CERN-INTC-2023-059 / INTC-P-673
26/09/2023

EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Laser spectroscopy of neutron-deficient thulium isotopes

September 26, 2023

B. Cheal[1], L. V. Rodríguez[2,3], S. Bai[4], K. Blaum[3], P. Campbell[5], K. Chrysalidis[6], R. F. García Ruíz[7], P. F. Giesel[8], R. Heinke[6], P. Imgram[9], K. Koenig[9], D. Lange[3], T. Lellinger[2,9], D. Lunney[10], K.M. Lynch[5], B.A. Marsh[6], P. Müller[9], W. Nazarewicz[11], R. Neugart[3,12], G. Neyens[13], L. Nies [2], W. Nörtershäuser[9], R. D. Page[1], P. Plattner[3], P. G. Reinhard[14], L. Renth[9], S. Rothe[6], R. Sánchez[15], Ch. Schweiger[3], L. Schweikhard[8], S. Stegemann[6], T. Stora[6], S. M. Wang[16], X. F. Yang[4], D. T. Yordanov[10]

[1] *Oliver Lodge Laboratory, University of Liverpool, UK.*
[2] *Experimental Physics Department, CERN, Geneva, Switzerland.*
[3] *Max-Planck-Institut für Kernphysik, Heidelberg, Germany.*
[4] *School of Physics and State Key Laboratory of Nuclear Physics and Technology, Peking University, Beijing, China.*
[5] *School of Physics and Astronomy, The University of Manchester, Manchester, UK.*
[6] *Engineering Department, CERN, Geneva, Switzerland.*
[7] *Massachusetts Institute of Technology, Cambridge, MA, USA.*
[8] *Institut für Physik, University of Greifswald, Greifswald, Germany*
[9] *Institut für Kernphysik, Technische Universität Darmstadt, Darmstadt, Germany.*
[10] *IJCLab, IN2P3/CNRS-Université Paris-Saclay, Orsay, France*
[11] *Department of Physics and Astronomy and FRIB Laboratory, MSU, USA.*
[12] *Institut für Kernchemie, Universität Mainz, Mainz, Germany.*
[13] *Instituut voor Kern- en Stralingsfysica, KU Leuven, Leuven, Belgium.*
[14] *Institut für Theoretische Physik II, Universität Erlangen-Nürnberg, Erlangen, Germany.*
[15] *GSI Helmholtzzentrum für Schwerionenforschung GmbH, Darmstadt, Germany.*
[16] *Fudan University, Shanghai 200438, People's Republic of China.*

Spokesperson: Bradley Cheal, bradley.cheal@liverpool.ac.uk
Co-Spokesperson: Liss Vázquez Rodríguez, liss.vazquez.rodriguez@cern.ch
Contact person: Liss Vázquez Rodríguez, liss.vazquez.rodriguez@cern.ch

Abstract: This proposal aims to study the neutron-deficient isotopes of thulium using laser spectroscopy. This is following 6 shifts of radioactive beam time that were awarded as part of INTC-I-245 (and INTC-I-246) to measure yields and the spectroscopic efficiency and sensitivity. We aim to study the mean-square charge radius and electromagnetic moments of isotopes down to ^{147}Tm. In doing so, this will constitute the first measurement of the mean-square charge radius of a proton emitting nucleus.

Requested shifts: 21 shifts of radioactive beamtime (as 1 run)

1 Physics Motivation

High-resolution collinear laser spectroscopy is an indispensable tool in the study of ground and isomeric state nuclear structure. Model independent access is provided to the nuclear magnetic dipole and electric quadrupole moments, the nuclear spin and the mean-square charge radius. It therefore constitutes a unique probe of single particle and collective phenomena. Of much recent theoretical attention have been attempts to reproduce trends and features of the mean-square charge radius across the nuclear chart simultaneously, through ongoing developments in Density Functional Theory (DFT) [1, 2, 3]. For example, around the calcium region [4, 5, 6], nickel region [7, 8], and tin region [9, 10]. The ability to reproduce phenomena such as the ubiquitous odd–even staggering along isotope chains, or upward kinks seen at shell closures have been used as a basis to assess and develop various functionals of a Skyrme and Fayans type. Still more recently, these have been applied to isotopes of palladium, marking the first detailed analysis of deformed open shell nuclei [11]. We now propose to extend these studies into the neutron-deficient rare earth region of the nuclear chart.

Nuclear existence is limited on the neutron-deficient side by the proton drip line, but how the structure of the nucleus changes as this is approached is one of the perennial questions of nuclear physics research. While the existence of proton emission as a decay mode is well established, the properties of the nuclear state as this happens is not so clear. Laser spectroscopy has yet to be performed on a proton emitter. Of primary interest would be the first measurement of the nuclear mean-square charge radius for such an isotope and for those in the neighbourhood. The spatial extent of the proton distribution of such a metastable state would be expected to increase. However, the amount of this increase will depend on the angular momentum content of the orbital occupied by the unstable proton and the proton separation energy. Theoretically, the description of the narrow proton resonance and its radius will require a coupling between DFT and an open-quantum system framework [12]. In the neutron-deficient rare earth region, the isotope ^{147}Tm is a promising candidate for study at ISOLDE, which has a 15% proton emission branch, while an alternative possibility would be ^{151}Lu. Elsewhere, the Liverpool group is also working towards a measurement of the proton emitter ^{53m}Co at JYFL, although this will be challenging.

Previous laser spectroscopic measurements of the ground states down to ^{157}Tm date from 1988 [13] with ^{153,154}Tm added in 2000 [14]. These measurements are summarised in table 1. None of the fundamental properties listed above are known below ^{153}Tm and nuclear spins are assigned only tentatively. Laser spectroscopy will enable these to be established unambiguously. The thulium chain additionally appears one rich in structural change, with the odd–A (even–N) isotopes displaying multiple changes in ground state spin. Understanding the systematics of the isomeric states, which are largely unexplored, may also help to theorise the nuclear spins of the isomeric states in ^{146}Tm [15, 16, 17], without which the proton spectra are hard to interpret. For lutetium, optical measurements only extend as far down as ^{161}Lu [18] and no such measurements in the wider region extend below the $N = 82$ shell closure. We therefore believe that the thulium isotopes en route to ^{147}Tm are of interest in themselves.

Similarly to the laser spectroscopy data available from ^{155}Tm towards the proton emitter candidate, the knowledge on binding energies is sparse. Due to the abundance of long-lived and possibly low-lying isomers, direct mass measurements are challenging as high resolving powers are required. At GSI, some ground state masses were measured by SHIPTRAP [19] and the ESR [20]. However, due to the low-lying nature of many isomers, it is not clear whether all states were resolved if present in the spectra. No experimental data on ground or isomeric states exists on ^{149,150}Tm, while several excitation energies for $^{151-157}$Tm were tentatively deduced from various decay experiments (see eg. AME2020 and ENSDF Database). This lack of data around ^{151}Tm prevents accurate determination of the strength of the $N = 82$ shell gap in the thulium chain and motivates direct mass measurements of $^{149-155}$Tm which will provide data to calculate the two-neutron separation energy S_{2n} and the two-neutron shell gap δS_{2n}. Furthermore, the wealth of isomeric states in almost all isotopes makes the region very interesting for benchmarking nuclear structure calculations.

2 August 2023 Beamtime

A Letter of Intent INTC-I-245 was submitted (see also INTC-I-246) and awarded six shifts of radioactive beam time. The purpose of this was two fold. Firstly to investigate the yields of neutron-deficient thulium isotopes, which had not been properly investigated since the installation of the PSB. Secondly, to identify a suitable spectroscopic transition and establish both the detection efficiency and appropriate sensitivity to nuclear properties.

A new design of target container (with Ta foils) was tested, in order to determine the yields with the PSB and also with a highly efficient RILIS scheme that has recently been developed for thulium. Members of the ISOLTRAP collaboration were able to do a fast analysis of the yields and isobaric contaminants. Unfortunately a brand new target unit was not available for these tests, lacked the requested mass marker, and had already deteriorated. Overwhelming levels of contamination were also observed for the most neutron-deficient isotopes, consisting of rare earth oxides. An attempt was made to break up these molecules using the RILIS "blaze laser" but this only exacerbated the problem.

Proceeding with the laser spectroscopy tests, the 313.2 nm $J = 4 \to J = 5$ ground state line of the thulium ion was explored. Although several other lines were accessible, this line proved efficient, with a spectroscopic efficiency of 1 photon per 1400 ions. This is without time for a full optimisation. Very intense resonances were observed for isotopes even for 900 fA of isobaric contamination being measured on the Faraday cup and no visible RILIS laser on/off effect. Measuring the stable isotope allowed the magnetic field at the centre of the nucleus (for both the ionic ground and excited states) to be calibrated, showing

A	I^π	$\tau_{1/2}$	μ (μ_N)	Q_s (b)	$\delta\langle r^2\rangle$
175	$(1/2^+)$	15.2 m	✓	—	✓
174	$(4)^-$	5.4 m	✓	✓	✓
174m	0^+	2.29 s	—	—	✓
173	$(1/2^+)$	8.24 h	✓	—	✓
172	2^-	63.6 h	✓	✓	✓
171	$1/2^+$	1.9 y	**−0.230(4)**	—	✓
170	1^-	128.6 d	**+0.247(4)**	**+0.74(2)**	**+0.070(14)**
169	$1/2^+$	stable	−0.2310(15)	—	0
168	3^+	93.1 d	**+0.226(11)**	**+3.23(7)**	**−0.084(4)**
167	$1/2^+$	9.25 d	**−0.197(2)**	—	**−0.126(3)**
166	2^+	7.7 h	**+0.092(1)**	**+2.14(3)**	**−0.209(3)**
166m	(6^-)	340 ms	?	?	?
165	$1/2^+$	30.06 h	**−0.139(2)**	—	**−0.250(2)**
164	1^+	2.0 m	**+2.37(3)**	**+0.706(51)**	**−0.347(6)**
164m	6^-	5.1 m	✓	✓	✓
163	$1/2^+$	1.810 h	**−0.082(1)**	—	**−0.404(2)**
162	1^-	21.70 m	**+0.068(8)**	**+0.69(3)**	**−0.537(5)**
162m	5^+	24.3 s	?	?	?
161	$7/2^+$	30.2 m	**+2.39(2)**	**+2.90(7)**	**−0.632(3)**
160	1^-	9.4 m	**+0.156(18)**	**+0.582(44)**	**−0.741(4)**
160m	$5^?$	74.5 s	✓	✓	✓
159	$5/2^+$	9.13 m	**+3.408(34)**	**+1.93(7)**	**−0.850(4)**
158	2^-	3.98 m	**+0.042(17)**	**+0.74(11)**	**−1.002(7)**
158m	(5^+)	≈20 s	?	?	?
157	$1/2^+$	3.63 m	**+0.475(15)**	—	**−1.093(8)**
156	2^-	83.8 s	✓	✓	✓
155	$11/2^-$	21.6 s	✓	✓	✓
155m	$1/2^+$	45 s	?	?	?
154	(2^-)	8.1 s	−1.14(2)	+0.4(9)	−1.486(19)
154m	9^+	3.30 s	+5.91(5)	−0.2(4)	−1.522(15)
153	$(11/2^-)$	1.48 s	+6.93(11)	+0.5(10)	−1.615(31)
153m	$(1/2^+)$	2.5 s	?	?	?
152	$(2)^-$	8.0 s	?	?	?
152m	$(9)^+$	5.2 s	?	?	?
151	$(11/2^-)$	4.17 s	?	?	?
151m	$(1/2^+)$	6.6 s	?	?	?
150	(6^-)	2.20 s	?	?	?
149	$(11/2^-)$	0.9 s	?	?	?
148	(10^+)	0.7 s	?	?	?
147	$11/2^-$	0.58 s	?	?	?

Table 1: Nuclear moments and charge radii known prior to this work. Quantities in bold were remeasured during the limited beam time attached to LoI-245 and new measurements are indicated with ticks. Previously unknown long-lived isomeric states in ^{161}Tm and ^{168}Tm were also observed.

that the hyperfine structure has high sensitivity to the magnetic dipole moment, and also leading to spectra that are clearly resolved. Likewise, measuring the hyperfine structure of a radioactive isotope ($I > 1/2$) confirmed sensitivity to the quadrupole moment. Measuring a third isotope enabled calibration of the atomic field and mass shift factors, showing a strong sensitivity of the isotope shift to the change in nuclear mean-square charge radius.

Although six shifts were awarded, we were beset by facility issues including a loss of protons for a significant portion of the time, mainly due to a PSB intervention, followed by a reduced proton intensity. We also encountered an issue with our setup which frequently limited us to only three rather than four PMTs being in use, which can be fixed offline. Nevertheless, the strength of the spectroscopic scheme meant that we were able to perform measurements of several isotopes and make the most of the available time.

Table 1 shows nuclear properties obtained prior to these tests. All quantities indicated in bold font were remeasured during the August tests. Despite the limited beam time the checkmarks indicate properties that have been measured for the first time, and analysis is underway. Additional measurements were made of ground state structures on both the neutron-deficient and neutron-rich sides (the latter up to ^{175}Tm despite the use of a Ta target) and also several isomeric states were measured for the first time. In the cases of ^{161}Tm and ^{168}Tm the states measured were not known experimentally to be isomeric. Measuring these with ease following many hours of protons not being available show them to be very long-lived.

3 Experimental Proposal

Mass measurements during the August 2023 beamtime showed that large levels of contamination are present for the very neutron-deficient isotopes. Other yield measurements in 2023 tested a LIST ion source. This uses a repeller electrode to suppress the release of contaminant ions before ionisation of the thulium atoms with RILIS takes place. While the yields of the desired element are generally lower, the technique greatly enhances beam purity. Too much contamination can cause an overfilling of ISCOOL.

Figure 1 shows a preliminary analysis of the thulium yield measurements using LIST. This indicates a yield of ^{147}Tm of approximately 400/s. With much higher beam purity and given the strength of the ionic transition, measurements with such count rates are feasible. For the 313.2 nm line, 100% of the decays occur back to the ground state allowing multiple excitations to occur.

We propose to continue measurements of all isotopes down from ^{155}Tm to ^{147}Tm. Having calibrated the atomic factors, the shape and location of the hyperfine structures can be calculated from estimates of the nuclear properties. In particular, the trend of the $11/2^-$ ground states can be tracked from the new measurement of ^{155}Tm, through ^{153}Tm, and the odd-A isotopes to ^{147}Tm. Measurements of ^{154}Tm and ^{153}Tm will be re-measurements of literature values (with the hyperfine structures therefore easy to calculate and locate)

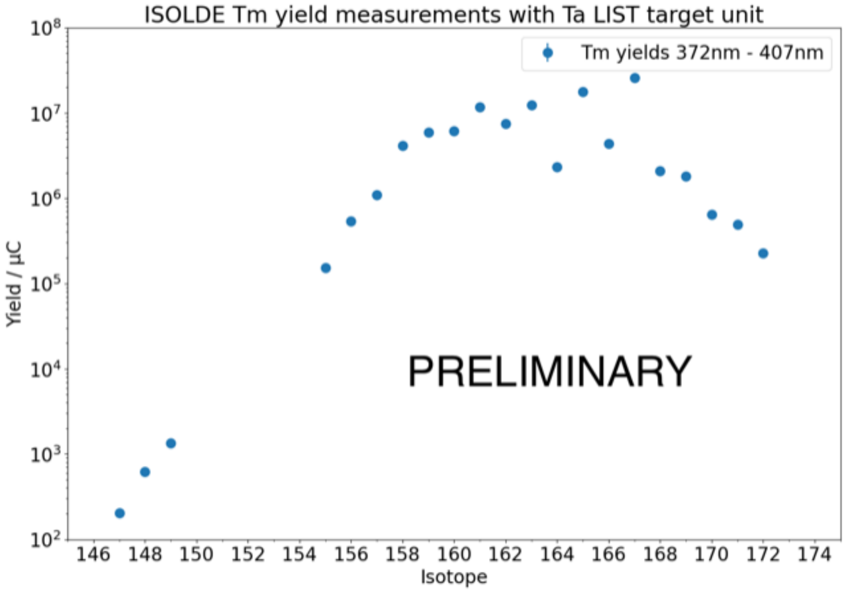

Figure 1: Analysis of measured thulium yields using a LIST ion source.

but with increased precision of the quadrupole moment and systematic errors on the radii minimised. For each isotope, typically three measurements of the location of the hyperfine structure are performed, with reference measurements of the stable isotope in between to ensure systematic errors are minimised.

Given the strength of the transition, expected purity of the beam but challenging yields, we request 1 shift to remeasure ^{155,154,153}Tm, 1 shift for each of ^{152}Tm and ^{151}Tm, 2 shifts for each of ^{150}Tm and ^{149}Tm, 3 shifts for ^{148}Tm and 8 shifts for ^{147}Tm.

While the strong contamination during the 2023 August beam time prevented effective use of the ISOLTRAP mass spectrometer for all requested cases, the use of the LIST will greatly reduce the contamination levels. This will enable the Multi-Reflection Time-of-Flight mass spectrometer (MR-ToF MS) to effectively separate the remaining contamination and the thulium ions of interest (up to intensity ratios of 10^3 to 1) for successive and pure Penning trap mass spectrometry. The Phase-Imaging Ion-Cyclotron-Resonance (PI-ICR) technique is fully capable of resolving all ground and isomeric states. Given the reported yields in Figure 1, we request 3 shifts for the mass measurements of ground- and isomeric states of $^{149-155}$Tm using the ISOLTRAP mass spectrometer, which is also used in support of the laser spectroscopy in terms of beam composition identification and intensity ratio measurements.

Summary of requested shifts: 21 shifts of radioactive beam plus 2 shifts of stable beam to optimise the setup.

References

[1] P.-G. Reinhard, W. Nazarewicz, *Phys. Rev. C* **105**, L021301 (2022).

[2] J. Hur, *et al.*, *Phys. Rev. Lett.* **128**, 163201 (2022).

[3] P.-G. Reinhard, W. Nazarewicz, *Phys. Rev. C* **106**, 014303 (2022).

[4] R. F. Garcia Ruiz, *et al.*, *Nature Physics* **12**, 594 (2016).

[5] A. J. Miller, *et al.*, *Nature Physics* **15**, 432 (2019).

[6] Á. Koszorús, *et al.*, *Nature Physics* **17**, 439 (2021).

[7] R. P. de Groote, *et al.*, *Nature Physics* **16**, 620 (2020).

[8] S. Malbrunot-Ettenauer, *et al.*, *Phys. Rev. Lett.* **128**, 022502 (2022).

[9] M. Hammen, *et al.*, *Phys. Rev. Lett.* **121**, 102501 (2018).

[10] C. Gorges, *et al.*, *Phys. Rev. Lett.* **122**, 192502 (2019).

[11] S. Geldhof, *et al.*, *Phys. Rev. Lett.* **128**, 152501 (2022).

[12] S. M. Wang, W. Nazarewicz, *Phys. Rev. Lett.* **126**, 142501 (2021).

[13] G. Alkhazov, *et al.*, *Nuclear Physics A* **477**, 37 (1988).

[14] A. E. Barzakh, *et al.*, *Phys. Rev. C* **61**, 034304 (2000).

[15] T. N. Ginter, *et al.*, *Phys. Rev. C* **68**, 034330 (2003).

[16] A. P. Robinson, *et al.*, *Eur. Phys. J. A* **25**, 155 (2005).

[17] M. N. Tantawy, *et al.*, *Phys. Rev. C* **73**, 024316 (2006).

[18] U. Georg, *et al.*, *Eur. Phys. J. A* **3**, 225 (1998).

[19] C. Rauth, *et al.*, *The European Physical Journal Special Topics* **150**, 329 (2007).

[20] Y. Litvinov, *et al.*, *Nuclear Physics A* **756**, 3 (2005).

DESCRIPTION OF THE PROPOSED EXPERIMENT

The experimental setup comprises: COLLAPS and ISOLTRAP.

Part of the	Availability	Design and manufacturing
COLLAPS installation	☒ Existing	☒ To be used without any modification
ISOLTRAP setup	☒ Existing	☒ To be used without any modification

HAZARDS GENERATED BY THE EXPERIMENT

Hazards named in the document relevant for the fixed COLLAPS installation. The ISOLTRAP setup has safety clearance, the memorandum document 1242456 ver.1 "Safety clearance for the operation of the ISOLTRAP experiment" by HSE Unit is released and can be found via the following link: *https://edms.cern.ch/document/1242456/1*.