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

First online laser spectroscopy study of promethium isotopes

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Abstract: We propose to perform the first online, in-source laser spectroscopy study of the long chain of $^{132-153}$Pm ($N = 71 - 92$), for which only $^{143-147}$Pm were previously studied off-line. From them, changes in nuclear mean-squared charge radii and electromagnetic moments will be extracted. The data will provide insight into three phenomena: a potentially large and sharp transition from near-spherical ground states to deformed ones with $\beta_2 = 0.3 - 0.4$ at $N \sim 75$, predicted by theory; how the $N \sim 90$ shape transition manifests in the ground state properties of the promethium chain; and probe the possible existence of predicted triaxial and octupole degrees of freedom in the neutron-deficient and neutron-rich regions, respectively.

Summary of requested shifts: [23] shifts, (split into [1] runs over [1] years)

1 Motivation for studying promethium isotopes and previously known data

The structure of the rare-earth nuclei near the $Z = 62$ proton midshell are known for their transitionary nature. Whilst isotopes near $N = 82$ have spherical shapes, changes to strongly deformed ground states have been observed on both sides of $N = 82$. For instance, a well-known shape transition occurs at $N \sim 90$ and is clearly seen in the charge radii of neutron-rich europium and terbium chains shown in Fig. 1(a) [1]. While many chains have been extensively studied using laser spectroscopy, a unique exception is the promethium chain ($Z = 61$), for which the known isotopes span a wide range of neutron numbers ($N = 67 - 102$) above and below the spherical $N = 82$ shell closure.

Currently, there are only limited data available on the ground-state properties of the promethium isotopes. Indeed, the nuclear spin and parity (I^π) assignments across the chain are uncertain, and the magnetic dipole (μ) and electric quadrupole (Q) moments were measured for the long-lived isotopes $^{143-149,151}$Pm only [2–7]. The results from these studies, made using various complementary methods in the 1960s and 70s, indicated a large increase in the quadrupole moment of ^{151}Pm ($N = 90$) [3]. However, apart from ^{147}Pm, these data have large uncertainties of order $\gtrsim 10\%$. Only two precision hyperfine

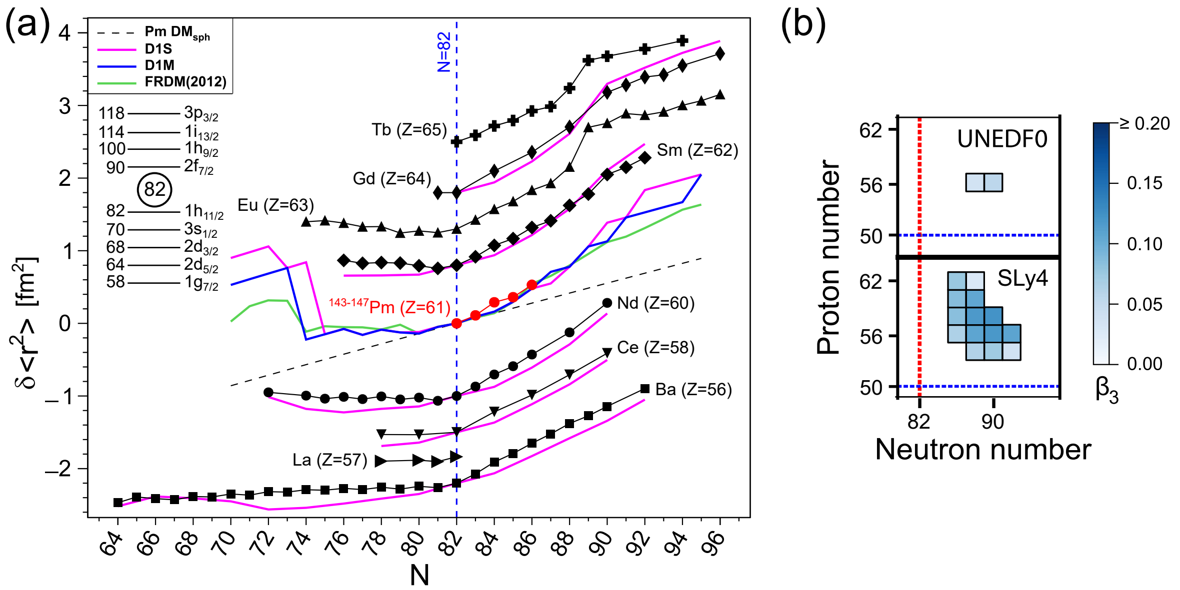

Figure 1: (a) Experimental nuclear mean-squared charge radii in the rare-earth region (black data points) [1] including the $^{143-147}$Pm (red data points) [8], and schematic spherical shell model energy-level diagram for the region. The isotopic chains are arbitrarily offset from each other for clarity. The black dashed line represents the $\delta\langle r^2\rangle$ values for spherical promethium nuclei calculated using the Droplet Model [9, 10], the green, pink and blue line are those based on β_2 predictions from the FRDM [11], and HFB calculations using the D1M [12] and D1S [13] interactions, respectively. (b) Ground state β_3 values predicted by DFT calculations using the UNEDF0 and SLy4 interactions (adapted from [14]). We propose to study the properties of $^{132-153}$Pm ($N = 71 - 92$).

structure (hfs) and isotope shift (IS) studies have been made using laser techniques, on the long-lived isotopes $^{143-147}$Pm [8, 15]. These isotopes were observed to have properties consistent with spherical configurations [see Fig. 1(a)].

Whilst strong departures from spherical trends have been observed in the proton-rich europium, samarium and neodymium isotopes, the ground and isomeric state properties of the promethium isotopes represent a clear gap in our knowledge in this region. Yet, as outlined below, the promethium isotopes are of interest for testing the sometimes conflicting predictions made by several different theoretical studies.

1.1 Neutron-rich isotopes

1.1.1 N=90 shape transition

The $N = 86 - 90$ region is well known for a transition from spherical to well-deformed ground states with $N > 88$ [16]. This rapid change was suggested as evidence for a nuclear phase transition when crossing $N = 90$ [17], or even as a case of "phase coexistence" [18]. It has been linked to the nearby "nearly doubly-magic" ^{146}Gd ($Z = 64$, $N = 82$), for which an increased proton shell gap between the $\pi d_{5/2}$ and $\pi h_{11/2}$ orbitals near $N = 82$ reduces the effective proton valence space [19]. This shell gap decreases as the $\nu h_{9/2}$ orbital fills with increasing neutron number, allowing for strong $p - n$ interactions with $\pi h_{11/2}$ protons.

The nature of this transition between spherical and deformed shapes has an observed Z dependence, with the promethium isotopes lying directly on the border between smooth, weak transitions observed for $Z = 56$, 58 and 60, and a larger jump-like change for $Z = 62 - 67$ [see Fig. 1(a)]. Hartree-Fock Bogoliubov (HFB) calculations using the D1S interaction [13] perform reasonably well at modelling the trends in changes in mean-squared charge radii ($\delta\langle r^2\rangle$) in even-even isotopes in the region [see Fig. 1(a)], and predict a smooth but rapid onset of deformation in the promethium chain from near-spherical ground states near $N = 82$, to large deformations of $\beta_2 \sim 0.3 - 0.4$. A similar onset is predicted by Finite Range Droplet Model (FRDM) calculations [11], albeit to smaller deformations of $\beta_2 \sim 0.25$.

Therefore, new IS and hfs measurements of $^{148-153}$Pm ($N = 87-92$) are critical for filling the current gaps in the data and testing the model predictions. Complementary data on I and μ extracted from the hfs measurements will provide additional insight into the configurations of the odd particles. These will provide strong additional constraints for testing the predictions of theory in this region of rapidly evolving nuclear structure.

1.1.2 Octupole collectivity

In recent years, the discovery of nuclear ground states with exotic octupole (i.e. pear shaped) deformations [20] has been a topic of particular interest for nuclear structure physics [21]. Octupole collectivity is expected in regions of the nuclear chart where single-particle orbitals with $\Delta\ell = \Delta j = 3$ lie close to the Fermi surface. In the lanthanide region, the proximity of the low-lying $\pi d_{5/2}$ & $\pi h_{11/2}$ proton, and $\nu f_{7/2}$ & $\nu i_{13/2}$ neutron orbitals is expected to enhance octupole correlations, possibly leading to pear-shaped ground states. However, theoretical predictions vary on the extent of this region of octupole collectivity and the magnitude of the possible deformation, as illustrated by the differences between the UNEDF0 and SLy4 calculations shown in Fig. 1(b) [14].

Experimental signatures of octupole collectivity are typically associated with near-degenerate, parity-doublet rotational bands, and electric octupole and dipole transition strengths [$B(E3)$s and $B(E1)$s] that are enhanced by the asymmetric nuclear charge distributions. Complementary evidence can be found in μ and $\delta\langle r^2\rangle$ values extracted from hfs and IS measurements. As the parity of states in the intrinsic frame of asymmetric nuclei is not well-defined, mixing between states of opposite parities becomes possible. The μ value is sensitive to such mixing, and can therefore be used to identify the possible presence of octupole collectivity [22, 23]. An inversion in the odd-even staggering (iOES) systematics relative to the "normal" trend in $\delta\langle r^2\rangle$ has been observed in regions of expected octupole collectivity, such as those with $Z \sim 88$, $N \sim 132$ [24]. This inversion of the staggering was qualitatively explained by introducing $\beta_3 \neq 0$, assuming odd-N isotopes have larger octupole deformations than their even-N neighbours.

The promethium isotopes with $N \sim 88$ lie at the northern border of an expected region of octupole collectivity. An iOES in radii was previously observed in nearby $^{153-155}$Eu ($N = 90 - 92$) [25], which coincides with the observation of parity doublet bands in ^{154}Eu [26]. For the promethium chain, small but stable octupole deformations were predicted for $^{146-150}$Pm ($N = 85 - 88$) using the microscopic-macroscopic approach [11, 27]. Furthermore, parity-doublet bands were observed in in-beam studies of ^{151}Pm [28–30], however these data have been described both with [31] and without [32] octupole degrees of freedom. Measurements of μ, Q and $\delta\langle r^2\rangle$ would provide new information on the possible presence of octupole collectivity, and additional tests for the different models.

1.2 Proton-rich isotopes

1.2.1 N<75 shape transition

There are long-standing model predictions for a strongly-deformed region in the very light rare-earth isotopes [33, 34]. In particular, deformed shell model calculations indicate a large and sudden jump in deformation close to the proton drip line, with an especially pronounced transition between $N = 74$ and 75 in the promethium chain (see Fig. 1 of [35]). Modern FRDM and HFB codes also predict a dramatic shape transition for promethium isotopes, as shown in Fig. 1(a). However, whilst the HFB calculations predict a step-like change from β_2 ∼0.2 to ∼ 0.4 [13], those of the FRDM display a smoother, albeit still strong transition from $\beta_2 = 0.23$ to 0.33 [11].

Indeed, in-beam studies of $A \sim 130$ nuclides have observed band structures and excited state lifetimes consistent with a transition from sphericity near $N = 82$ to strong, presumably prolate ($\beta_2 > 0.25$) deformations in the $N < 75$ region [36–38]. However, the available $\delta\langle r^2\rangle$ and Q data are limited to moderately deformed cases, for $N > 74$ europium, $N > 76$ samarium, and $N > 72$ neodymium isotopes [39–43]. In each case, a gradual and smooth onset of deformation was observed with reducing N.

As shown in Fig. 1 of [35], the expected region of strong deformation starts at ^{135}Pm ($N = 74$, $T_{1/2} = 19$ s). Our proposed IS and hfs measurements will be used to determine the presence and magnitude of the jump in deformation, its precise location, and whether the ground states remain strongly deformed in the lightest cases as the predictions show in Fig. 1(b).

1.2.2 Triaxiality

Whilst the deformed nuclei with $N > 90$ are thought to possess axially symmetric prolate deformations, nuclei with $A = 130 - 140$ are known for exhibiting properties associated with triaxial degrees of freedom [44–46]. The ground state I^π, μ and Q values are sensitive to the degree of triaxiality. To illustrate this effect, results from particle+triaxial rotor model calculations [47] for ^{135}Pm ($N = 74$) are shown in Fig. 2, at a fixed quadrupole deformation of $\beta_2 \approx 0.2$. Here, the (a) excitation energy, (b) μ, (c) g-factor and (d) Q values of the lowest-lying $I^\pi = 3/2^+$, $5/2^+$ and $7/2^+$ states are plotted as function of the triaxial deformation parameter, γ. The results show that the ground state properties are sensitive to γ, with a change from $I^\pi = 5/2^+$ to $7/2^+$ for the ground state occurring at $\gamma = 35°$, for example, and significant variation in μ for $I^\pi = 3/2^+$ and $5/2^+$ states.

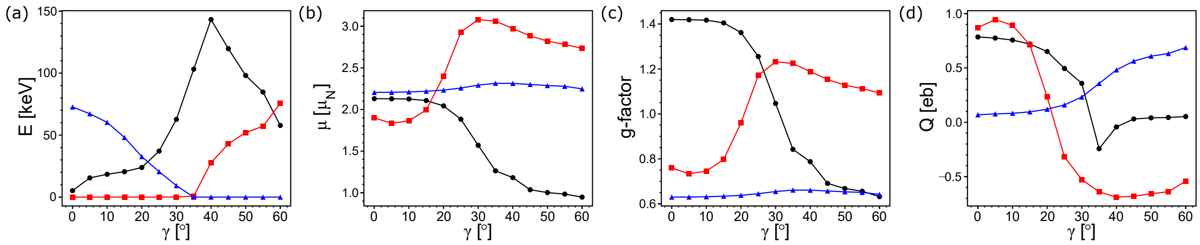

Figure 2: Particle+triaxial rotor model calculations for ^{135}Pm, plotting the (a) excitation energy, (b) μ, (c) g-factors and (d) Q values of lowest-lying $I^\pi = 3/2^+$ (black circles), $5/2^+$ (red squares), and $7/2^+$ (blue triangles) states as a function of the triaxial shape parameter, γ, assuming $\beta_2 \approx 0.2$.

However, the present I^π assignments for the neutron-deficient promethium isotopes are uncertain, and are primarily based on either systematics from neighbouring nuclei, or β-decay feeding patterns to states in neodymium daughters, the I^π assignments for which are also uncertain (see Table 1). Therefore, laser measurements from which nuclear moments and spins can be determined will probe possible triaxiality in promethium isotopes, and constrain the I^π assignments for the neodymium isotopes.

Table 1: Experimental ground-state spin and parity assignments taken from ENSDF for proton-rich samarium, promethium and neodymium isotopes.

	Ground state I^π assignment												
N	70	71	72	73	74	75	76	77	78	79	80	81	82
Sm ($Z = 62$)	0^+	$(5/2^+)$	0^+	$(3/2^+,5/2^+)$	0^+	$(9/2^-)$	0^+	$(9/2^-)$	0^+	$(1/2^+)$	0^+	$(3/2^+)$	0^+
Pm ($Z = 61$)	$(11/2^-)$	(3^+)	$(3/2^+)$	(2^+)	$(3/2^+,5/2^+)$	(5^-)	$(11/2^-)$		$(5/2^+)$	1^+	$5/2^+$	1^+	$5/2^+$
Nd ($Z = 60$)	0^+	$(5/2^+)$	0^+	$(7/2^+)$	0^+	$9/2^{(-)}$	0^+	$1/2^+$	0^+	$3/2^+$	0^+	$3/2^+$	0^+

2 Proposed measurements

We propose to perform in-source laser-spectroscopy studies to measure the hfs and IS of promethium isotopes. Based on our previous yield measurements from LOI246, we will be able to perform IS and hfs measurements from ^{132}Pm ($N = 71$) to ^{153}Pm ($N = 92$) using a Ta-foil plus LIST target module. Apart from ^{144}Pm which will be used as the reference isotope for our measurement, we will not repeat measurements of $^{143,145-147}$Pm as they were studied in [8]. The LIST module will be used to suppress surface-ionised beam contaminants that are typical to the lanthanide region, and would usually prohibit laser spectroscopy measurements [48]. Promethium ions will be produced using the three-step resonance ionisation scheme shown in Fig. 3(A) of [8], with the hfs and IS measurements performed by scanning the 452 nm, $J = 5/2^+ \rightarrow 5/2^+$ transition. Ions will be mass separated using one of the ISOLDE separators and delivered to the measurement devices discussed below.

A combination of the PI-LIST mode and an injection seeded laser will be used, with which resolutions of $\sim 200-300$ MHz should be achievable. This will allow us to flexibly switch between the high-resolution perpendicular illumination mode, and the high-efficiency, low-resolution (~ 1.5 GHz) "normal" LIST mode. Whilst the former will afford the precision needed to determine Q values, the latter will allow us to extend our IS measurements to most weakly produced cases furthest from the line of stability. Based on simulations with a minimum of 20 counts in the maximum of the hfs spectra, a precision of better than 3% for extracted μ values can be achieved for odd-mass cases and 10% for the odd-odd isotopes. Similarly, at worst a $\sim$ 20% precision will be achievable for Q values in the high-resolution PI-LIST mode. For the IS, uncertainties of $\sim$ 20 MHz in high-resolution and 50-100 MHz in low-resolution mode will be reachable. These values are sufficient to explore the physics motivations outlined in the previous sections.

The hfs spectra will be recorded by ion counting using the ISOLDE Decay Station (IDS), ISOLTRAP's multi-reflection time-of-flight mass spectrometer (MR-ToF MS), a magneToF detector, or the ISOLDE Faraday cup (FC) where applicable. IDS will be equipped with 12 clover detectors ($>$ 6% detection efficiency at 1 MeV) and an array of plastic scintillators for beta tagging ($\gtrsim$ 60% efficiency). Whilst the FC and magneToF will be used for scanning high-purity cases, IDS and the MR-ToF-MS will provide additional selectivity and sensitivity in cases where prohibitive contamination is present in the beam. Additionally, IDS can be used for cases with known isomeric states with overlapping hfs spectra, which will be separable through characteristic decay tagging. Our combined approach to ion counting has the added benefit of providing additional spectroscopic data from mass and decay measurements that can be made in parallel to the primary laser study.

Firm I assignments will be possible based on the number of observed transitions for $I = 3/2$ and $5/2$ cases such as ^{135}Pm (10 and 15 possible, respectively). For cases with $I > 5/2$, the quality of the fit in high-resolution mode will also allow unambiguous determination of I. The $\delta\langle r^2\rangle$, μ and Q values across the long chain will be extracted and compared to predictions from theory. The data will help elucidate how the deformation of the ground states in promethium isotopes evolve above and below $N = 82$, and whether sharp or smooth shape transitions are present. Systematics in the extracted $\delta\langle r^2\rangle$ and moments will be used to identify potential evidence for ground state triaxiality in the neutron-deficient cases, and octupole collectivity in the neutron-rich isotopes.

3 Summary of requested shifts

The following shift request is based on the measured yields from the LOI246, an extrapolation of those yields for the cases furthest from the line of stability, and our previous experience performing hfs scans with IDS, ISOLTRAP, and the MagneToF.
We request **18 shifts for the IS and hfs measurements**, which are summarised in Table 2. This allows for at least a minimum of 20 counts to be collected in the peak of each hfs spectrum which, as discussed above, allows for extraction of all desired observables with high enough precision. At least two scans will be made for each isotope to remove any systematic uncertainties that could be introduced by the scanning direction. The requested time accounts for regular reference scans of ^{144}Pm, the time for setup between measurements, and the time to tune the separator magnets between different masses.
In addition to the aforementioned dedicated measurements, **3 shifts are requested for**: beam tuning from the ISOLDE separator to the experimental setups and optimisation of the laser systems. Furthermore, **we request an additional 2 shifts for** the optimization of the LIST conditions in normal and perpendicular illumination mode.
Therefore, **in total we request 23 shifts** to complete the proposed measurements.

Table 2: Summary of requested shifts for laser scanning.

A	$T_{1/2}$	Scanning mode	New measurements	Shifts
132	6.9 s	LIST	I, $\delta\langle r^2\rangle$, μ	2
133, 134	$\geq$ 5 s	LIST/PI-LIST	I, $\delta\langle r^2\rangle$, μ, Q	2
135	49 s	PI-LIST	I, $\delta\langle r^2\rangle$, μ, Q	1.5
136	107 s	PI-LIST	I, $\delta\langle r^2\rangle$, μ, Q	1
137	2.4 mins	PI-LIST	I, $\delta\langle r^2\rangle$, μ, Q	1
138	3.3 mins	PI-LIST	I, $\delta\langle r^2\rangle$, μ, Q	0.5
139	4.1 mins	PI-LIST	I, $\delta\langle r^2\rangle$, μ, Q	0.5
140	9.2 s	PI-LIST	$\delta\langle r^2\rangle$, μ, Q	0.5
141	20.9 mins	PI-LIST	$\delta\langle r^2\rangle$, μ, Q	0.5
142	40.5 s	PI-LIST	$\delta\langle r^2\rangle$, μ, Q	0.5
144	363 days	PI-LIST	reference isotope	1.5
148	5.4 days	PI-LIST	$\delta\langle r^2\rangle$	0.5
149	53 hrs	PI-LIST	$\delta\langle r^2\rangle$, Q	0.5
150	2.7 hrs	LIST/PI-LIST	I, $\delta\langle r^2\rangle$, μ, Q	1
151	28.4 hrs	LIST/PI-LIST	$\delta\langle r^2\rangle$	1
152	4.1 mins	LIST	$\delta\langle r^2\rangle$, μ	1.5
153	5.3 mins	LIST	$\delta\langle r^2\rangle$, μ	2
			Total:	**18**

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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: IDS, ISOLTRAP
  - ☒ 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 in LIST mode)	Minimum required rate at experiment (pps)	$t_{1/2}$
$^{135-149}$Pm	> 500	0.2	49 s - 18 y
^{132}Pm	~ 3	0.2	6.3 s
^{133}Pm	~ 40	0.2	13.5 s
^{134}Pm	~ 250	0.2	5.0 s
^{150,151}Pm	> 100	0.2	2.7 h - 28 h
^{152}Pm	~ 2	0.2	4.1 min
^{153}Pm	~ 0.8	0.2	5.2 min

- Full reference of yield information: LOI246 - yields of $^{132-134}$Pm and ^{152,153}Pm have been extrapolated from the measured yields of $^{135-151}$Pm through the total efficiencies dependency on the half-lives.
- Target - ion source combination: *Ta foil target with a LIST module*
- RILIS? (*YES*)
  - ☒ Special requirements: *isomer selectivity, LIST, PI-LIST, ultra-narrow bandwidth laser scanning using an injection-seeded laser, fibre transport of CW laser light from CRIS, laser shutter access, etc.*

- Additional features?
  - ☐ Neutron converter: No
  - ☐ Other: (*quartz transfer line, gas leak for molecular beams, prototype target, etc.*)
- Expected contaminants: Well-known surface ionised contamination in lanthanide region, should be suppressed (10^6) by LIST, selectivity of ISOLTRAP and IDS can filter out any remaining as demonstrated in LOI219.
- Acceptable level of contaminants: *The LIST module will suppress the vast majority of surface-ionised contamination (*$\sim 10^6$ *suppresion factor), whilst the selectivity of IDS and ISOLTRAP's MR-ToF MS will be able to filter out any remaining contamination.*
- 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? *No*