EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Laser spectroscopy of neutron-deficient thulium isotopes

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Abstract: In proposal INTC-P-673, 21 shifts were requested to perform high-resolution laser spectroscopy of neutron-deficient thulium isotopes. Among several motivations the ultimate physics goal is to reach the proton emitter ^{147}Tm. The INTC recommended the award of 13 shifts (IS740) to begin measurements down to ^{148}Tm and requested an Addendum then be submitted for additional time. Here we report on the beam time of September 2024 and seek the required shifts.

Summary of requested shifts: 18 shifts, (split into 1 run over 1 year)

1 Introduction

This proposal aims to perform laser spectroscopy of neutron-deficient isotopes of thulium, to obtain precise model-independent values for the nuclear magnetic dipole moments, electric quadrupole moments and mean-square charge radii of the ground and isomeric states. Additionally mass measurements were proposed with the added advantage of using ISOLTRAP to assess beam purity. An initial exploration as part of LOI INTC-I-245, while very fruitful in terms of measurements, revealed that beams of isotopes below ^{155}Tm suffered from overwhelming contamination, identified using ISOLTRAP as primarily rare-earth oxides. We therefore proposed the use of a LIST ion source to suppress the isobars even though the yields of the thulium isotopes are also reduced by around two orders of magnitude.

The physics cases were outlined in the original proposal (INTC-P-673) and in principle endorsed by the INTC. In brief, we wish to extend measurements of the nuclear moments and mean-square charge radii into this region of the nuclear chart for the first time to provide a stringent test of Density Functional Theory (DFT) calculations [1, 2, 3]. Ongoing developments have been the subject of intense theoretical and experimental efforts to calculate nuclear radii across the nuclear chart including the calcium [4, 5, 6], nickel [7, 8] and tin regions [9, 10], to deformed open shell nuclei [11] and now also the nuclear moments [12].

The primary aim is to measure moments and, in particular, the mean-square charge radius of a proton emitting nucleus for the first time. The spatial extent of the proton distribution of such a 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. The isotope ^{147}Tm, which has a 15% proton emission branch, has been identified as the most promising candidate for measurement at ISOLDE. Theoretically, the description of the narrow proton resonance and its radius will require a coupling between DFT and an open-quantum system framework [13]. However, it is important to measure the systematics of the radii leading down to ^{147}Tm in order to reliably assess the increase above the trend, particularly beyond the $N = 82$ shell closure where the signature “upward kink” in the course of the charge radii may be expected. Meanwhile the systematics of the isomeric states, the spins of which can be determined through laser spectroscopy, may help to theorise the nuclear spins of states in ^{146}Tm [14, 15, 16], without which the proton spectra are hard to interpret. The multiple changes in ground state spin along the thulium chain already point to there being much structural change. Finally, precision mass measurements below ^{155}Tm are sparse, with the wealth of low-lying isomeric states complicating existing data but making the region interesting for complementary benchmarking of nuclear structure calculations. From this can be calculated the two-neutron separation energy S_{2n} and the two-neutron shell gap δS_{2n}, allowing a determination of the strength of the $N = 82$ shell gap.

2 Summary of the 2024 beam time

During August 2023 only 4 shifts were required to select an efficient optical transition in the thulium ion (the 313.2 nm $J = 4 \rightarrow J = 5$ line from the ground state), calibrate its atomic constants for extracting the nuclear moments and mean-square charge radii and perform measurements of all isotopes from ^{175}Tm (a reasonable upper limit for a Ta target) down to ^{155}Tm. Moreover, several isomeric states were measured for the first time. In a few cases their existence had not been established or the assigned spin was shown to be incorrect.

Following the full proposal, 13 shifts of beam time were scheduled in September 2024. A tantalum foil target was again used but this time with a LIST ion source. This resulted in a substantial and critical suppression of the isobaric contamination but unfortunately also reduced the yields to lower than expected values. However, the spectroscopic transition used turned out to be far more efficient than previously thought and was established to yield 1 photon per every 50 ions. This transition is also closed, meaning that it does not branch out and permits multiple laser excitations. In principle the COLLAPS beam line could be extended in the future to further exploit this.

Measurements were made of ^{155}Tm which were a substantial improvement on the previous beam time, and the isomeric state could be measured for the first time. Unfortunately, after only a day into the run, the target heating circuit then failed. A new target was then quickly assembled and placed online. We are very grateful to TISD and the operators for achieving this. Our measurements continued, including measurements of ground and isomeric states of both ^{154}Tm and ^{153}Tm. In addition, ^{162}Tm and ^{164}Tm were measured more extensively to understand additional peaks that had been observed previously and which relate to isomeric states. A second measurement was also taken of ^{160}Tm after the spin of the isomeric state in literature was revealed to be incorrect during the 2023 run.

Yield checks when the first target was in position gave a total flux of 700 ions/μC for ^{152}Tm and 200 ions/μC for ^{151}Tm. Indeed a measurement of the ^{152}Tm ground state was obtained with ease, and two peaks of the weakly-produced isomeric state were identified. Only three peaks are required for the three unknowns of the magnetic dipole moment, electric quadrupole moment and mean-square charge radius. While considering whether to zoom in on the third peak for extra statistics, initial scans were made of ^{151}Tm. Again, clear peaks were identified in the spectra. Example spectra taken during the beamtime are shown in Figure 1. Unfortunately, it became apparent that during the measurement of these last two isotopes, the target had degraded significantly, affecting shorter-lived isotopes in particular. A tenfold reduction in the yield of ^{152}Tm to 70 ions/μC was concluded and yet the stronger ground state peaks could still be measured with ease.

In these circumstances, the remainder of the beam time was passed to RILIS for a preliminary exploration of in-source spectroscopy tests with LIST. The degradation of the target was also confirmed at this time. ISOLTRAP was unfortunately not operational due to a technical fault which may have been related to the power interruption that affected the CERN site and wider area a week or so before.

3 Request of additional shifts

Although the yields for the lighter isotopes were not firmly established, it is clear that the measurements of ^{152m}Tm and ^{151g,m}Tm will be achieved. This marks the shell closure. It is also likely that ^{150}Tm will be possible, and with a half life of 2.2 s does not require proton triggering of the data acquisition and ISCOOL bunch release. We request 6 shifts for making these measurements, in addition to the 3 shifts that were previously requested for mass measurements of $^{149-155}$Tm, where the PI-ICR method will be capable of resolving all ground and isomeric

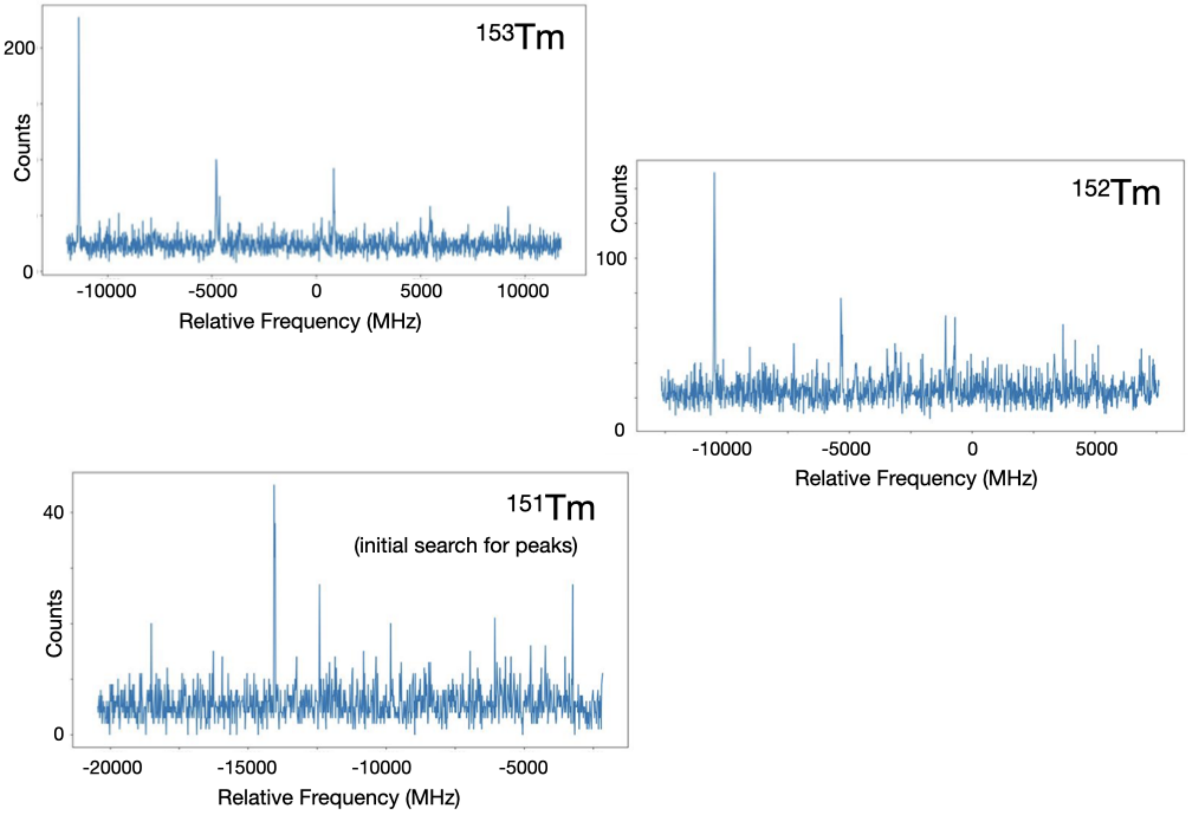

Figure 1: Example spectra of ^{153}Tm and ^{152}Tm taken in the 2024 beamtime. First peak search in ^{151}Tm.

states. As before, ISOLTRAP will also be able to serve as a diagnostic tool for the beams from (PI)LIST whilst heading towards ^{147}Tm.

For further laser measurements, although COLLAPS has not identified a definite end-point of which subsequent isotopes are feasible, the yields for ^{149,148,147}Tm beyond this point remain uncertain. In-source measurements have traditionally been higher in sensitivity but suffer from low resolution. In-source measurements with PI-LIST may offer an appropriate compromise. Although the sensitivity will be higher than that of collinear laser spectroscopy in its current form, the resolution will remain significantly lower, but with linewidths improved to the order of a few 100 MHz [17]. This can lead to peaks not being resolved, which is particularly problematic where there are uncertainties regarding the spin (or in at least one case where the "definite" spin assignment proved to be incorrect).

Analysing such spectra is problematic as the hyperfine peak locations depend on five parameters, those being the centroid and the hyperfine A and B coefficients (which relate to the magnetic dipole and electric quadrupole moments, respectively) for both the upper and lower states of the atomic transition. However, this is simplified once the hyperfine coefficients are measured for one isotope, as the ratio between upper and lower states of the transition is constant (isotope independent). We therefore propose to use COLLAPS to perform a re-measurement of just two radioactive isotopes and the single stable isotope as a reference, to ascertain these two ratios with precision, for the atomic transition that will form the first step of the PI-LIST scheme. A radioactive isotope is required since the stable isotope ^{169}Tm is spin $I = 1/2$ and therefore has no quadrupole moment or hyperfine B coefficients. This will also serve to precisely calibrate all atomic factors which are necessary to relate the isotope shifts and A and B parameters to changes in mean-square charge radius, magnetic dipole moments and electric quadrupole moments for the atomic line. Most likely this will be the 372 nm atomic transition. Since the (ionic) line used for the collinear work is 313 nm, these laser systems can be set up in parallel since one requires a frequency-doubled titanium sapphire laser and the other a frequency-doubled dye laser. These measurements would only take 1 shift since isotopes closer to stability may be chosen (or even shortly after protons have been switched off).

Figure 2 shows the measured yield values for thulium isotopes using LIST, following some uncertainty. The yields for the lightest isotopes are an extrapolation only, but are commensurate with the subsequent measurements for ^{152}Tm and ^{151}Tm. It therefore remains to be seen what yields may ultimately be achieved and be measurable using PI-LIST (or COLLAPS). An additional efficiency loss of around a factor 10 is expected when going from standard collinear LIST to PI-LIST, i.e. high-resolution, operation mode. Additionally to exploiting PI-LIST, using the ISOLDE decay station (IDS) as a sophisticated detector setup would aid in suppression of potential remaining ion background that can not be filtered out in single ion counting. Explorations with LIST on remaining contamination in this mass regime during the recent experiment revealed remaining rates of the order of 10 to 100 non-Tm ions per second, which would make scanning of structures with Tm ion peak rates around 1 ion/s challenging. The gain in selectivity by using decay tagging is expected to outmatch additional efficiency loss. Favorable gamma-ray multiplicities following the decay of the isotopes of interest, as well as the considerably longer half-lives of the contamination identified by ISOLTRAP during LoI 245 give additional confidence in this approach. Joint experimental campaigns of RILIS laser scanning and IDS detection are well established at ISOLDE [18, 19], and have proved the feasibility of low-yield experiments using this technique.

The four components of the proposal are therefore:

1. Collinear laser spectroscopy of ^{152m,151g,151m,150}Tm using the ionic transition.
2. Collinear laser spectroscopy of two radioactive isotopes on an atomic line to calibrate A and B ratios.
3. Stopping at ^{150}Tm to attempt spectroscopy of ^{149}Tm onwards using PI-LIST and IDS.
4. Use ISOLTRAP for mass measurements from ^{155}Tm downwards, as well as a useful diagnostics tool of the PI-LIST beams.

In summary we request 3 shifts for ^{152m,151g,m}Tm and 3 shifts ^{150}Tm for with COLLAPS (ionic line), 3 shifts for the mass measurements of $^{149-155}$Tm, 1 shift for spectroscopy of two radioactive isotopes (atomic line) using COLLAPS to calibrate atomic factors and hyperfine ratios, 1 shift of setup and optimization of beam transport and PI-LIST parameters for measurements with IDS, 4 shifts for hyperfine structure and isotope shift measurements with PI-LIST and IDS on ^{150,149}Tm, and finally 3 shifts of exploration towards ^{147}Tm.

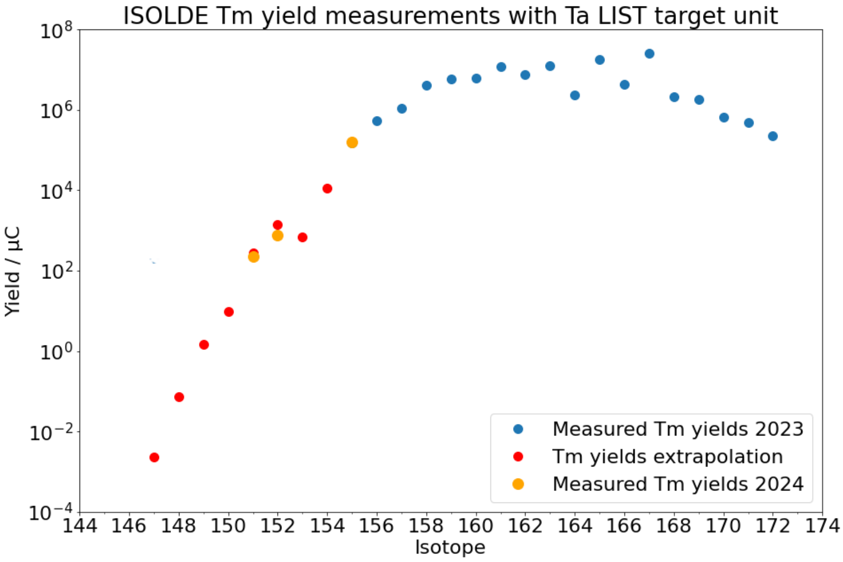

Figure 2: Analysis of measured thulium yields using a Ta LIST target unit. (J. Wessolek, private communication.)

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4 Details for the Technical Advisory Committee

4.1 General information

Describe the setup which will be used for the measurement. If necessary, copy the list for each setup used.

- ☒ Permanent ISOLDE setup: *COLLAPS, ISOLTRAP, IDS*
  - ☒ To be used without any modification
  - ☐ To be modified: *Short description of required modifications.*
- ☐ Travelling setup (*Contact the ISOLDE physics coordinator with details.*)
  - ☐ Existing setup, used previously at ISOLDE: *Specify name and IS-number(s)*
  - ☐ Existing setup, not yet used at ISOLDE: *Short description*
  - ☐ New setup: *Short description*

4.2 Beam production

For any inquiries related to this matter, reach out to the target team and/or RILIS (please do not wait until the last minute!). For Letters of Intent focusing on element (or isotope) specific beam development, this section can be filled in more loosely.

- Requested beams:

Isotope	Production yield in focal point of the separator (/μC)	Minimum required rate at experiment (pps)	$t_{1/2}$
Isotope 1 Isotope 2 Isotope 3			

- Full reference of yield information (*J. Wessolek, private communication*)
- Target - ion source combination: Ta foil target - PI-LIST ion source
- RILIS? *Yes*
  - ☒ Special requirements: *PI-LIST, narrow-band laser scanning*
- Additional features?
  - ☐ Neutron converter: (*for isotopes 1, 2 but not for isotope 3.*)
  - ☐ Other: (*quartz transfer line, gas leak for molecular beams, prototype target, etc.*)
- Expected contaminants: *Isotopes and yields*
- Acceptable level of contaminants: (*Not sensitive to stable contaminants (IDS), limited by ISCOOL overfilling (COLLAPS)*)
- Can the experiment accept molecular beams? *No*
- Are there any potential synergies (same element/isotope) with other proposals and LOIs that you are aware of?

4.3 Shift breakdown

The beam request only includes the shifts requiring radioactive beam, but, for practical purposes, an overview of all the shifts is requested here. Don't forget to include:

- Isotopes/isomers for which the yield need to be determined
- Shifts requiring stable beam (indicate which isotopes, if important) for setup, calibration, etc. Also include if stable beam from the REX-EBIS is required.

An example can be found below, please adapt to your needs. Copy the table if the beam time request is split over several runs.

Summary of requested shifts:

With protons	Requested shifts
Yield measurement of isotope 1 Optimization of experimental setup using isotope 2 Data taking, isotope 1 Data taking, isotope 2 Data taking, isotope 3 Calibration using isotope 4	
Without protons	Requested shifts
Stable beam from REX-EBIS (after run) Background measurement	

4.4 Health, Safety and Environmental aspects

4.4.1 Radiation Protection

- If radioactive sources are required:
  - Purpose?
  - Isotopic composition?
  - Activity?
  - Sealed/unsealed?
- For collections:
  - Number of samples?
  - Activity/atoms implanted per sample?
  - Post-collection activities? (*handling, measurements, shipping, etc.*)

4.4.2 Only for traveling setups

- Design and manufacturing
  - ☒ Consists of standard equipment supplied by a manufacturer
  - ☐ CERN/collaboration responsible for the design and/or manufacturing
- Describe the hazards generated by the experiment:

Domain	Hazards/Hazardous Activities		Description
Mechanical Safety	Pressure	☐	[pressure] [bar], [volume][l]
	Vacuum	☐	
	Machine tools	☐	
	Mechanical energy (moving parts)	☐	
	Hot/Cold surfaces	☐	
Cryogenic Safety	Cryogenic fluid	☐	[fluid] [m3]
Electrical Safety	Electrical equipment and installations	☐	[voltage] [V], [current] [A]
	High Voltage equipment	☐	[voltage] [V]
Chemical Safety	CMR (carcinogens, mutagens and toxic to reproduction)	☐	[fluid], [quantity]
	Toxic/Irritant	☐	[fluid], [quantity]
	Corrosive	☐	[fluid], [quantity]
	Oxidizing	☐	[fluid], [quantity]
	Flammable/Potentially explosive atmospheres	☐	[fluid], [quantity]
	Dangerous for the environment	☐	[fluid], [quantity]
Non-ionizing radiation Safety	Laser	☐	[laser], [class]
	UV light	☐	
	Magnetic field	☐	[magnetic field] [T]

Workplace	Excessive noise	☐	
	Working outside normal working hours	☐	
	Working at height (climbing platforms, etc.)	☐	
	Outdoor activities	☐	
Fire Safety	Ignition sources	☐	
	Combustible Materials	☐	
	Hot Work (e.g. welding, grinding)	☐	
Other hazards			