EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Letter of Intent to the ISOLDE and Neutron Time-of-Flight Committee

Yield measurements for lanthanide elements with Ta-foil target and a LIST ion source

September 27, 2022

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Abstract: We intend to measure production yields of different lanthanide isotope chains from a Ta-foil target coupled with the Laser Ion Source and Trap (LIST). Currently, data with 1.4-GeV protons is scarce in this region of the nuclear chart, hindering the possible proposals of physics motivated experiments.

Requested shifts: 70 shifts, split into multiple runs over 3 years (up to LS3)

1 Introduction

The Resonance Ionization Laser Ion Source (RILIS) [1] at ISOLDE is the most commonly used ion source, due to its chemical selectivity and good ionization efficiency. The step-wise excitation and subsequent ionization of the atoms of interest can either enhance the ionization efficiency of elements with low to medium high ionization potentials (IP) (5-6.5 eV) or even be the sole source of ionization for elements with higher IP, without causing significant isobaric contamination. This is a distinct advantage over e.g. a plasma ion source. In the standard configuration, ionization takes place inside a hot cavity, usually made from tantalum. Elements with low enough IP can still get surface ionized and therefore increase the isobaric background. In order to avoid this effect, the laser ionization region can be decoupled from the hot cavity in a geometry known as LIST (Laser ion Source and Trap) [2, 3]: Ions produced inside the hot cavity are deflected by repelling electrodes while the effusing atoms can be ionized outside the hot cavity. Ion confinement and extraction is guaranteed by guiding them through an RF quadrupole structure towards the field of the extraction electrode. Operating the repeller electrodes with negative voltages in the so-called ion guide (IG) mode, acting as pre-extraction from the hot cavity, enables an easy in-situ switch to the standard RILIS operation mode with higher efficiency, but without contaminant suppression.

An additional advantage of the LIST is the possibility to off-set the laser(s) from the central axis of the ion source and deflect them perpendicularly. This way, Doppler-broadening caused by the high temperatures required for volatilization and extraction of the atoms, can be reduced and high-resolution laser spectroscopy becomes possible [4, 5]. Coupled to the low background due to the suppression of the surface ions, this method allows measurements even with low yields. Recently, this has been successfully demonstrated with the measurement of Ac isotopes [6].

2 Motivation

Laser spectroscopy has been a key technique at ISOLDE since the very beginning and has been used to investigate the nuclear shape and its evolution across wide isotopic chains. The RILIS has been used in the past in extensive in-source laser spectroscopy campaigns, adding significantly to the publication success of ISOLDE (e.g., [7–9]). The main limitation, so far, was the Doppler-broadening, allowing only for heavy elements (>150 amu) to be investigated with sufficient resolution (down to 1 GHz) to reliably extract nuclear parameters as the magnetic moment μ_I and spectroscopic quadrupole moment Q_s and to pin down changes in mean square charge radii $\delta < r^2 >$ from dense hyperfine structure spectra. The Perpendicularly Illuminated (PI)-LIST has been shown to allow for resolution as low as 100 MHz [4]. The applicability of the PI-LIST for high-resolution laser spectroscopy has also been demonstrated at the RISIKO mass-separator at Mainz University for nuclear structure investigations on technetium (Tc) [10], holmium (Ho) [5] and promethium (Pm) [11]. Very recently this year, its first-time application at ISOLDE-GPS on Ac was very successful.

The PI-LIST collaboration does not aim to propose laser spectroscopy experiments in areas where other setups at ISOLDE are better suited to perform measurements. Instead, we propose to investigate areas in the nuclear chart where a clear advantage of using the PI-LIST exists. One such area of interest is given in the region of the elements known as lanthanides. High-resolution fast beam laser spectroscopy experiments at ISOLDE (and elsewhere) commonly require neutralization of the delivered radioactive ion beam. Due to their rich atomic structure, caused by the open d- and f-shells, neutralization of lanthanides distributes electrons among a multitude of excited states, thereby drastically reducing the availability of atoms in a single experimentally desired electronic configuration. Even though multiple states can also be thermally populated in a hot cavity ion source, the distribution concentrates on only a few low-lying states, reducing losses into non-accessible configurations. Clearly, for these cases the PI-LIST can offer higher efficiency and is therefore better suited.

Additionally, standard RILIS ion beams in the lanthanide region usually suffer from non-negligible surface-ionized contamination of these medium-IP elements, which are intrinsically suppressed by using LIST. A $\mathrm{UC_X}$ target-LIST campaign in April 2022 showed promising results for the lanthanide ytterbium, proving loss factors and contamination suppression capabilities within expectations. Production yields around mass 162 even exceeded those measured with a Ta target-RILIS ion source unit, where production is predicted to be higher. This already points towards a good suitability of the LIST for this part of the nuclear chart. Following laser scheme development work over the last years, high achievable ionization efficiencies under ideal conditions in the range of multiple 10 % for Tb [12], Dy [13], Ho [14], Tm [15], Yb [16], and Lu [17] were reported, as well as the first-time experimental determination of the ionization potential of Pm with minuscule artificial samples [18].

The physics motivation for this region of the nuclear chart can be divided, roughly, into the ones outlined below. Fig. 1 illustrates the situation by showing currently available experimental data on the changes in mean square charge radii $\delta < r^2 >$. Once respective parts of the LOI have been completed, full proposals with detailed physics motivations will be submitted.

Nuclear shape evolution at the N = 82 shell and Z = 64 subshell closure

One of the main interests in the lanthanides is the effect of a "sub-magic" proton subshell closure at Z = 64 (Gd) on the nuclear structure systematics either side of the N = 82 shell closure. All of the lanthanide isotope chains cross this region and therefore provide great evidence of the nuclear evolution depending on the shell structures. In the region of N > 82 for elements close to Z = 64, a distinct jump-like change of deformation happens where the influence of the subshell closure diminishes for N > 88 [20]. This is especially evident in the two stable isotopes of Eu (Z = 63, stable isotopes at N = 88 and N = 90), which show a strong difference in trend of deformation. For Tm with Z = 69 this kink vanishes and a smooth trend is observed for increasing neutron numbers [21].The lower-Z boundary of this area is yet to be established, possibly still being present at Z = 61 (Pm) where no experimental data is available. Also for the Z = 88 chain (Nd), available data shows a sharp increase of the radii, but the presumed kink position is not mapped out yet.

Isotopes with N < 82 exhibit odd-even staggering (OES). Even though it was predicted

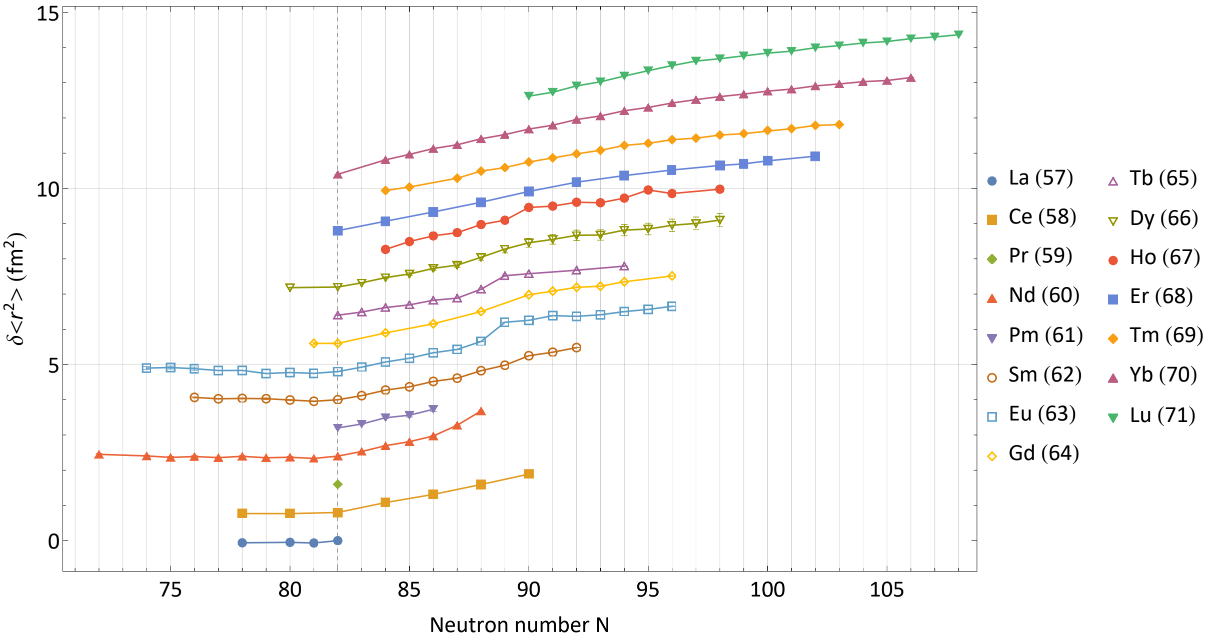

Figure 1: Experimentally determined changes in ground state mean square charge radii in the lanthanide region (proton number Z in brackets) taken from [19], with Pm data added from [11]. Constant offsets between datasets at N = 82 neutron shell closure and connecting lines added to guide the eye. Areas of interest are outlined in the text.

that reaching the subshell closure would stabilize the nucleus and therefore smooth out deformation, no such behavior has been observed [22]. A remarkable effect is evident in a high-spin ($I = 11/2$) ^{141}Sm isomer at N = 79, where the strongly pronounced OES of the ground state almost completely vanishes [23]. Similar isomers exist in the Dy isotopic chain [24] and potentially in further even-Z nuclei. Predicting these strong effects has been established as a good benchmark for validity of different nuclear theory models, namely the choice of suitable effective nuclear forces [25]. Investigating these isomers will become possible with the beam purification provided by LIST.

Below the N = 82 shell closure, there is strong evidence on the transition from spherical to strongly deformed nuclei at N < 75. However, radii and deformation (accessible via quadrupole moments) measurements in this region are restricted to moderately deformed nuclei (Eu: N > 74; Sm: N > 76). In each case only gradual development of deformation was observed. Just below, the jump-like change of deformation was predicted for Eu, Sm, and Pm [26], e.g., $\beta(^{135}\mathrm{Pm}) = 0.198$, $\beta(^{136}\mathrm{Pm}) = 0.292$. In contrast, for the adjacent Nd chain a gradual change of deformation was observed down to N = 72. It is of high interest to check this prediction as well as to receive valuable information on spins, magnetic and quadrupole moments in this ”new” region of deformation.

Possible octupole deformation in the region of Z = 56, N = 88

Small stable octupole deformation ($\beta_3 \neq 0$) was predicted for $^{146\text{-}149}$Pm [27]. There are nuclear spectroscopic evidences of possible octupole deformation also for ^{151,150,153}Pm [28–31]. However, in [32] nearly all properties of the corresponding nuclei, which have been taken as evidence for octupole deformation, were understood assuming $\beta_3 = 0$, namely, decoupling parameters, magnetic moments, and splittings between "parity doublets". To elucidate the situation it is of importance to measure μ, Q and $\delta < r^2 >$ for $^{146\text{-}153}$Pm, noting that magnetic moments have been shown to be a sensitive probe for mixing of nearly-degenerate opposite-parity states occurring with octupole deformation [33].

It is well known that in the Ra/Th region (Z $\sim$ 88, N $\sim$ 132) a strong correlation exists between the existence of presumed octupole deformation and inverse odd-even staggering in the charge radii [34–36]. There is some experimental evidence that such inversion takes place exactly in those nuclei for which reflection asymmetric shapes were theoretically predicted and supported also by spectroscopy results in Ra/Th [37]. This inversion was qualitatively explained by introducing $\beta_3 \neq 0$ and assuming that for the odd-N nuclei β_3 is larger than for the even-N neighbors. The same effect was found for $^{153\text{-}155}$Eu [38], i. e., in the region under consideration. Remarkably, for these Eu nuclei some evidence on octupole deformation also exists [39]. Studies of the radii for lighter-Z elements such as the Pm isotopic chain are needed to confirm whether the odd-even reversal noted for the Eu isotopic chain persists in these nuclei with possible octupole deformation.

Proton-emitting nuclei

In 1982, the neutron-deficient Tm and Lu isotopes, ^{147}Tm (Z = 69) and ^{151}Lu (Z = 71), were the first two ground-state proton emitters to be discovered [40, 41]. With half-lives of 580 and 80 ms, respectively, we hope to be able to demonstrate sufficient yields for their investigation at ISOLDE.

Measurement of the spin, electromagnetic moments and charge radii of the neutron-deficient Tm and Lu isotopes will allow the role of deformation in the proton decay process to be thoroughly investigated. By charting the trend of the charge radii towards and across the proton drip line (at ^{149}Tm and ^{155}Lu, respectively), the effect of the unbound proton on the charge radii can be studied. As the charge radii are incredibly sensitive to the change in proton distribution as the drip line is approached, this will allow new phenomena past the proton drip line to be uncovered.

For many proton emitters, the rate of proton decay can only be explained by the inclusion of nuclear deformation. For ^{150}Lu and ^{151}Lu, only with an assignment of oblate deformations could the the proton decay rates be described [42–44]. Similarly, a deformed framework was also used to interpret the nuclear structure of the transitional nucleus ^{147}Tm [45], where in this region of the proton drip line (between Tm and Ho) a rapid transition between spherical and highly deformed shapes is predicted [46]. In such cases, it was only by comparison between experimental values (e.g. proton decay rates) and theoretical calculations that assignments of nuclear spin and deformation could be deduced. In order to provide definitive answers, unambiguous nuclear-model-independent measurement of the nuclear spin and deformation (by means of quadrupole moment and charge radii) is required.

We would like to thank Anatoly Barzakh and Kara M. Lynch for their help in summarizing the possible physics cases.

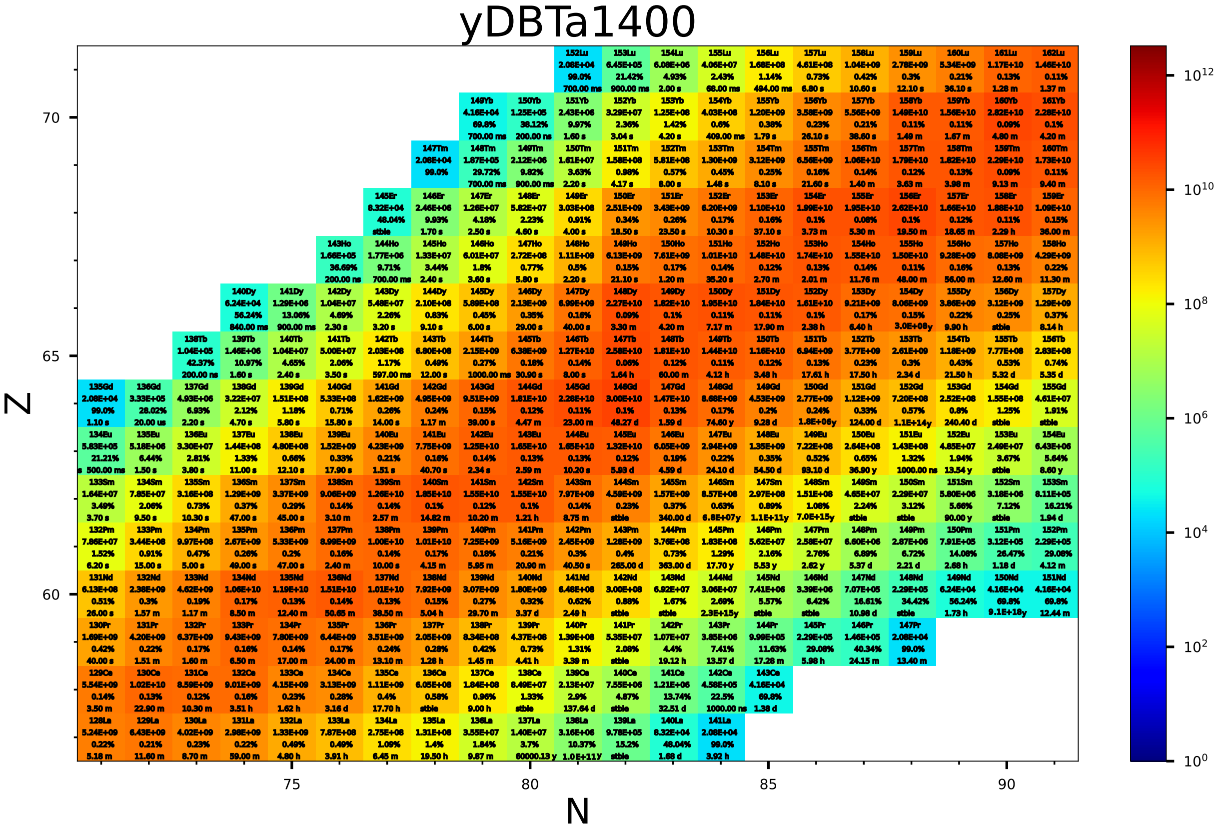

Figure 2: In-target production (particles per μC) using 1.4 GeV protons on a standard ISOLDE Ta foil target as predicted by FLUKA for the area of interest.

3 Method

Ionization will be provided through resonance laser ionization with the LIST. To measure the respective RIB production yields, we will use primarily the ISOLDE Fast Tape Station. In addition, other tools, i.e. Faraday cups, (time-resolved) single ion counting with a MagneTOF detector and collections with off-line γ-analysis, may be employed where applicable. For each element, production yields of several isotopes will be measured, ideally by recording release curves. For elements where the release parameters can be determined via release curves, wider production-yield predictions are directly possible by calculating the half-life dependent release efficiency and by utilizing in-target production simulation data (ABRABLA, FLUKA - see fig. 2). Since yield measurements of several isotopes per chain are foreseen, the quality of the predictions can be evaluated by comparison. In cases where release parameters cannot be deduced, the release efficiency vs. half-life dependency can be extracted by measuring production yields from isotopes across different half-life regimes. This method eliminates the necessity to measure full isotopic chains to provide yield data for dedicated physics proposals.

We propose to start with the yield measurements of elements which have been studied already at ISOLDE and expand the available data. One of these elements is Dy, which is regularly extracted from Ta-foil targets for medical isotope production and in-source laser spectroscopy has already been carried out in parallel [24]. At the MEDICIS facility,

the efficient extraction of Tm has been demonstrated [15], making it another ideal candidate for a first measurement campaign with the scope to extend knowledge of nuclear parameters towards the neutron shell closure.
Additionally, following the recent groundwork on Pm [11], yield information would enable to possibly extend the isotope chain towards the area of interest outlined above.
Cases known to be more difficult due to their chemical behavior such as refractivity or strong absorption into molecular sidebands as expected for e.g. Gd and Tb are proposed to be addressed at a later stage. Significant increase of target operation temperature can be a path to be investigated.

4 Summary of requested shifts

We request a total of 70 shifts, spread out over multiple runs over the next 3 years. We would like to limit the yield measurement campaigns to about 3 elements per run but distribute the runs across the next yars of proton availability at ISOLDE. For our first run, we will set out to determine the yields of Dy, Tm and Pm. With these elements we will attempt to verify our proposed method for reducing the required number of isotopes measured per element. We will use the LIST in order to determine the loss-factors and contaminant suppression capabilities and provide the most complete results possible for the proposals to come.

Summary of requested shifts:

- 0.5 shifts for the yield measurement/release curve of one single isotope for at least 8 isotopes per element: 4 shifts per element
- restricting the isotopes to ±5 amu around the Z=64 proton subshell closure: 11 elements
- 1 shift for accelerator setup and beam tuning for each run: 4 shifts (if we assume 3 elements per run)
- 2 shifts per element for laser frequency tuning and LIST optimization

gives $4 \times 11 + 4 + 22 = 70$ shifts, spread out over 3 years (up to LS3) and several runs.

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Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

The experimental setup comprises: (*name the fixed-ISOLDE installations, as well as flexible elements of the experiment*)

Part of the	Availability	Design and manufacturing
ISOLDE Fast tape station	☒ Existing	☒ To be used without any modification
GLM chamber for collections	☒ Existing	☒ To be used without any modification

HAZARDS GENERATED BY THE EXPERIMENT (if using fixed installation:) No additional hazards.

Average electrical power requirements (excluding fixed ISOLDE-installation mentioned above): No additional requirements.