23.20.-g Electromagnetic transitions – 25.85.Ca Spontaneous fission – 27.60.+j $90 \leq A \leq 149$

Our study of the ^{110}Mo$_{68}$ nucleus [1] provided new arguments supporting the idea [2] that nuclear deformation in the $38 < Z < 46$, $58 < N < 70$ region is due to the correlated occupation of the $h_{11/2}$ neutron and $g_{9/2}$ proton shells. The maximum deformation is observed along the line from $^{100}_{38}$Sr$_{60}$ towards $^{114}_{46}$Pd$_{68}$, as marked in fig. 1 (region I). It changes from strongly prolate in Sr and Zr isotopes (due to deformation-driving, low-Ω orbitals of the $\nu h_{11/2}$ and $\pi g_{9/2}$ parentage), through a triaxial in Mo and Ru isotopes (where mid-Ω subshells are active) towards low deformation of still unknown sign in Pd isotopes. Recent experiments claim prolate deformation in Pd [3,4], while the theory points to an oblate deformation at $N \geq 70$ [5,6] and predicts the most pronounced prolate-oblate transition around $Z = 40$.

The knowledge of the deformation in this region is relevant for studies of the astrophysical r-process [6,7]. The approximate position of the r-process path is sketched in fig. 1 based on refs. [7,8]. For the closed-shell nuclei, ^{78}Ni and ^{132}Sn, the path is located at a distance, ΔN, of approximately thirteen neutrons from the centre of the stability line (the accuracy of the ΔN determination is 1 to 2 units). For nuclei between ^{78}Ni and ^{132}Sn, ΔN is larger, with a maximum around Ru, as seen in the diagram on the right-hand side of fig. 1. This effect can be caused by nuclear deformation, expected around the middle of the

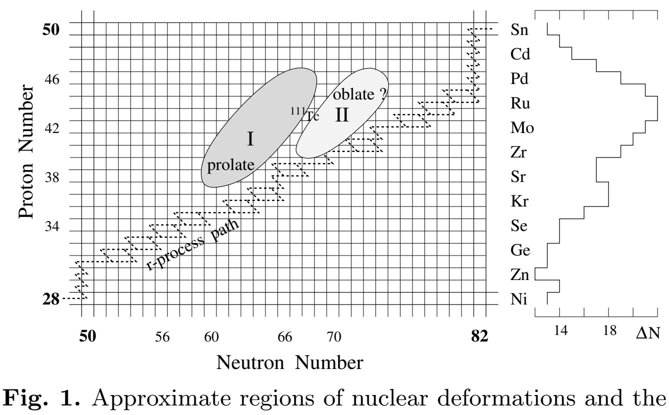

Fig. 1. Approximate regions of nuclear deformations and the approximate position of the r-process path in the $28 < Z < 50$, $50 < N < 82$ region. To the right, the distance, ΔN, from the stability line to the path, is shown for various isotopes.

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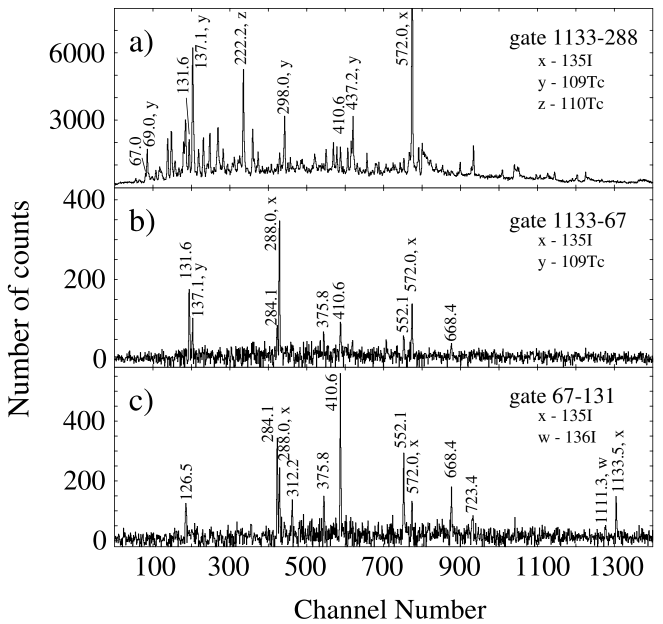

Fig. 2. Coincidence spectra of γ-rays following the fission of ^{248}Cm, gated on lines in ^{135}I and ^{111}Tc. Lines are labeled with their energies in keV. See text for more explanations.

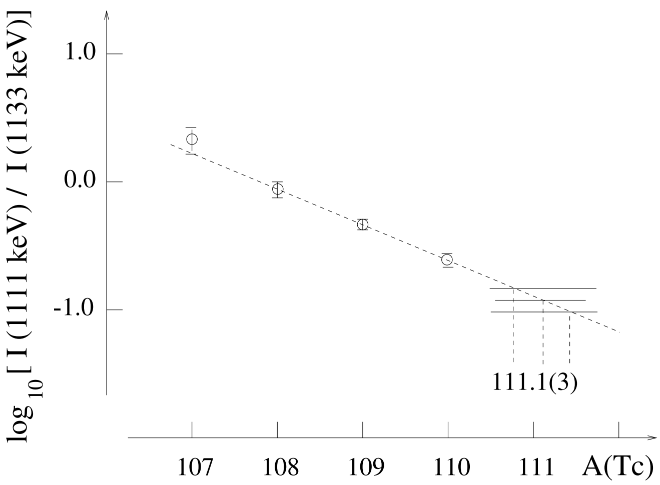

Fig. 3. Mass correlation diagram for Tc isotopes and the identification of the ^{111}Tc nucleus. See text for more explanations.

shell. In deformed nuclei, binding energies of nucleons increase. Consequently the r-process path tends to go round the region of deformation [7], moving away from the stability line.

A close inspection of fig. 1 shows that between ^{78}Ni and ^{90}Se ΔN is approximately constant, in accord with the expectation that up to the $Z = 34$ and $N = 56$ subshell closures, nuclei are spherical. The first increase of ΔN is observed around $Z = 35$ (Br and Kr), where the path moves up to $N = 66$. This first maximum in the ΔN diagram coincides with strongly prolate deformation around $^{100}_{38}$Sr$_{60}$ (region I). Interestingly, afterwards, the predicted path stays at the middle of the neutron shell, moving along $N = 66$ from ^{102}Kr up. This effect may correspond to the local maximum observed in the solar r-process composition around mass $A = 103$ (cf. fig. 2 in ref. [7]). It would also agree with our suggestion [1] that in very neutron-rich Sr and Zr isotopes the deformation decreases, after attaining the maximum around $N = 64$ (in Mo isotopes this drop in deformation is clearly evidenced [1]).

At $Z = 39$ (Y isotopes) the r-process path moves again to the neutron-rich side and ΔN increases to a maximum of 22 at $Z = 43$ (Tc isotopes). It is an interesting question as to whether this maximum could be associated with the hypothetical region of oblate deformation (II) predicted at $N \geq 70$ [5,6] and sketched in fig. 1 to the right-hand side of the deformation "ridge" (I). In ref. [1] we provided the first hint that above $N = 68$ the deformation may increase again. It is now important to gather more information, which could verify the existence of this new region of deformation, while the future studies with radioactive beams may tell about the sign of this deformation.

In technetium nuclei, which are next to the Mo isotopic chain, one can reach experimentally $N = 68$ neutrons and, eventually observe a transition from region I to the predicted region II of deformation. Therefore, we have undertaken a systematic study of neutron-rich Tc isotopes in order to find and study ^{111}Tc, the $N = 68$ isotone of ^{110}Mo, located between regions I and II, as marked in fig. 1. First, we studied lighter Tc isotopes to verify some unexpected properties, reported previously [9–12]. Our investigation of ^{107}Tc [13] and ^{109}Tc [14] provided regular systematics of excitation energies in Tc isotopes, which then served as a guide in a search for ^{111}Tc. In this Letter we report on the first observation of excited states in ^{111}Tc. These results were obtained from the analysis of multiple coincidences of prompt γ-rays from the spontaneous fission of ^{248}Cm, measured with EUROGAM2 array of anti-Compton spectrometers. More information about the experiment and analysis methods is given in refs. [15,16].

It is expected that ^{135}I is the strongest fission fragment partner to the ^{111}Tc nucleus. The 1133.5 keV high-energy line in ^{135}I [17] provides a good, clean gate to search for ^{111}Tc. Figure 2a shows a γ-ray spectrum double gated on the 1133 keV and 288 keV lines of ^{135}I, observed in our triple-gamma coincidences. Apart from the known lines in ^{135}I and complementary Tc isotopes, lighter than ^{111}Tc, there are new lines at 67.0 keV, 131.6 keV and 410.6 keV, which do not belong to ^{135}I. A spectrum double gated on the 67.0 keV and 1133.5 keV lines, shown in fig. 2b, indicates that the 67.0 keV transition belongs to a Tc isotope because only lines of ^{135}I and new lines at 131.6 keV, 284.1 keV and 552.1 keV are seen there. Further gates, as the one in fig. 2c, revealed more new lines forming a band, which is in coincidence with the 1133.5 keV and 288.0 keV lines of ^{135}I but also with the 1111.3 keV line of ^{136}I [18]. This observation confirms the assignment of the new cascade to a Tc isotope and allows the identification of this isotope. We have calculated the ratio, R, of the intensity of the 1111.3 keV line in ^{136}I to the intensity of the 1133.5 keV line in ^{135}I, observed in spectra double

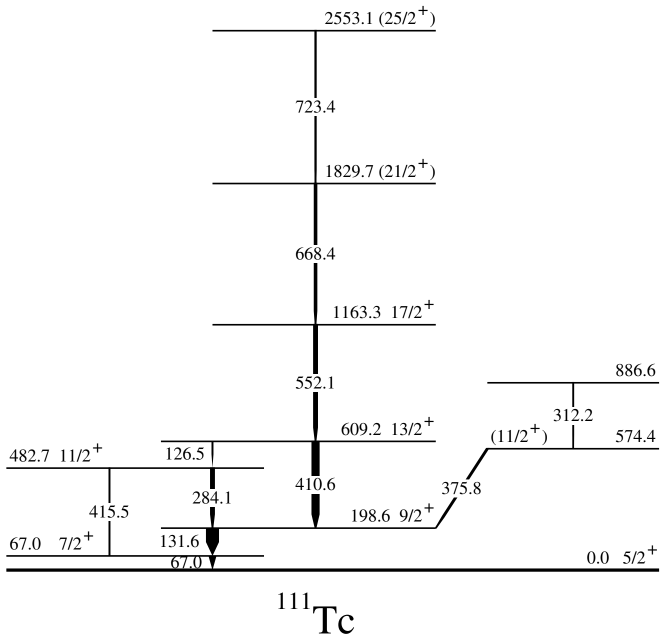

Fig. 4. Partial level scheme of ^{111}Tc as obtained in the present work. Transition and excitation energies are given in keV.

gated on gamma lines belonging to various Tc isotopes. Due to the correlation between masses of the complementary fission fragments [2], the ratio R observed in these spectra depends on the mass of the gated Tc isotope. In fig. 3 we show the ratio R as a function of the mass of the gated Tc isotope. Because of the large variation in R, we show $\log_{10}(R)$ values. The dependence is nearly linear. The dashed line represents a straight-line fit to the data points available for ^{107,108,109,110}Tc [13]. We assume that this line can be extrapolated to mass $A = 111$. The ratio for the new cascade is represented by a horizontal bar at $\log_{10}(R) = -0.93(8)$. The intersection of this value with the linear fit determines the mass of the unknown Tc isotope to be 111.1(3). This indicates that the new cascade belongs to the ^{111}Tc nucleus.

The proposed partial level scheme of ^{111}Tc, as obtained in this work, is displayed in fig. 4. The order of the transitions at the top of the cascade has been proposed according to their intensities, which are given in table 1.

We have determined angular correlations between transitions in the yrast cascade of ^{111}Tc, using techniques described in ref. [15]. The results are shown in table 1. Assuming that the 410.6 keV transition is a stretched quadrupole, as discussed below, we concluded that the 552.1 keV transition is a stretched quadrupole while the 131.6 keV transition corresponds to a $\Delta I = 1$ change in spin. Since no half-life longer than 10 ns has been observed in connection with the 552.1 keV transition, we concluded that it has an $E2$ multipolarity. The angular correlation for the 67.0-131.6 keV cascade is consistent with both lines corresponding to $\Delta I = 1$.

We found the conversion coefficient for the 67.0 keV transition from a spectrum measured by Low-Energy Pho-

Table 1. Properties of γ transitions in ^{111}Tc, as observed in this work. Ref. E_γ denotes the second transition in a cascade, for which the angular correlation has been determined.

E_γ (keV)	I_γ (rel.)	A_2/A_0	A_4/A_0	Multi-polarity	Ref. E_γ (keV)
67.0	30(8)	0.04(3)	−0.03(3)	$M1+E2$	131.6
131.6	100(5)	−0.06(2)	0.04(3)	$\Delta I = 1$	410.6
126.5	32(5)				
284.1	40(5)				
312.2	20(5)				
375.8	20(5)				
410.6	70(8)				
415.5	15(5)				
552.1	40(5)	0.09(3)	−0.01(3)	$E2$	410.6
668.4	27(5)				
723.4	20(5)				

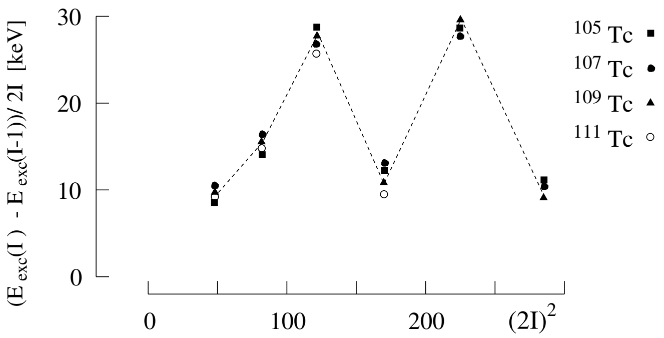

Fig. 5. Staggering in the yrast cascades of odd-A Tc isotopes. The data are from this work and refs. [11,13,14]. Dashed lines are drawn to guide the eye.

ton detectors used together with EUROGAM. In a spectrum double gated on the 410.6 keV and 131.6 keV lines only the 67.0 keV gamma line and the corresponding technetium X-ray, K_α line at 18.3 keV were present (for more details about this technique, see ref. [13]). The resulting value, $\alpha_K = 1.9(5)$ indicates that the 67.0 keV transition is of a $M1+E2$ character since the theoretical α_K values are 0.3, 0.8 and 3.2 for an $E1$, $M1$ and $E2$ transition, respectively.

The parity change connected with the 131.6 keV transition has not been determined experimentally but there are other arguments indicating its $M1+E2$ character. The yrast cascade observed in ^{111}Tc resembles closely analogous yrast cascades in lighter Tc isotopes, which are interpreted as decoupled bands based on the $5/2^+[422]$ orbital originating from the $g_{9/2}$ proton shell. In these bands a pronounced staggering is observed. This is shown, after ref. [11] in fig. 5, where we also included the new data found for ^{111}Tc (open circles). The new cascade fits well the trend if one assigns spins $7/2^+$, $9/2^+$ $11/2^+$ and $13/2^+$ to the 67.0, 198.6, 482.7 and 609.9 keV levels, respectively. Considering the $M1+E2$ character of the 67.0 keV transition we assign spin $5/2^+$ to the 0.0 keV level in ^{111}Tc and propose that this level is the head of the yrast band, corresponding to the $5/2^+[422]$ proton configuration. This

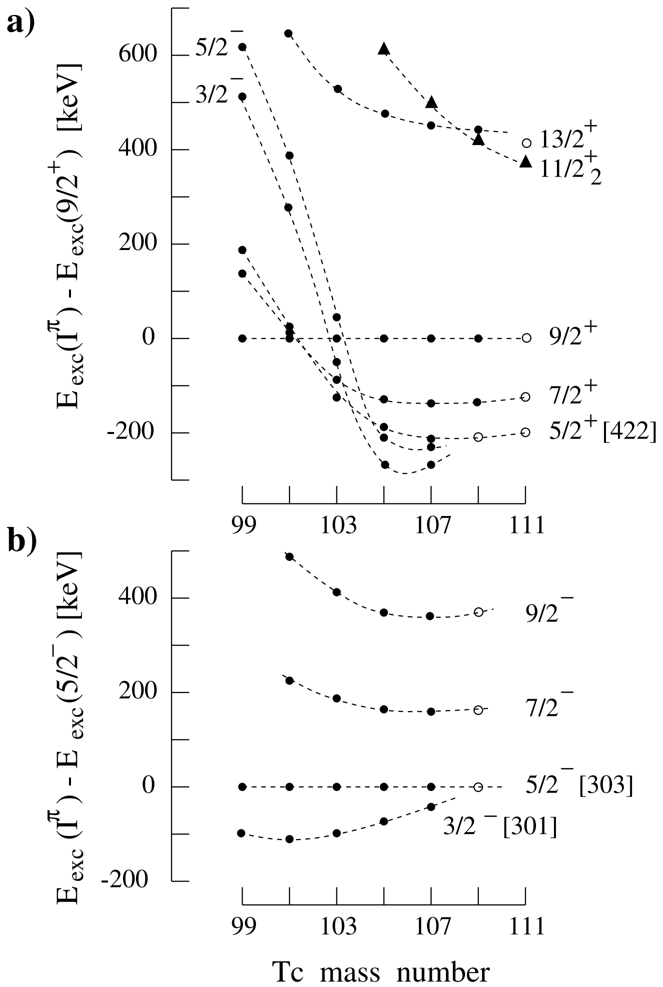

Fig. 6. Excitation energies of various levels in odd-A Tc isotopes, shown relative to the $9/2^+$ yrast level. The data are from this work and refs. [11–14,19]. The dashed line is drawn to guide the eye.

is consistent with the systematics of $5/2^+[422]$ bands in $^{99-109}$Tc isotopes, shown in fig. 6a, where the 0.0 keV and the 67.0 keV levels (open circles) smoothly extend the trend for the $5/2^+$ and $7/2^+$ keV excitations, respectively. In this figure we included the $5/2^+$ level in ^{109}Tc (open circle). This level, suggested previously as $5/2^-$ [4], was assigned positive parity, based on the α_K conversion coefficient, found for the 69.0 keV transition in ^{109}Tc in our recent study [14].

It is an interesting question to ask if the $5/2^+[422]$ proton configuration forms the ground state in ^{111}Tc. Figure 6a does not provide a definite answer to this question due to the lack of data for the $3/2^-$ and $5/2^-$ excitations in both the ^{109}Tc and ^{111}Tc nuclei. The $5/2^-[303]$ orbital is observed systematically in the lighter Tc isotopes, $^{99-107}$Tc. The excitation energy of this configuration, calculated relative to the $9/2^+$ band heads in these isotopes is shown in fig. 6a. This picture suggests that the $5/2^-[303]$ orbital is expected near the Fermi level also in ^{109}Tc and ^{111}Tc.

We found a $5/2^-$ band in ^{109}Tc [14]. Figure 6b shows excitation energies in $5/2^-[303]$ bands of Tc isotopes with the new data for ^{109}Tc included. The new band in ^{109}Tc fits the systematics well, which supports its interpretation as the $5/2^-[303]$ proton configuration. No decay of this band was seen, however. Our observational limit for an $E1$ transition from the $5/2^-[303]$ level to the $5/2^+[422]$ band head in ^{109}Tc is about 20 keV. Therefore, in ^{109}Tc the $5/2^-[303]$ band head either forms the ground state or is placed very close to it. This is consistent with the trend seen in fig. 6a and b, which suggests that the $3/2^-[301]$ excitation, which forms ground states in ^{105}Tc and ^{107}Tc, is expected to be very close to the $5/2^-[303]$ level in ^{109}Tc and probably is located above the $5/2^-[303]$ level in ^{111}Tc. In ^{111}Tc neither the $3/2^-[301]$ nor the $5/2^-[303]$ excitations were found. This suggests they are non-yrast in this nucleus. Therefore, the likely ground-state configuration in ^{111}Tc corresponds to the $5/2^+[422]$ orbital. What we can definitely say is that the spin of the ground state in ^{111}Tc is not (7/2, 9/2) as reported in the nuclear data base [12].

Since the $5/2^+$ yrast band in ^{111}Tc is very similar to analogous bands in lighter Tc isotopes we conclude that it corresponds to the $5/2^+[422]$ proton excitation in a prolate-deformed potential, as observed in lighter Tc isotopes. It is also worth noting that the deformation in this band does not decrease compared to ^{109}Tc. In ^{111}Tc the first in-band $E2$ transition has lower energy than in ^{109}Tc, as seen in fig. 6a, where we show excitation energies of the $13/2^+$ yrast levels in Tc isotopes. This observation suggests that the deformation region extends beyond $N = 68$.

There are excitations seen in ^{111}Tc, which suggest that the deformation above $N = 68$ may be oblate. Several low-energy levels found in $^{105-109}$Tc [3,11,14] and ^{111}Tc show features as observed in gamma-vibrational bands in ^{111}Rh and ^{113}Rh isotones [20]. In fig. 6a we show the $11/2^+_2$ levels in Tc isotopes. The dependence of their excitation energies on the neutron number closely resembles that of $11/2^+_2$ levels in the Rh isotopes, interpreted as 2^+ gamma-vibrational states coupled to $7/2^+$ levels of yrast bands [20]. As in Rh nuclei, the excitation energy of the $11/2^+_2$ state in Tc isotopes quickly decreases with the neutron number. This could correspond to the expected transition from prolate to an oblate shape, predicted to become yrast at $N \geq 70$. A mixing between the gamma band and the yrast band in ^{111}Tc, where these two configurations are close to each other, could explain the apparent lowering of the $13/2^+$ yrast level in this nucleus.

After this work was completed, the authors of ref. [11] reported again on Tc nuclei [21], changing multipolarities of the 65.8 keV (^{107}Tc) and 69.4 keV (^{109}Tc) transitions from $E1$ to $M1 + E2$, as found in our studies [13,14].

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