EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

In-source laser resonance ionization spectroscopy of neptunium and plutonium

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Mia Au[1,2], Anastasia Borschevsky[3], Katerina Chrysalidis[1], Raphaël Crosa-Rossa[3], Christoph Düllmann[2,4,5], Reinhard Heinke[1], Asar Jaradat[1], Magdalena Kaja[2], Bruce Marsh[1], Iain Moore[6], Andrea Raggio[6], Sebastian Rothe[1], Simon Stegemann[1], Darcy van Eerten[7], Clemens Walther[7]

[1]*SY-STI, CERN, Switzerland*
[2]*Johannes Gutenberg-Universität Mainz, Germany*
[3]*Rijksuniversiteit Groningen, Groningen, Netherlands*
[4]*GSI Helmholtzzentrum für Schwerionenforschung, Germany*
[5]*Helmholtz Institute Mainz, Germany*
[6]*University of Jyväskylä, Finland*
[7]*IRS, Leibniz Universität Hannover, Germany*

Spokesperson: Mia Au mia.au@cern.ch, Magdalena Kaja mkaja@uni-mainz.de
Contact person: Mia Au mia.au@cern.ch

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Abstract: Studying the actinide region of the nuclear chart is motivated by interest in nuclear properties and fundamental physics research; however this has so far been limited by the production of the mostly radioactive species. We propose to use in-source laser resonance ionization spectroscopy (RIS) to evaluate laser ionization schemes and to measure isotope shifts and hyperfine structures for isotopes of neptunium and plutonium. Coupled with simulations of in-target production mechanisms, measurements of release and yields will provide the community with information on the production and extraction of neptunium and plutonium beams and availability of the Np and Pu isotopic chains at ISOLDE. From measurements of isotope shifts, changes in mean-squared charge radii can be extracted for the neptunium and plutonium isotopic chains up to mass 241. State-of-the-art theoretical calculations of the atomic factors will be used to extract nuclear properties from measurements.

Requested shifts: 16 shifts over 1 run

1 Introduction

Preliminary target and ion source studies [1] have identified the possibility of laser-ionizing neptunium and plutonium beams produced from uranium carbide targets. For several of these above-target-mass isotopes, in-target production simulations predict reaction pathways that favor pre-irradiation and subsequent decay before delivery of these actinide beams. To enable proper planning of experiments with isotopes where the proton number is greater than that of the target nucleus, the production mechanisms must be identified and understood.

High-resolution laser spectroscopy of plutonium at the University of Jyväskylä and the Johannes Gutenberg-Universität Mainz compared the two techniques of in-source resonance ionization spectroscopy (RIS) and collinear resonance ionization spectroscopy (CRIS), demonstrating the capabilities of laser spectroscopy to access fundamental nuclear structure properties of the actinides through the measurement of the isotope shifts [2]. The isotopes included in this study were $^{238-244}$Pu, obtained from a sample produced at Mainz. The hyperfine structure was additionally studied for isotopes $^{238-242}$Pu using a MOX-fuel sample [3].

At ISOLDE, isotopes can be produced from direct reactions with the driver beam and reactions of secondary particles but also from decay, potentially giving access to further neutron-deficient species and overlapping the region of plutonium isotopes previously studied up to mass 241 [4]. Measurements of the hyperfine structure of ^{237}Np have been performed recently at Mainz and are pending publication. To our knowledge, high resolution in-source laser spectroscopy of neptunium has not yet been completed along an isotopic chain.

2 Motivation

Actinide elements offer opportunities for advancement in disciplines including fundamental research of nuclear properties, experimental and theoretical atomic physics, nuclear medicine and chemistry. The LISA (Laser Ionization and Spectroscopy of Actinides) Marie Sklodowska-Curie Innovative Training Network (ITN) aims to develop knowledge of the actinide region of the nuclear chart through investigations on properties ranging from ionization potentials, electron affinities and atomic levels to nuclear structure, with industry, experiment, and theory [5].

Extracting nuclear charge radii through laser spectroscopy can distinguish regions of the nuclear chart where changes in nuclear deformation occur by measuring the evolution of the nuclear size and shape along an isotopic chain. For the actinides, nuclei at the shell closure N = 126 are spherical. Above the shell closure, regions of deformed nuclei show trends in nuclear structure that offer the possibility for comparison against theoretical nuclear models, for example Figure 1. Extending nuclear models to better descriptions of properties such as the nuclear charge density [6] requires better experimental data in the actinide region, where isotope shift data is missing for more exotic isotopes and some elements, notably neptunium.

The use of high resolution laser spectroscopy techniques would additionally allow the hy-

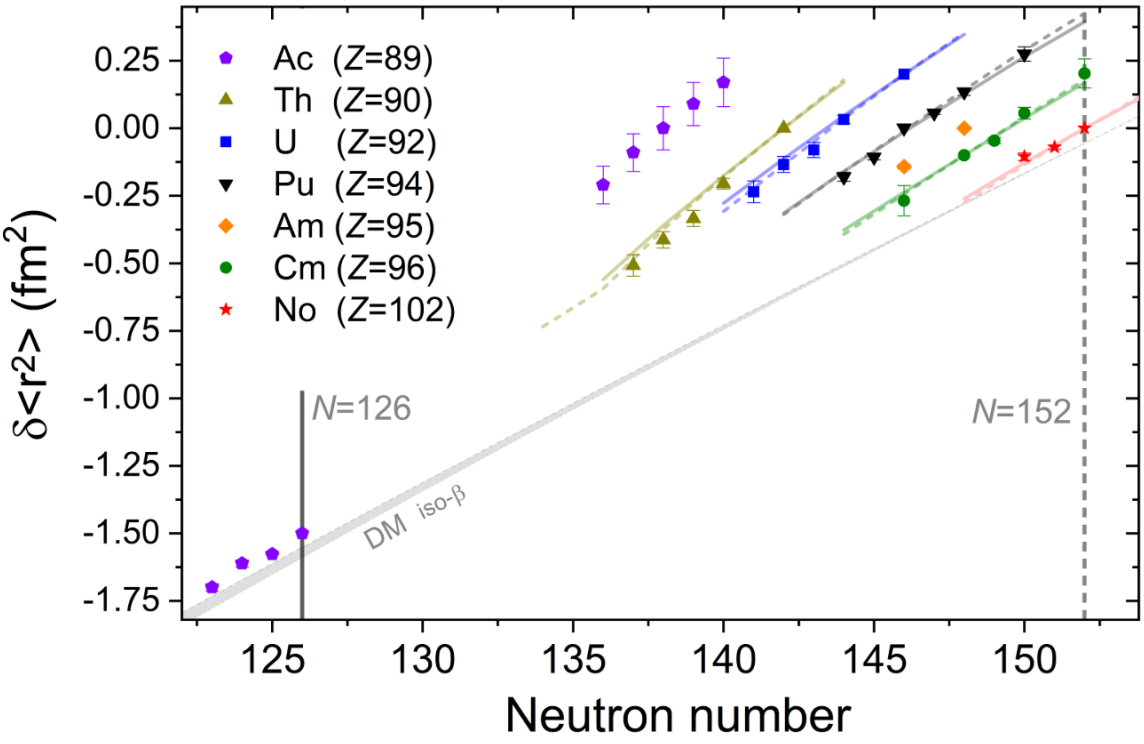

Figure 1: Figure taken from [7, 8] showing changes in the mean square charge radii based on laser spectroscopy data for the Ac to No, data taken from [9–11]. Theoretical predictions by an energy density functional approach are given by the lines. For orientation the iso-beta line for a droplet model (DM) is also shown. Vertical lines represent nuclear-shell or sub-shell closures.

perfine structure of the neptunium and plutonium isotopes to be measured, providing a comprehensive probe into underlying nuclear properties including nuclear spins, electromagnetic moments, sizes and shapes.
Furthermore, neutron-deficient actinides have been highlighted for static octupole deformation. Direct experimental evidence of this behaviour has been reported only for a few Ra isotopes [12–14] via Coulomb excitation studies. Atomic spectroscopy is not able to provide a direct measurement in this context, but the comparison with nuclear energy density functional (EDF) models where the octupole degree of freedom is taken into account can produce valuable insights [11]. In order to validate these models a comparison with experimental data from heavier actinides is needed.
Radioactive molecules present another opportunity for a rich experimental program, as shown by recent studies on radium monofluoride [15]. Several actinide molecules have been identified as candidates to investigate important questions in fundamental physics, including studies of electric dipole moments (EDMs) [16, 17] and nuclear magnetic quadrupole moments (MQMs) to probe time-reversal and parity symmetry violation in molecules [18, 19]. Due to the experimental difficulty in production, preparation and handling encountered with the actinides, our knowledge of the electronic and chemical properties of molecular actinide compounds is mostly based on theoretical investigations. Some theoretical predictions are already available for certain actinide species like the oxides [20, 21]. Such theoretical studies encounter difficulties in accurately describing the relativistic effects and the high density of electronic states, which are present in many of these species. There is thus significant interest in obtaining experimental data on actinide energy levels for the validation of theoretical methods.
Considering the extensive interest in actinide species at ISOLDE, information on production pathways, ion source operation and laser ionization schemes is critical for the success of future experiments.

3 Method

In a recent study, we used surface ion sources and pre-irradiated uranium carbide targets at ISOLDE to observe laser-ionized beams of neptunium and plutonium using a developmental neptunium laser scheme and a plutonium laser scheme published by Kneip et al. [22]. With lower formation enthalpy of the mono-atomic gaseous state and lower negative adsorption enthalpy on the tantalum surfaces of the target container material, we predict plutonium and neptunium to be the least refractory of the light actinides [23] and therefore the easiest to release in atomic form for laser spectroscopy.
To see Np from the pre-irradiated target commissioning study, a first excitation step of 25277.663 cm^{-1} (413.4 nm) from the $5f^46d7s^2$, J=11/2 ground-state to an odd-parity, $5f^46d7sn$p J=9/2 state was used. In broadband mode, this first excitation step was proven to be resonant for a number of attempted second excitation steps. In Figure 2 three different ionization schemes are compared that were later investigated off-line at Mainz University. The scheme used in the previous study is the least intense, suggesting that we could have an improvement of signal-to-background using a different scheme.
Plutonium was studied with a first step of 24188.4 cm^{-1} from the $5f^67s^2$, J=0 ground

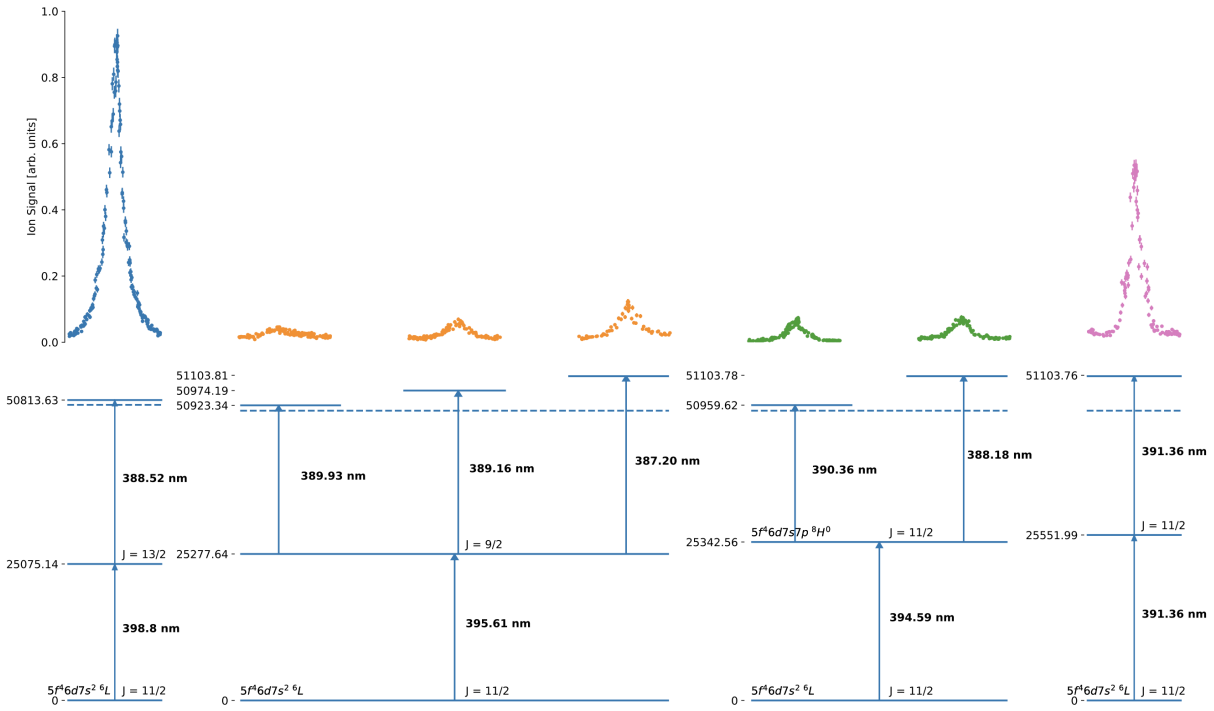

Figure 2: Intensities of different two-step ionization schemes for Np showing ion count rate against the total energy of the auto-ionizing state in cm^{-1}. [24]

state to the $5f^5 6d^2 7s$ J=1 state followed by a second step of 24690.3 cm^{-1} above the ionization potential to an auto-ionizing state. Another 3-step laser ionization scheme has been recently published for plutonium, indicating impressive overall ionization efficiencies of close to 50% [25]. This scheme has not yet been attempted at ISOLDE.
The 413.4 nm transition was scanned in plutonium isotopes 236, 239, 240 and on A=241 (Figure 3) using the grating Ti:Sa. Peaks are broadened beyond the expected Doppler width of around 2 GHz by the inherent spectral width of the utilized lasers and potential power broadening, resulting in Gaussian distributions with peak widths (FWHM) of approximately 5 GHz. The clear shift in peak centroid between isotopes already shows the possibility for determination of isotope shifts in this low-resolution configuration. Mass separation and contamination from the uranium target material have been identified as the main limiting factors on these masses.
Recent studies have demonstrated the capability of the Laser Ion Source and Trap (LIST) ion source [26] online at ISOLDE to suppress surface-ionized uranium by at least a factor of 10,000 (down to the detector noise limit) while decreasing the intensity of laser-ionized species by a factor 15 (Tl) to 60 (Yb, Sn). Furthermore, the ability to achieve resolution down to 200 MHz in a specialized operation mode, at cost of significant efficiency loss factor in the order of 1,000, has been demonstrated. The LIST nevertheless also offers the operation mode resembling standard RILIS operation with highest possible efficiency, but no contamination suppression or resolution enhancement. We propose to use the LIST ion source to validate suppression factors of uranium and possible molecular contaminants and evaluate the efficiency loss for neptunium and plutonium.

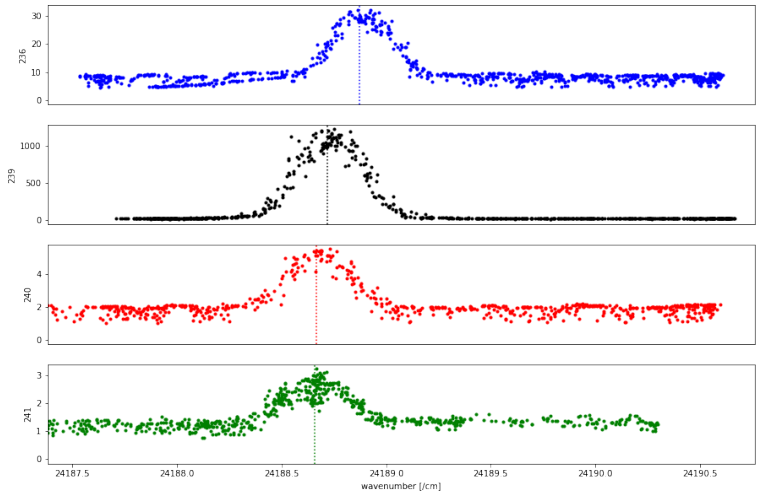

Figure 3: Combined resonance scans of the 413.4 nm transition of ^{236,239,240,241}Pu at ISOLDE with indication of the resonance's centroid (dotted lines). Intensity on GLM MagneToF detector in fA vs. wavenumber of frequency doubled Ti:Sa light. The shift in the centroid frequency (isotope shift) is clearly visible.

Alongside the proposed measurements, high accuracy calculations of the field and mass shifts and the hyperfine structure parameters of the transitions of interest in Np and Pu will be carried out. These calculations will be based on the state-of-the-art relativistic coupled cluster (RCC) [27, 28] and configuration interaction (CI) [29] approaches.

4 Summary of requested shifts:

We request a uranium carbide (UCx) LIST target and ion source unit to reach neptunium and plutonium isotopes while suppressing uranium. 1 shift is requested for taking the beam to the GLM beamline with a single-ion counter and optimization of the laser and ion source parameters and uranium suppression. Four shifts are then requested to evaluate two different plutonium laser ionization schemes and five shifts are requested for plutonium isotope shift and yield measurements. One shift is requested for evaluation of the neptunium scheme before 5 shifts are requested for measuring the isotope shifts and yields on other Np isotopes.
For a total of **16** requested shifts.
This project has received funding from the European's Union Horizon 2020 Research and Innovation Programme under grant agreement number 861198 project 'LISA' (Laser Ionization and Spectroscopy of Actinides) Marie Sklodowska-Curie Innovative Training Network (ITN).

Table 1: Breakdown of shift request

Masses	Measurement	Laser setup [nm (cm^{-1})]	Shifts
Setup			1
^{236,239,240}Pu	Scheme evaluation	420.77 (23765.98) [25] 818.91 (12211.3) 792.79 (12613.62)	2
		413.4 (24188.4) [22] 405.02 (24690.3)	2
$^{228-241}$Pu	isotope shifts, yields	""	5
^{237}Np	Scheme evaluation	398.801 (25075.15) [30] 388.523 (25738.52)	1
$^{228-241}$Np	isotope shifts, yields	""	5
Total			16

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Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

The experimental setup comprises: GLM

Part of the	Availability	Design and manufacturing
GLM beamline	☒ Existing	☒ To be used without any modification ☐ To be modified
	☐ New	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing
Single ion counter	☒ Existing	☐ To be used without any modification ☒ To be modified
	☐ New	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing

HAZARDS GENERATED BY THE EXPERIMENT (if using fixed installation:) Hazards named in the document relevant for the fixed [COLLAPS, CRIS, ISOLTRAP, MINIBALL + only CD, MINIBALL + T-REX, NICOLE, SSP-GLM chamber, SSP-GHM chamber, or WITCH] installation.

Additional hazards:

Hazards		RILIS	GLM
Thermodynamic and fluidic			
Pressure	[pressure][Bar], [volume][l]		
Vacuum			
Temperature	[temperature] [K]		
Heat transfer			
Thermal properties of materials			
Cryogenic fluid	[fluid], [pressure][Bar], [volume][l]		
Electrical and electromagnetic			
Electricity	3000 [V], $10^{-5}\mu$[A]		
Ionizing radiation			
Target material [material]		UC_x	
Beam particle type (e, p, ions, etc)		p^+	
Beam intensity		2 μA	

Beam energy		1.4 GeV	
Cooling liquids	[liquid]	water	
Gases	[gas]		
Calibration sources:	☐		
• Open source	☐		
• Sealed source	☐ [ISO standard]		
• Isotope			
• Activity			
Use of activated material:			
• Description	☐		
• Dose rate on contact and in 10 cm distance	[dose][mSV]		
• Isotope			
• Activity			
Non-ionizing radiation			
Laser		☒	
UV light			
Microwaves (300MHz-30 GHz)			
Radiofrequency (1-300 MHz)			

Hazard identification:

Average electrical power requirements (excluding fixed ISOLDE-installation mentioned above): [make a rough estimate of the total power consumption of the additional equipment used in the experiment]