EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Addendum to the Letter of Intent INTC-I-243

Addendum - In-source laser resonance ionization spectroscopy of neptunium and plutonium

September 26, 2023

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Abstract: The study of the actinide elements is motivated by interest in nuclear properties and fundamental physics research, but is limited by the production of the mostly radioactive species. In this addendum to LOI243 (INTC-I-243), we propose to

complete the in-source laser resonance ionization spectroscopy (RIS) using the Perpendicular Illumination (PI) mode of the LIST ion source, using the laser schemes evaluated in LOI243, 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 have identified the possibility of laser-ionizing neptunium and plutonium beams produced from uranium carbide targets [1]. 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.

In LOI243, we attempted to study neptunium and plutonium isotopes using in-source spectroscopy with the LIST. We were successful in demonstrating the suppression of uranium surface ionization and extracting yields in LIST mode for plutonium and neptunium, all calculated yields are shown in tables 1 and 2, respectively.

Isotope	$FES [cm^{-1}]$	$SES [cm^{-1}]$	$AI \ [cm^{-1}]$	Yield [ions/s]
²³⁶ Pu				29700
237 Pu				50
238 Pu	23765.98	35977.28	48590.9	990
239 Pu				118800
240 Pu				4000

Table 1: Extracted yields for each plutonium isotope.

Isotope	$FES \ [cm^{-1}]$	AI $[\rm cm^{-1}]$	Yield [ions/s]
²³⁷ Np			496
$^{238}\mathrm{Np}$	25075.14	50812.46	157
$^{239}\mathrm{Np}$			55
²³⁵ Np			10
$^{237}\mathrm{Np}$	25277.64	50909.33	300
$^{239}\mathrm{Np}$			38

Table 2: Extracted yields for each neptunium isotope for each excitation scheme.

For the first neptunium scheme the second laser was on the shoulder of the auto-ionizing (AI) state, recent studies [2] showed that the maximum of this state should be neptunium selective, thus we predict an increase in the presented yields by a factor of 2 by ionization via an AI of 50818.57 cm^{-1} .

2 Motivation

In LOI243, we had an old target unit (#752 UC_x LIST) and we were not able to go into PI-mode. We have broadband scans of $^{237-239}$ Np and $^{236-241}$ Pu, but there was leaking from neighboring masses on the GPS, occasionally giving extra peaks in the spectrum.

Our aim is to improve the old data with the HRS and using a new UCx target for highresolution ionization spectroscopy using the PI-mode of the LIST ion source, based on the known yields for these elements. This technique would allow the hyperfine 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.

3 Method

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 [3] and therefore the easiest to release in atomic form for laser spectroscopy. We consider the option to test new target and line heating configurations as well as new target materials to extend the range of reachable isotopes.

Due to leaking from neighboring masses on the GPS, we expect to have an improved mass resolving power using the HRS, as well as using a fast beam gating or laser-related time resolved data-taking.

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) [4, 5] and configuration interaction (CI) [6] approaches.

4 Summary of requested shifts:

We request a new uranium carbide (UCx) LIST target and ion source unit to reach neptunium and plutonium isotopes while suppressing uranium. 2 shifts are requested for taking the beam from HRS to a detector installed either in CB0 or after the ISCOOL with a single-ion counter and optimization of the laser and ion source parameters and uranium suppression. The remaining shifts are for improving the data acquired from the original LOI243 with the LIST-mode in the next way: 2 shifts are requested for plutonium isotope shift and yield measurements and 5 shifts are then requested for high-resolution studies in PI-mode; 2 shifts are requested for neptunium isotope shift and yield measurements and 5 shifts are then requested for high-resolution studies in PI-mode.

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 3: Breakdown of shift request			
Masses	Measurement	Laser setup $[nm (cm^{-1})]$	Shifts
Setup			2
		420.77 (23765.98) [7]	
$^{228-241}$ Pu	isotope shifts, yields	$818.91 \ (12211.3)$	2
		792.79(12613.62)	
Pu	PI-LIST mode	22.22	5
228-241Np	isotope shifts, yields	398.801 (25075.15) [8]	2
		388.523(25738.52)	
Np	PI-LIST mode	,, ,,	5
Total			16

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For a total of 16 requested shifts.

References

- M. Au, M. Athanasakis-Kaklamanakis, L. Nies, R. Heinke, K. Chrysalidis, U. Köster, et al. Production of neptunium and plutonium nuclides from uranium carbide using 1.4-GeV protons. Phys. Rev. C 107 (2023), 064604. DOI: 10.1103/PhysRevC.107. 064604.
- [2] In preparation. personal communication. 10.06.2023.
- [3] Jochen Ballof. Radioactive Molecular Beams at CERN-ISOLDE. Presented 09 Dec 2021. 2021.
- [4] Pi A. B. Haase, Ephraim Eliav, Miroslav Iliaš, and Anastasia Borschevsky. Hyperfine Structure Constants on the Relativistic Coupled Cluster Level with Associated Uncertainties. The Journal of Physical Chemistry A 124 (2020). PMID: 32202783, 3157–3169. DOI: 10.1021/acs.jpca.0c00877.
- [5] F. P. Gustafsson, C. M. Ricketts, M. L. Reitsma, R. F. Garcia Ruiz, S. W. Bai, J. C. Berengut, et al. *Tin resonance-ionization schemes for atomic- and nuclear-structure studies*. Phys. Rev. A **102** (5 Nov. 2020), 052812. DOI: 10.1103/PhysRevA.102.052812.
- [6] E.V. Kahl and J.C. Berengut. ambit: A programme for high-precision relativistic atomic structure calculations. Computer Physics Communications 238 (2019), 232– 243. DOI: https://doi.org/10.1016/j.cpc.2018.12.014.
- [7] Alfredo Galindo-Uribarri, Yuan Liu, Elisa Romero Romero, and Daniel W. Stracener. *High efficiency laser resonance ionization of plutonium*. Scientific Reports 11 (2021), 23432. DOI: 10.1038/s41598-021-01886-z.

[8] S. Raeder, N. Stöbener, T. Gottwald, G. Passler, T. Reich, N. Trautmann, et al. Determination of a three-step excitation and ionization scheme for resonance ionization and ultratrace analysis of Np-237. Spectrochimica Acta Part B: Atomic Spectroscopy 66 (2011), 242-247. DOI: https://doi.org/10.1016/j.sab.2011.02.002.

DESCRIPTION OF THE PROPOSED EXPERIMENT

Please describe here below the main parts of your experimental set-up:

Part of the experiment	Design and manufacturing				
If relevant, write here the	\boxtimes To be used without any modification				
name of the <u>fixed</u> installa-	\Box To be modified				
tion you will be using [Name					
fixed/present ISOLDE installation:					
e.g. COLLAPS, CRIS, ISS, Miniball					
etc]					
If relevant, describe here the name	□ Standard equipment supplied by a manufacturer				
of the flexible/transported equipment	\Box CERN/collaboration responsible for the design				
you will bring to CERN from your In-	and/or manufacturing				
stitute					
[Part 1 of experiment/ equipment]					
[Part 2 of experiment/ equipment]	□ Standard equipment supplied by a manufacturer				
	\Box CERN/collaboration responsible for the design				
	and/or manufacturing				
[insert lines if needed]					

HAZARDS GENERATED BY THE EXPERIMENT

Additional hazard from flexible or transported equipment to the CERN site:

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

	Magnetic field	[magnetic field] [T]
	Excessive noise	
Workplace	Working outside normal working hours	
workplace	Working at height (climbing platforms,	
	etc.)	
	Outdoor activities	
	Ignition sources	
Fire Safety	Combustible Materials	
	Hot Work (e.g. welding, grinding)	
Other hazards		
Other nazarus		