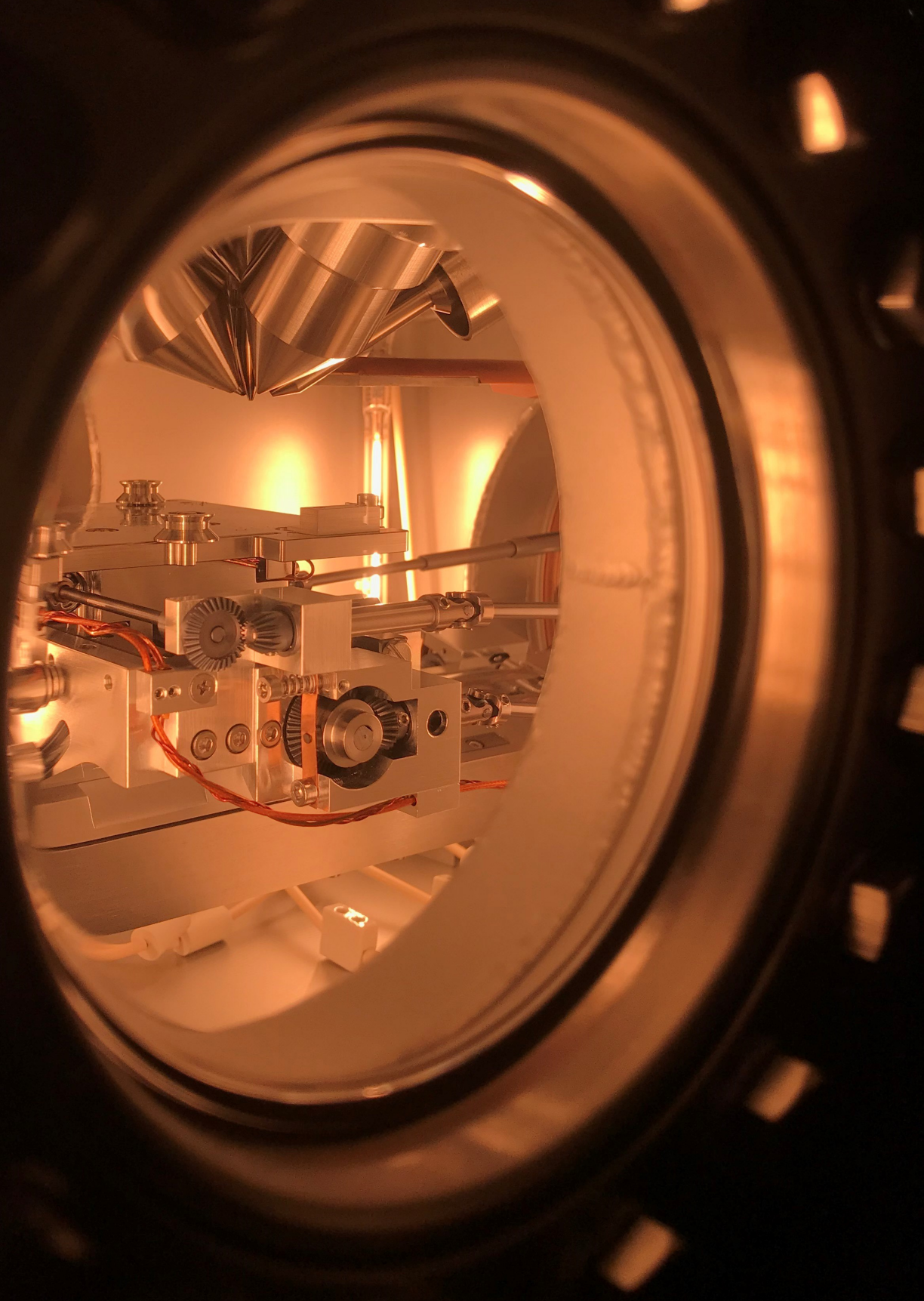




LASER IONISATION AND SPECTROSCOPY OF ACTINIDES

**ANNUAL NEWSLETTER**

**SEPTEMBER 2021**



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*This Marie Skłodowska-Curie Action (MSCA) Innovative Training Networks (ITN) receives funding from the European Union's H2020 Framework Programme under grant agreement no. 861198*



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## NOTE FROM THE EDITORS

Laser Ionization and Spectroscopy of Actinides, LISA, is the name of the Marie Skłodowska-Curie Innovative Training Network (ITN) formally established under grant agreement no. 861198. The LISA network was kicked off 1 November 2019 and an interdisciplinary team of researchers was assembled to tackle the projects encompassed in the research action. This first edition of the LISA annual newsletter offers a glimpse into the activities and accomplishments of the network this past year.

Taking the lead are the 15 early stage researchers (ESRs) who have begun their PhD programs within the LISA project and are based at different institutions across Europe at the beneficiaries listed below. The newsletter highlights work from this diverse group of young scientists with a wide variety of backgrounds and experiences.

In this, our first newsletter, we showcase the foundation of a new training network. It starts with a review of the past year, as our ESRs have adjusted to new lives in new countries in the strangest of times. The projects are all connected in work packages, which are each given an introduction. In our feature articles, we tackle important topics at the intersection between fundamental physics and impact to society. The accomplishments of the ESRs in their research are highlighted both on an individual level, as well as through collaborative efforts in the secondments already taking place.

We hope you enjoy this newsletter, and encourage you to follow our activities on social media, where you'll find updates on upcoming trainings, events, and research.

### Editors:

Mia Au  
Darcy van Eerten

### LISA Editorial Board:

Thomas Elias-Cocolios  
Isabelle Marie Fontaine  
Bruce Marsh

## NOTE FROM THE EDITORIAL BOARD

From the off-handed "What if?" to the realisation of this training network, much water has flowed through our pumps, many codes have been compiled, and time has been flying by. However, for the last year, the steam engine of our ESRs has kick-started this training network with an enthusiasm we could only have hoped for.

All of the ESRs have started their contract amidst confinement and other strange conditions, with researchers forced to telework instead of learning in the laboratory - if they could even reach their institution - or with secondments delayed or cancelled because of travel restrictions. Nonetheless, the information gathered by Darcy and Mia under the rainbow of this newsletter is testament to the resilience of our ESRs and their passion for the research and training offered by LISA.

This promises to deliver even more surprises in the years to come!

Bruce Marsh, Coordinator & Thomas Elias Cocolios, Training Officer.

### BENEFICIARIES



### PARTNERS

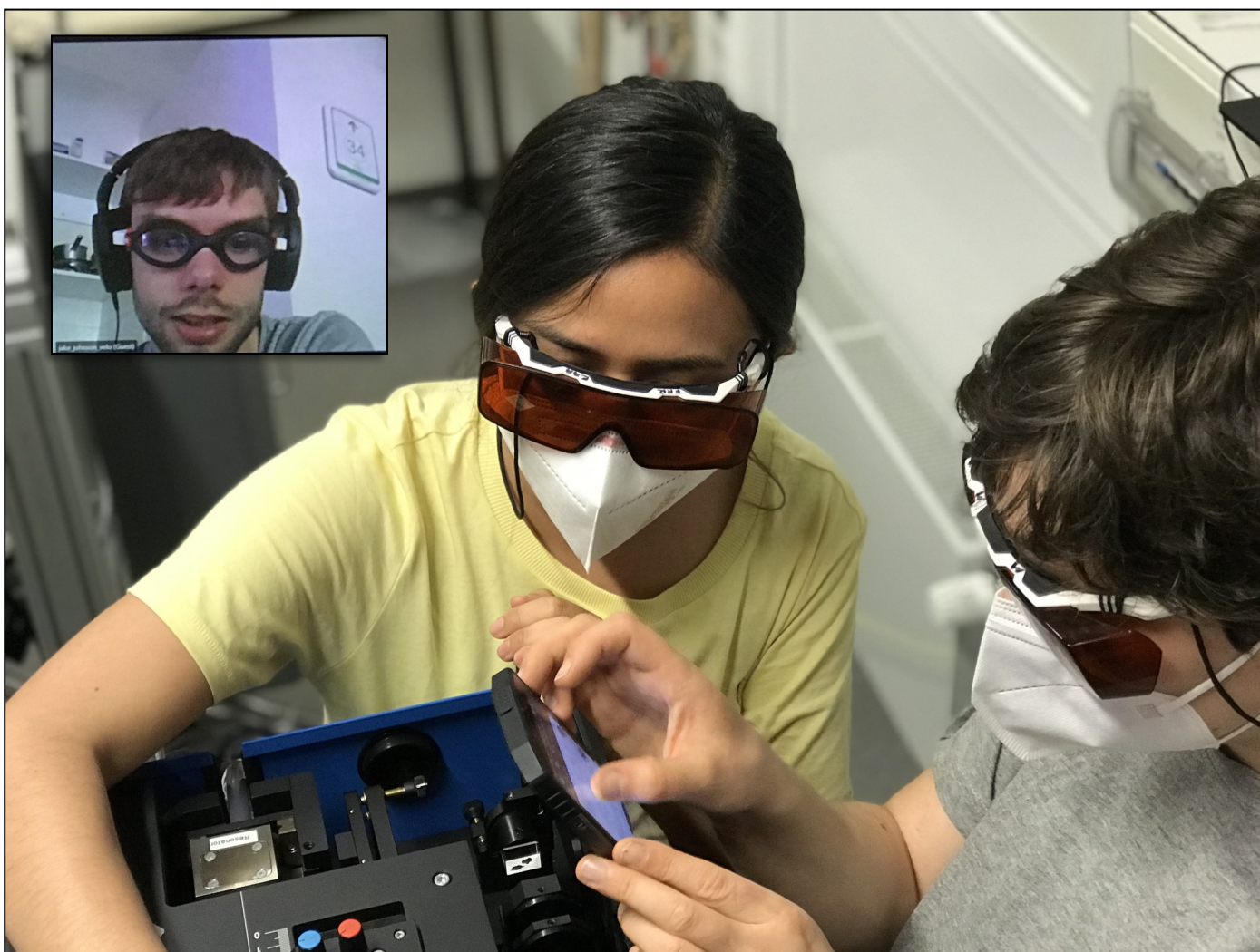


# YEAR IN REVIEW

**C**onnection is the all-important word in a year characterized by the pandemic, one that many feared would be lost in a budding new network. It has been incredibly exciting to see how the LISA network has dedicated itself to making those connections happen in spite of the challenges.

Though we have predominantly met in virtual spaces, members of the network have also been able to meet in person. Some ESRs have already started their various secondments, hosted at institutions where other ESRs are. Notably this has brought together ESRs at CERN in Switzerland, where many will be doing part of the work related to their PhD through secondments. Other hubs of LISA activity include Mainz in Germany and Leuven in Belgium, where multiple ESRs and affiliates are based.

The launch of the network began in earnest with the **Kick-Off Training** in November 2020. Almost all ESRs had started their PhD programmes at their respective institutions by then, though some were prevented from moving internationally due to the pandemic. The meeting included lightning speed introductions from the ESRs on their projects, our first opportunity to see how our diverse research topics interconnect. As part of the training, workshops had been organized on science communication and outreach, hosted by



*Dr. Elisa Romero Romero (HIM) and Elli Rickert (HIM) show Jake Johnson (ESR 1, KU Leuven) the cavity of a dye laser. Though participating online, Jake made sure to wear his goggles to acknowledge laser safety protection procedures.*



@claranellist and @Kurz\_Gesagt, about their experiences communicating scientific concepts to an international audience through YouTube and social media. Clearly conveying your ideas and adapting to your audience is also essential to success in a business context, as Fiona Reid demonstrated in her workshop on business, entrepreneurship and innovation.

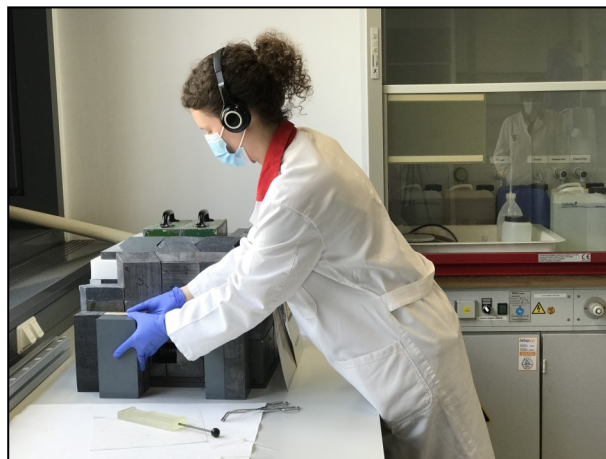
While for the ESRs this was the start of the LISA project, the Kick-Off also coincided with the **Mid-Term Review**, which saw the supervisors and beneficiaries come together to discuss the work that has gone into developing the programme, the recruitment process, and milestones ahead.

The first **LISA General Training** traded in the December Christmas Markets of Strasbourg for our desks across Europe (and Mexico!) as we once more met for a virtual training. The LISA affiliates gave their introductions, and the ESRs collaborated within their work packages to create posters, which can be seen on our website. The training featured lectures on the social impact of actinides (in medicine, energy production, and the development of a thorium nuclear clock), and the application of lasers (in trace analysis, additive manufacturing, micromachining and surface functionalisation), as well as laser safety training given by IREPA LASER. We kept the holiday spirit alive by sending out 'Secret Santa' presents, with cookies, chocolates and books arriving in the mail.

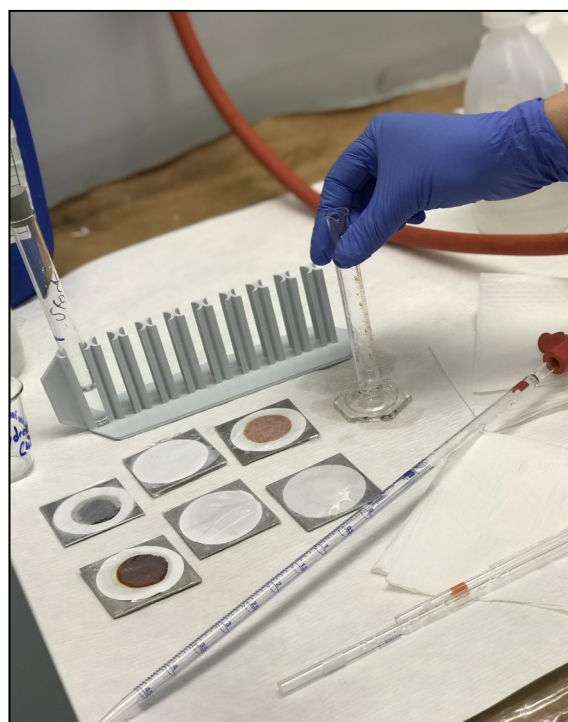
The winter months saw the work of the ESRs accelerating and starting on collaborations. Hosted virtually by ESR Julius Wessolek at M Squared Lasers in Glasgow, a workshop on **Injection-Seeded Ti:Sa Lasers** was held in March, bringing together LISA partners who are working on and developing Ti:Sa laser systems. By May, many ESRs and affiliates were presenting their work at a virtual workshop on the **Atomic Structure of the Actinides and Related Topics**, proceedings of which can be found in the MDPI open access journal 'Atoms' in December 2021.

Lockdown restrictions began to lift in many countries by the end of spring, renewing hope that we might be able to have the specialized laboratory training on **Nuclear Chemical Techniques and Laser Resonance Ionization** in person in Mainz at the start of June. With half the ESRs and affiliates already in Germany, this meant a hybrid training could be possible where the other half attended online. A collaboration between three institutes at Mainz, the Institute of Physics, Institute of Nuclear Chemistry, and the Helmholtz Institute Mainz, a total of six lab experiments were rotated through in groups of three or four. By pulling together all the available tools: cameras, microphones, wireless headsets, laptops, tablets, phones, masks, gloves, and the unabashed enthusiasm of participants and instructors, this hybrid workshop could be considered a rousing success.

In a year of change, it is the connections we make that ground us. Provided with a strong foundation through training and support, the coming years will see remarkable advances in the research done by our ESRs and affiliates. We look forward to what the next years will bring, and especially the opportunities to meet in person altogether.



Jessica Warbinek (ESR 10, GSI) performing neutron activation analysis on Ag at the Nuclear Chemistry Institute in Mainz.

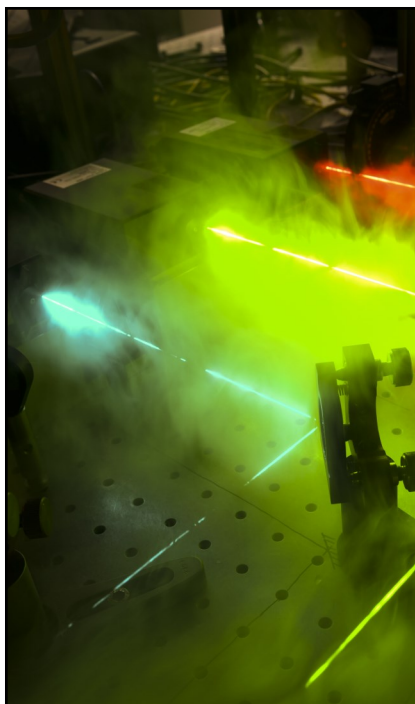


Views from inside the fume hood were shown on camera for the online participants.



## Novel Techniques and Technologies for Actinide Research

An introduction to Work Package 2  
by Mia Au



Hübner Photonics

**T**he actinide elements are all radioactive, making them notoriously difficult to produce and study. Because of this, they have evaded experiment despite an abundance of compelling motivation. The researchers in WP2 are tackling this problem through two approaches: developing new techniques for actinide production and developing laser technologies to enable further actinide research.

The ISOL (Isotope Separation On-Line) method of isotope production is already successfully used at several facilities within the LISA network to provide a sizable catalogue of isotopes available for research. At the ISOLDE facility at CERN, two LISA ESRs are working on developments to use the ISOL technique for the actinides. Bianca Reich, ESR 2, researches in-source resonance spectroscopy techniques such as the PI-LIST (Perpendicularly-illuminated laser ion source and trap) [1]. The PI-LIST is an ion source specifically for high resolution in-source spectroscopy, aiming to improve capabilities for in-source spectroscopy of actinide elements produced using the ISOL method.

In close collaboration, Mia Au, ESR 3, works on molecular beams with ISOLDE targets and ion sources, developing techniques to deliver the actinide elements to the ion source from the target. Formation of volatile molecules could be one extraction mechanism for refractory elements such as the actinides, in addition to being an interesting field of research in their own right. Target and ion source developments for the LISA project aim to extract and deliver actinides from ISOL targets to be studied with laser spectroscopy.

In-gas jet laser ionization spectroscopy is another avenue to producing and studying the actinides [2]. In this method, the main production mechanisms are heavy-ion fusion evaporation reactions from a beam impacting a thin target, creating heavy elements which are stopped in a buffer gas cell before mass separation. At the S3 LEB facility at GANIL, Anjali Ajayakumar, ESR 8, works with spectroscopy of the ions in a gas jet formed through a “de Laval” nozzle. To cope with ionization schemes for the actinides, Anjali is developing a narrow linewidth, high repetition rate, injection locked titanium sapphire (Ti:Sa) laser system for high resolution laser spectroscopy with improved sensitivity.

The first letter in LISA is a not-so-subtle clue regarding the critical importance of lasers in actinide research: it all depends on lasers. Two ESRs are working in collaboration with LISA industrial partners to advance laser technology for actinide research. Julius Wessolek, ESR 9, is working on Ti:Sa lasers for ion beam spectroscopy at M Squared Lasers. Development of a low-noise high-power tuneable pulsed narrow linewidth Ti:Sa amplifier has huge potential for facilitating ion beam spectroscopy. Julius’s developments on the fully automated SolTiS system aim to provide a market-grade laser solution to the challenge of actinide laser spectroscopy.

Dye lasers use a dye molecule as the lasing medium. These dyes can occasionally be difficult to operate and troubleshoot, but the lasers are indispensable for many laser schemes. Mitzi Urquiza, ESR 15, at Hübner Photonics is working on a solid-state tuneable CW laser, developing alternatives to CW dye lasers and building on the standard C-WAVE concept at Hübner Photonics.

The development of technologies to produce and study the actinides gives a foundation on which to build new experiments. The impact will be noticeable in the results of the coming years, as we search for conclusions about these fascinating but evasive elements.

### “it all depends on lasers”

#### Ti:Sa Laser:

- titanium-doped sapphire crystal
- tuneable 690-950 nm (red)
- double frequency for blue

#### Dye Laser:

- fluid chemical dyes
- tuneable depending on dye

#### CW vs pulsed:

- continuous wave, lower power
- pulsed light, higher power

[1] Heinke, R., *HI*, 238 (2017): 127.

[2] Yu. Kudryavtsev et al., *NIM B*, 297 (2013): 7-22



## Societal Applications

An introduction to Work Package 3  
by Darcy van Eerten

**F**antastic solutions can be imagined in the course of doing fundamental research. However, it is the physical work of experimentation that must be done to bridge the gap between what is possible, and what is practical. Within LISA, it is the limitations imposed by investigating and handling the actinides, along with operating complex laser systems, that pose significant practical challenges.

Radioisotopes have long been known to be remarkably effective tools in the fight against cancer.  $^{225}\text{Ac}$  has been proposed as a candidate isotope to be used in pre-clinical trials for targeted alpha therapy. Its decay properties, alpha decay with a 10-day half-life, make it ideal to provide impactful radiation of cancer cells at a short range, in a medically convenient timescale.

Jake Johnson, ESR 1, at KU Leuven in Belgium has been working on producing  $^{225}\text{Ac}$ . It is especially important in a pre-clinical trial that the source is an isotopically pure sample of  $^{225}\text{Ac}$ . Contamination with the more easily produced  $^{227}\text{Ac}$  can be dangerous for the patients and nullify the effects of the treatment. This can be achieved through the ISOL\* method at CERN MEDICIS, which will be able to isolate  $^{225}\text{Ac}$  in small amounts. With improvements to the process, the eventual aim is to provide a reliable source for pre-clinical trials across Europe.

The use of radioisotopes in medicine has completely shifted the landscape of cancer diagnostics and therapeutics over the past half century. The benefits to society are evident, and enhanced knowledge in the fundamental nuclear physics of the isotopes in question is of particular relevance. But when we talk about the actinides, we must acknowledge the legacy of fissionable materials. Nuclear physics has allowed us to tap into the vast amounts of energy that can be produced for the benefit of society, but it has also allowed us to cause substantial harm.

Environmental contamination by radionuclides is of particular concern when considering the legacy of nuclear accidents and nuclear weapons testing, but is also a relevant factor to consider in nuclear energy production. At the IRS in Hannover, Germany, Darcy van Eerten, ESR 12, works on rL-SNMS\*, a method to perform ultra-trace analysis of actinides in environmental samples.

rL-SNMS combines the sensitivity and versatility of a commercial TOF-SIMS\* with the selectivity of laser excitation schemes. In this way, ultra-trace levels of actinides can be distinguished from the environmental background. Testing the range and speed of elements detectable and establishing a standard methodology will allow for more diverse samples to be analysed with higher confidence.

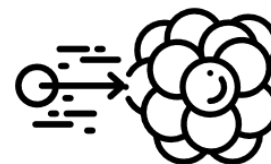
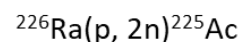
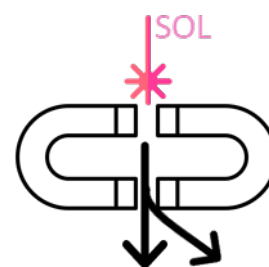
Within this work package, the LISA4Society project will be documenting the advances made in these efforts and making them accessible to the general public. Half the ESRs will be following the journey of Ac production as it is produced and transported to a hospital for actual use. The other team will be heading to the Chernobyl Exclusion Zone in June 2022 and learning about the lasting legacy of radionuclides in the environment.

The application of scientific advances to societal issues comes down to the practicality of the process. Additional challenges lie in navigating relevant regulations surrounding the transportation and use of radionuclides. With experiments, experience, and the inexhaustive hope that our machines don't break down, we start to slowly build the bridge between possibility and reality.

### Milking stockpiles



DISCONTINUED



TARGET PROBLEMS

Current actinium use is hampered by a dwindling supply. Enhancing the production process at CERN MEDICIS could improve the isotope supply for clinical trials.

\*

**ISOL— isotope  
separator on-line**

**rL-SNMS—resonant  
laser secondary  
neutral mass  
spectrometry**

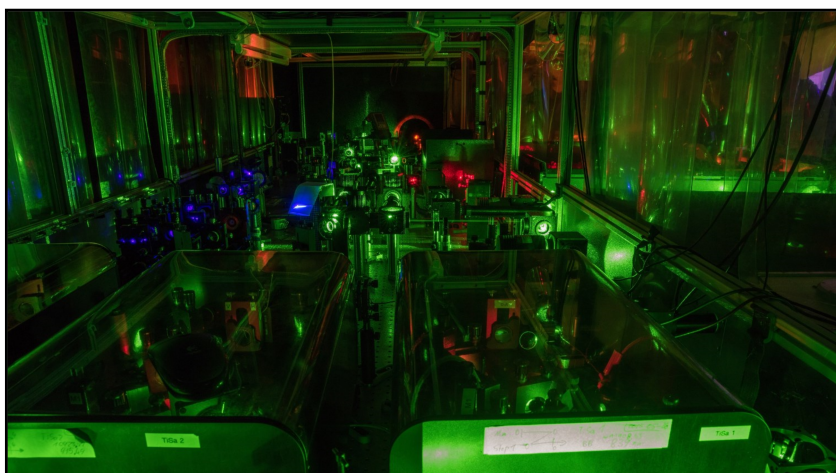
**TOF-SIMS—time-of-  
flight secondary  
ion mass  
spectrometry**



# Enhanced Understanding of Actinide Structure

An introduction to Work Package 4

by Mia Au



The RILIS laboratory at ISOLDE-CERN (2018) showing three TiSa lasers

**W**hat makes an actinide an actinide? The 15 actinides are the chemical elements with atomic numbers 89 through 103, starting with actinium and ending with lawrencium. The idea of the actinides as a chemical series was proposed in 1944, kicking off decades of theoretical and experimental research on actinide chemistry and materials. Now, the set of elements we call the actinides is known to scientists as a transition series, in which electrons are added to a specific electronic subshell. This special structure of the electrons—the atomic structure—gives rise to all the interesting chemical phenomena exhibited exclusively by the actinides.

Atomic structure is one of the most fundamental keys to truly understanding an element. The researchers in LISA's WP 4 aim to shed some light (literally!) on the atomic structure of these special elements, both through experimental and theoretical approaches.

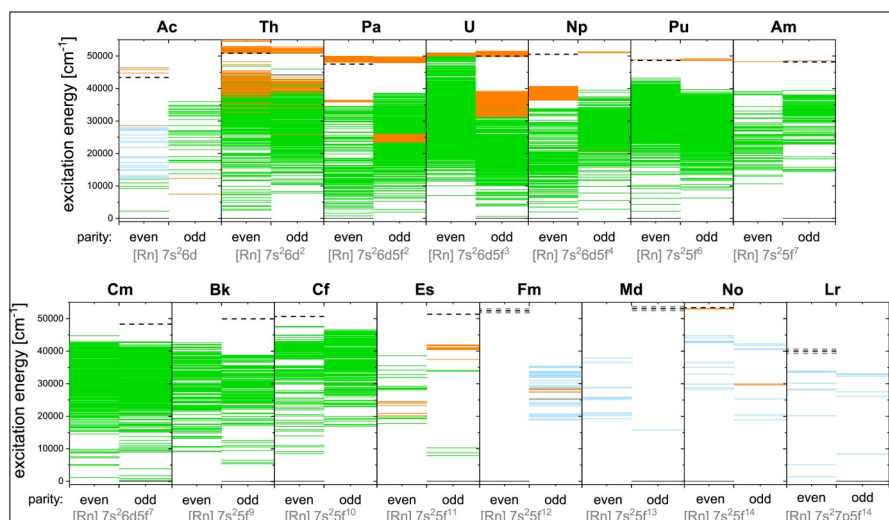
Ionization potentials offer one way to access atomic properties. At Johannes Gutenberg-Universität Mainz, Magdalena Kaja, ESR 5, works on laser spectroscopic investigations on the actinide atomic and nuclear structures, identifying resonance ionization schemes and determining ionization potentials of actinide elements.

As another probe of the actinide atomic structure, Miranda Nichols, ESR 6, studies negative ions at the University of Gothenburg, in Sweden. Electron affinity measurements identify the energy released by adding an electron to a neutral atom, giving information about the atomic structure. Miranda's work on collinear laser photodetachment spectroscopy of negative ions aims to access these properties for the actinide elements.

With many components to take into account, solving relativistic equations for heavy elements like the actinides is no easy task. Raphaël Crosa-Rossa, ESR 14, works at Rijksuniversiteit Groningen, in the Netherlands, on relativistic coupled

cluster and configuration interaction investigations to predict, study and validate atomic energy level structures of the actinides.

Theoretical predictions include ionization potentials and electron affinities, as well as hyperfine structure parameters. These numbers tie back into all the experiments on the actinides, helping guide and interpret experimental results, while also using experimental results to extract and infer more information about the actinide atomic structure.



Actinide level overview showing atomic lines in the actinide elements, taken from M. Block et al., PPNP 116 (2021), 103834 © 2020 Elsevier B.V.



# Exploring the Limits of Nuclear Existence

An introduction to Work Package 5  
by Darcy van Eerten

**I**n certain corridors of certain departments of Physics and Chemistry you will find a poster that is more ubiquitous than any other. More common than conference posters, more common than print outs of xkcd comics: it is the chart of nuclides. It is the fundamental resource of all researchers for whom the periodic table is merely the starting point. To see its use in action, fingers tracing up and down the elements and isotopes, is to marvel at the millions of hours of fundamental research condensed into a simple set of numbers and colours. It has been said that each square on the chart is the result of at least one doctoral thesis, if not multiple.

The exploration of nuclear existence continues, and not just at the far reaches of the nuclide chart but to the fundamental nuclear structure of the isotopes already discovered. This structure is what allows us to distinguish between isotopes that are otherwise chemically identical and potentially utilize their unique properties.

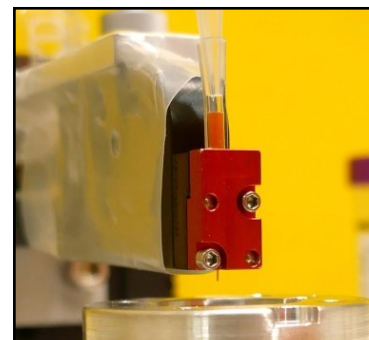
Optical spectroscopy methods over the last decades have aimed to “fill in” the nuclide chart with data on the nuclear structure of isotopes such as hyperfine structure, charge radii, nuclear moments, and nuclear spin. Within LISA, the researchers in WP5 are tackling the experimental challenges of isolating and measuring actinides and trans-actinides to benchmark theoretical calculations of nuclear properties. These challenges include the design and optimization of actinide targets, the methodology of in-gas-jet resonance ionization spectroscopy, and the measurement of actual actinide and transactinide isotopes.

To measure isotopes, you must first have isotopes to measure. Lauren Reed, ESR 11, at JGU Mainz will be developing actinide targets for use within the LISA project. Two methods for this have been identified, electrodeposition and drop-on-demand. While electrodeposition is a well-established technique, and may be optimized for certain target preparation, drop-on-demand provides the opportunity for higher yields and precision, but may lead to less stable performance.

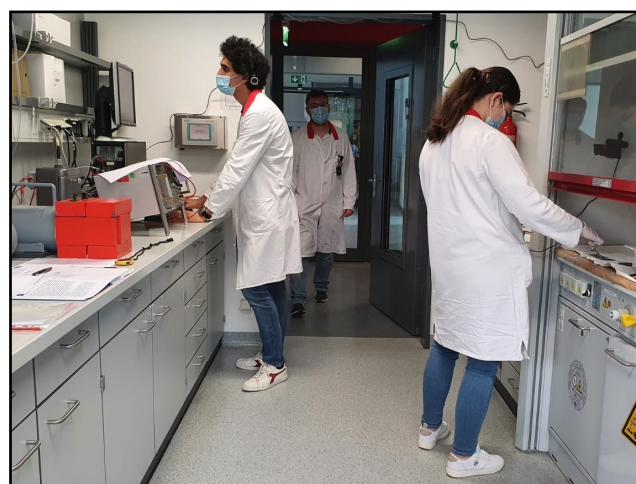
Two more projects are then devoted to the study of nuclear structure with the help of in-gas-jet methodology and other production techniques. Andrea Raggio, ESR 4, is at the University of Jyväskylä investigating fundamental ground state structure, including hyperfine structure and the octupole deformation of neutron deficient Th and U.

At the furthest reach of the actinides, Jessica Warbinek, ESR 10, at GSI Helmholtz is looking into the short-lived isotopes of nobelium and lawrencium. These heavy elements are produced in a fusion reaction, making them particularly challenging to handle due to their low yields and short lifetime. By employing alpha decay spectroscopy in combination with in-gas-jet techniques, hyperfine structures and ionization states can be found for these exotic elements.

The impact of having nuclear structure information at your fingertips cannot yet be known. The research could lead to ever more exotic elements at the edge of what is possible, or the development of more efficient production techniques for somewhat less exotic, but still rare elements. Even the most mundane library of known constants and excitation schemes can form the basis of cascading works that could transform science.



Drop on Demand system at JGU Mainz

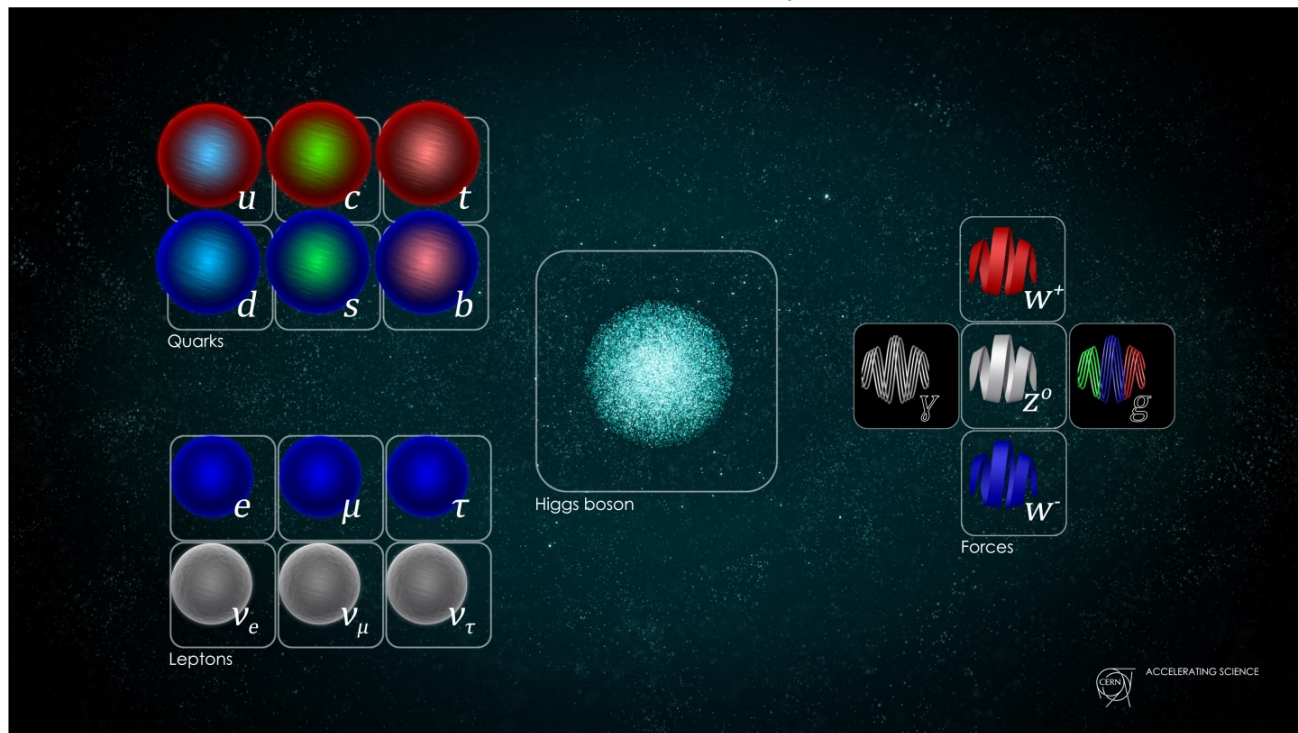


Andrea Raggio, ESR 4 (left) and Lauren Reed, ESR 11 (right) working together at JGU Mainz Nuclear Chemistry Department during the LISA training in June. Read more about their collaboration on page 18.



## ***LISA: Connections with physics beyond the standard model***

By Mia Au



Particles of the Standard Model of Particle Physics. Image by Daniel Dominguez, CERN [1]

**T**he big question: *How does our universe work?* The Standard Model of particle physics describes the interactions of fundamental particles that behave according to fundamental forces to make what we understand as matter. For decades, the Standard Model has endured many tests and survived as our best tool to explain many experimental results. So, what's missing?

One of the outstanding questions left unexplained by the standard model is the baryon asymmetry problem: why do we see more matter than antimatter? Looking for answers to these questions goes into BSM ("Beyond the Standard Model") Physics.

One condition that might result in production of matter and antimatter at different rates is violation of charge-parity symmetry (CP violation) or T-symmetry (CPT violation). CP violation can appear in the Standard Model through studies of the weak interaction, but it doesn't account for the observed matter-antimatter asymmetry. Atoms and molecules provide many ways to search for more answers [2].

Atomic parity violation is amplified for heavy atoms by orders of magnitude, scaling steeply with nuclear charge and motivating further spectroscopy of heavy nuclei and their isotopes. Atomic parity violation can probe a unique part of the Standard Model, making it complimentary to the investigations in high-energy physics.

Another condition that might explain the phenomenon of matter/antimatter asymmetry is the existence of an EDM (electric dipole moment) in any fundamental particle. Experiments that measure the upper limit of electric dipole moments can constrain the EDM as a source of asymmetry, putting experimental results beyond the reach of many BSM models of physics. The current limit on the electron EDM was measured in 2014 by the ACME collaboration [3] using beams of thorium monoxide — an actinide molecule.

[1] CERN <https://home.cern/science/physics/standard-model> (2020).

[2] M. Safronova, et al., *RMP* 90: 025008, (2018).

[3] J. Baron et al., *Science*, 343, 6168 : 269-272, (2014).

[4] V. Flambaum et al *PRL* 113 : 10, (2014).

[5] N. Hutzler et al. *Snowmass LOI*, <https://arxiv.org/abs/2010.08709>, (2020).

[6] L. Skripnikov et al., *PCCP*, 22, 33 18374-18380, (2020).

[7] R. Garcia Ruiz et al., *Nature* 581 : 396-400, (2020)

[8] MIT <https://web.mit.edu/radiomolecules/> (2021)



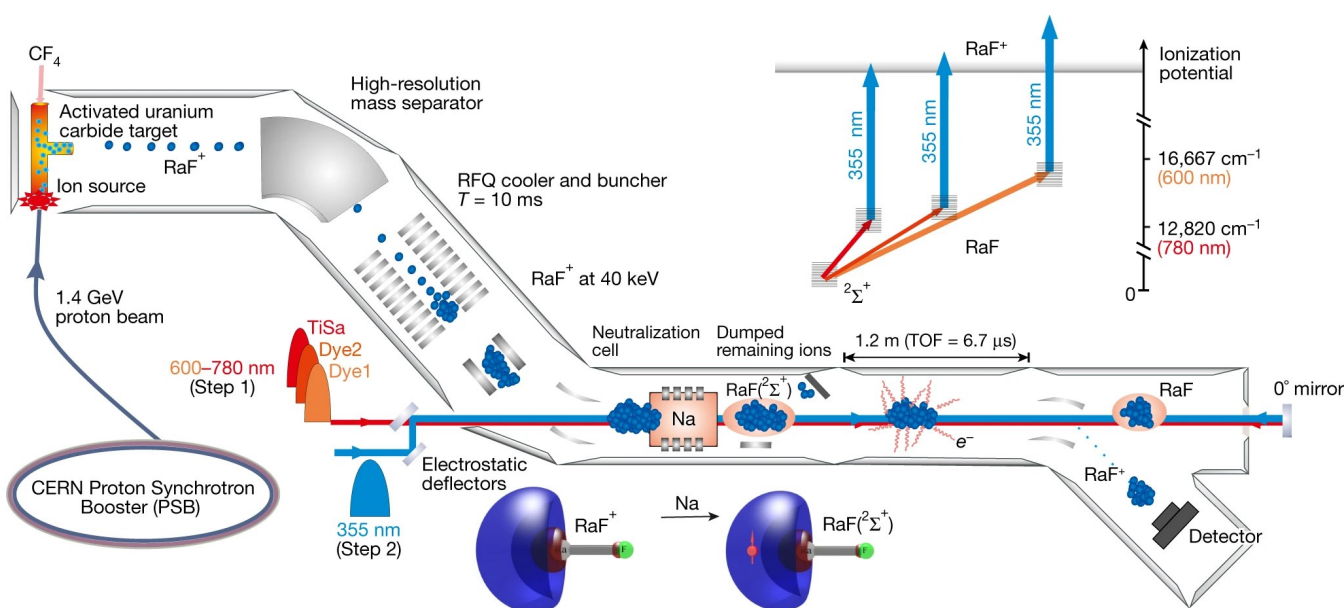
Time and parity violating energy shifts can be observed in some molecules as a result of nuclear contributions including the Schiff moment and the magnetic quadrupole moment. These observables can be connected theoretically to underlying time and parity-violating mechanisms in the nucleus, giving access to information about fundamental parameters involved in the matter-antimatter asymmetry problem. The nuclear Schiff moment is predicted to be large for actinide atoms, giving researchers another reason to dive into this region of the nuclear chart. The study of these actinide molecules promises “dramatic impact on the modern understanding of the nature of CP-violating fundamental interactions” [4].

Molecules feature dramatic energy shifts due to strong electric polarization, which can be exploited to connect underlying sources of CP violation with observed nuclear magnetic quadrupole moments [5]. The magnetic quadrupole moment can also be expressed in terms of fundamental parameters that feature in the matter-antimatter asymmetry problem, giving another potential way for measurements of molecules to improve our current limits on some of these underlying T,P-violating parameters.

There is significant potential for BSM physics through some of the nuclei being probed as part of the LISA project. Octupole deformation in some heavy actinide nuclei can lead to tremendously enhanced sensitivity of hadronic CP violation. Molecular species with these deformed nuclei could be orders of magnitude even more sensitive to these symmetry violations.

Recent work at ISOLDE with radioactive beams of radium monofluoride [6] has shown the capability of laser-spectroscopy experiments at such facilities to extract information from beams of radioactive molecules. The experiment was a collaborative endeavour requiring efforts in theory, radioactive ion beam production, and laser spectroscopy techniques. A community has grown from these efforts, bringing together theorists, technical experts, and experimentalists at events such as the New Opportunities for Fundamental Physics Research with Radioactive Molecules [8].

Within the LISA project, investigations regarding the production of some of these promising actinide molecules are already underway, while theorists tackle the challenge of predicting and understanding these molecules, and experimentalists develop new techniques to access information from the beams. These developments within the LISA program will contribute greatly to the search for new physics.



Experimental scheme of the radium fluoride experiment at CERN-ISOLDE [7]. Radioactive radium isotopes were created by impinging protons on a target. Radium monofluoride cations (RaF<sup>+</sup>) were produced and extracted from the source. Cooled bunches of molecular ions were extracted and neutralized in flight by charge exchange. Neutral RaF molecules were overlapped with different laser beams and resonantly reionized molecules were deflected. Top right: resonance ionization scheme. Bottom: molecular orbital schematics. © Springer Nature (2020)

## A very brief history of actinides in the environment

By Darcy van Eerten



Decontamination during the Chernobyl accident. From the IAEA image bank, Copyright: IAEA, Photo Credit: USFCRFC

**T**o be “in Nuclear” - the field, the state of being - is to be assumed to be intimately familiar with its entire global history. This isn't entirely unreasonable, as its history is fairly short and comprehensive, with only a limited number of key players. Most of this knowledge comes with time and experience, often gained first-hand. “Young” in nuclear generally means anyone under 40, or those who weren't alive during the Chernobyl accident.

The actinides created in the span of a lifetime, sometimes called the Atomic Age, can have half-lives of thousands of years. Their complicated chemistry means some elements (U, Pu) can exist in multiple oxidation states, affecting whether they move quickly, or slowly in the environment. It is therefore inevitably the responsibility of the youth to understand and appropriately handle the actinides that have ended up in our environment. Knowing how they got to be there in

the first place, and how that affects their behaviour in the environment, is foundational to LISA's work.

### Natural sources

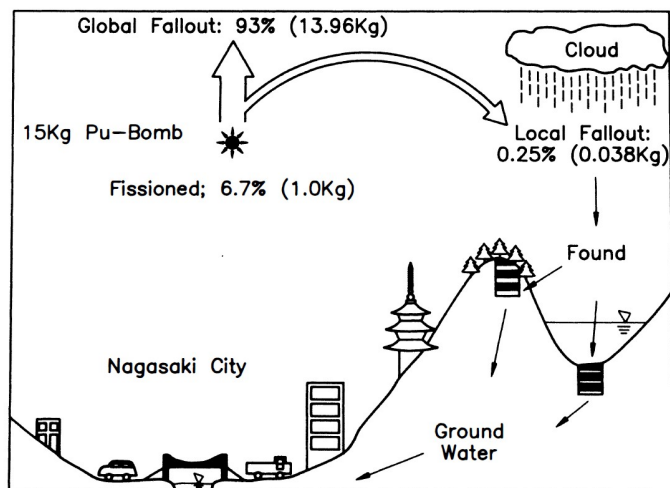
The actinides are of course united under a defining characteristic in that they are all radioactive, and so decay away after roughly five half-lives. To be considered primordial, the elements must have a half-life longer than the earth itself, or else derive from decay products in the following billions of years. Thorium and uranium are the only actinides to fit the former description.  $^{232}\text{Th}$  is the most abundant at 7.2 ppm in the earth's crust, with  $^{238}\text{U}$  and  $^{235}\text{U}$  (0.7% total U) closely following at 3 ppm. The world mean for the activities from these elements is 30 and 35 Bq/kg respectively, but higher at mining sites [1].

Technically speaking, there are more natural actinides, but they only contribute a vanishingly small percentage of the earth's crust.  $^{239}\text{Pu}$ , derived from neutron absorption by  $^{238}\text{U}$ , is the most abundant of these. Pre-1945,  $^{239}\text{Pu}$  could be found at roughly 1  $\mu\text{Bq/kg}$  in the earth's crust [2]. For reference, the German exemption limit under which you do not have to report handling of Pu to a competent authority is 0.1 Bq/g [3].  $^{244}\text{Pu}$  was created in the r-process alongside U and Th, but has a much shorter half-life of a mere 80 million years. An estimated 0.2-7 g of it remains in the entire earth's crust today [2].

### Nuclear Weapons

The dawn of the Atomic Age changes the picture rather drastically. Bombs that are released in the atmosphere are typically exploded several hundred meters in the air, and typically fission only a portion of the material. For the Nagasaki A-bomb, only 1.2 kg of plutonium was fissioned, while 13.8 kg was released into the atmosphere, contributing to what is known as “Global fallout” that can be measured as far as the Arctic [4].

The majority of the atmospheric tests were conducted in the 1950s and 1960s, after which a test ban was enforced and testing moved underground. Hardly discrete affairs, the locations for the tests were desolate or unpopulated areas, typically deserts and remote islands. The list includes Semipalatinsk (formerly Soviet Kazakhstan), Nevada (USA), Lop Nur (Xinjiang, China), the Sahara (formerly French Algeria), and many pacific islands colonized by the Americans,



Source of global fallout from the Nagasaki A-bomb. Kudo et al. [3].

(formerly French Algeria), and many pacific islands colonized by the Americans,



British, and French.

Some 6 tons of Pu were released during these years, predominantly in the northern hemisphere [5]. The air concentration has decreased exponentially since the end of atmospheric testing in 1970 to effectively zero today ( $<1$  nBq/m<sup>3</sup>). However, detectable amounts of Pu remain in the soil (in the order of Bq/kg, but higher at test sites [2]). Because weapons-grade Pu is over 90% <sup>239</sup>Pu, it remains a clear and important marker in nuclear forensics and radioecology to this day.

## The Nuclear Fuel Cycle

Global fallout is the standard upon which Pu measurements of the environment are measured. Significant deviation from either the concentration or isotope ratios indicates another source has to be considered. Nuclear forensics uses the multiple indicators of the nuclear fuel cycle (the actinides but also fission/activation products) to determine the origin of a given unknown sample. From uranium ore, to enriched uranium, to (mixed) fuel, to spent fuel, each step contributes elements or removes them. Plutonium and the so-called minor actinides, Am, Cm and even Cf, are bred in irradiated fuel and make up a small portion ( $<2\%$ ) of spent nuclear fuel.

Reprocessing plants separate the still usable U and Pu to reuse in fuel, or separate Pu for use in nuclear weapons. They have been relatively consistent sources of contamination due to regular discharge of effluent. Much has been written about Sellafield (Northwest of England), La Hague (Northern France), and Savannah River (South Carolina, USA), which are closely monitored by their respective regulatory bodies. Much more has been speculated about Mayak (Russia), which has a complicated legacy of multiple accidents and secrecy during Soviet operation.

This informs the debate on open and closed fuel cycles [6]. In an open cycle, the spent fuel is not treated further and stored in a geological repository after interim storage of some decades. A closed cycle envisions the spent fuel being reused in a fast neutron reactor, which are still in development. Both sides argue that environmental contamination is reduced, either by not involving reprocessing, or by reducing/eliminating the amount and nature of nuclear waste.

## Remediation

The accident in Chernobyl released actinides into the environment, atmospherically but also in the form of small fractions of intact fuel elements known as hot particles. These particles can be found throughout what is now the Chernobyl radiological reserve. A concrete sarcophagus was built within a year around the destroyed reactor number 4, which encased the worst of the contamination in concrete. The 'New Safe Confinement', essentially a large hangar replaced the sarcophagus in recent years.

A similar tactic was used in the former test site Semipalatinsk, where almost 300 kg of Pu in various states had been abandoned by the Russians when the USSR collapsed. The efforts to encase the Pu in concrete took over a decade - it's truly a fascinating story for anyone interested [7].

Actinide contamination in Fukushima appears to be limited to the rubble around the reactor and contaminated water used to cool the damaged fuel, although there are indications that some was still dispersed to vegetation nearby [8, 9]. Decontamination efforts in this area are ongoing in the process of decommissioning the powerplant.

## Conclusions

In a short span of time, we have inalterably changed the makeup of our planet through anthropogenic actinides. The activities in the early days of nuclear technology are still measurable today, and it is clear that the ban on atmospheric testing was absolutely necessary to prevent adverse health outcomes for the global population. But we should be careful that when we say something is measurable, it does not necessarily mean it is dangerous.

The quantification of risk, particularly of the actinides in the environment, is an ongoing challenge. How does one weigh up an edible mushroom from Chernobyl today with a poisonous mushroom anywhere else? Shouldn't we treat plastic, which will not degrade in our lifetime, as carefully as nuclear waste? We must continue to do the work needed to solve these questions, and in so doing advocate for the safety and prosperity of everyone.

[1] C. Nwankwo et al. *J. Radiat. Res. Appl. Sci.*, **8**, 2, (2015).

[2] D. Taylor, *Plutonium in the Environment*, A. Kudo (Editor) (2001).

[3] [http://www.gesetze-im-internet.de/strlrschv\\_2018/](http://www.gesetze-im-internet.de/strlrschv_2018/)

[4] A. Kudo et al., *Water Sci Technol*, **23**(1-3): 291-300 (1991).

[5] J. Harley, *Japan J Radiat Res*, **23**, 83-104 (1980).

[6] JRC, *EASAC policy report no. 24* (2014).

[7] E. Harrell et al., *Managing the Atom Project*, Belfer Center, August 15, (2013).

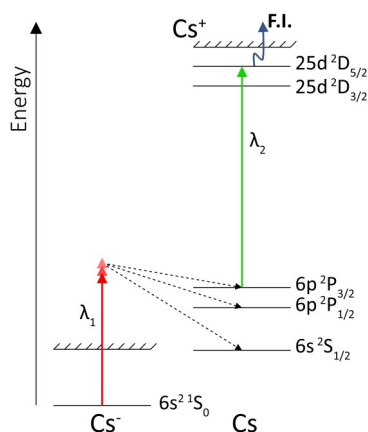
[8] Y. Koma et al., *Radiochim. Acta*, **107**(9-11): 965-977 (2019).

[9] S. Schneider et al., *Sci Rep* **3**, 2988 (2013).

# RESEARCH HIGHLIGHTS

## High Resolution measurement of electron affinity of Cs

By Miranda Nichols—ESR 6



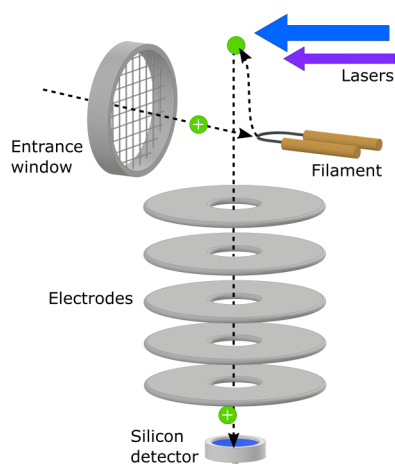
Cs excitation scheme.

The laser spectroscopy research group at the University of Gothenburg have recently submitted a paper to be published titled, “A high-resolution measurement of the electron affinity of Cs”. This paper presents an updated experimental value for the electron affinity of Cs via investigations of a partial photodetachment cross section for Cs. A resonance ionization scheme allowed for final-state selectivity which enabled them to improve the measurement by a factor of two from the previous most accurate published value.

The two-step excitation method used in this paper can be applied to heavier alkali metals such as francium, a near-actinide element. The electron affinity for Fr remains experimentally unknown. Measuring this would complete the EA measurements for the alkali metals and could help us understand more about electron-electron correlation and relativistic effects of heavy elements. These studies will pave the way for future studies on the actinides.

## Optimum Filament Setup for Efficient Lr Evaporation

By Jessica Warbinek—ESR 10



Principle of the RADRIS technique. Fusion products are collected and neutralized on a filament, followed by their evaporation, resonant laser ionization and detection via their alpha decay.

The heaviest actinides can only be accessed via fusion-evaporation reactions at large accelerator facilities and are only available in quantities of a few nuclei per second or less. Due to their short lifetimes, these exotic nuclei need to be studied directly after their production and separation from the primary beam.

Lately, the RADiation Detected Resonance Ionization Spectroscopy (RADRIS) allowed the first laser spectroscopic investigation of different nobelium isotopes. Here, produced recoil nuclei are collected and neutralized on a catcher filament in a gas cell, followed by their evaporation, resonant laser ionization and eventually their detection. Turning to the heaviest actinide, lawrencium, demands the adaptation of the filament due to disadvantages of the tantalum filament type previously used for nobelium evaporation.

Recently, the first on-line tests on the desorption of lawrencium from this new filament type have shown an efficient desorption as well as an adequate stability for laser spectroscopic investigations on lawrencium. It now enables on-line laser spectroscopy of lawrencium applying the RADRIS technique for the search of first experimentally determined atomic levels.

Hübner Photonics





## Application of fast scanning option for multi-actinide detection

By Darcy van Eerten—ESR 12

Hot particles from the Chernobyl exclusion zone are particularly interesting to measure on an individual level because of their unique form and varying composition. The actinide composition is dominated by  $^{238}\text{U}$ , but the trace levels of Pu and Am are different for each particle resulting from different irradiation times in the reactor.

Fast scanning of individual actinides in the particles has been made possible through the use of two-step excitation schemes and a diffraction-grating Ti:Sa laser in combination with time-of-flight mass spectrometry. By rotating on a motor, the diffraction grating can access the wavelengths required for exciting uranium, plutonium, and americium, all measured in a single day. This greatly reduces the time to measure each individual particle, demonstrating the versatility of the rL-SNMS technique.

## Experiences in the ISOL extraction of $^{225}\text{Ac}$ at CERN MEDICIS

By Jake Johnson —ESR 1

One of the objectives of the LISA network is the application of knowledge of actinide spectroscopy for societal benefit. We are harnessing laser ionization and mass separation to produce radio-isotopes such as  $^{225}\text{Ac}$  for cancer diagnostic and treatment at CERN MEDICIS [1]. Since commissioning in 2017, MEDICIS has been rapidly developing to expand the theranostic toolkit of radioisotopes and deliver them to partner institutions for preclinical trials. Recently, significant efforts have been invested to produce  $^{225}\text{Ac}$ , a highly promising candidate for targeted alpha therapy of metastases.

In two separate offline collections, samples of actinium were obtained from JRC Karlsruhe and then collected at MEDICIS using laser developments at the facility. Two actinium ionization schemes were employed simultaneously with an automated stabilization system, enabling constant and reliable ionization efficiency to be achieved during the collections. The samples were then sent to KU Leuven and SCK CEN where decay spectroscopy and dissolution tests were performed. The collected  $^{225}\text{Ac}$  could be dissolved into a solution appropriate for radio-labelling. Future efforts aim to supply greater quantities of  $^{225}\text{Ac}$  to support pre-clinical investigations.

## Developments for actinide beams at ISOLDE

By Mia Au—ESR 3

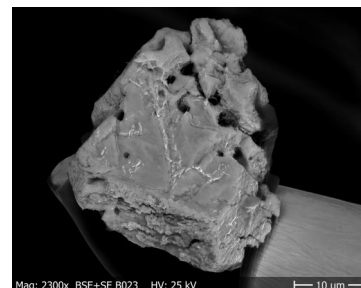
Developments for actinide production are critical to further study of the actinides. At ISOLDE, a previously irradiated uranium carbide (UCx) target was used to investigate actinide beams in the pre-proton commissioning period.

A vacuum chamber with a single-ion detector was purchased, assembled and installed in the ISOLDE GLM beamline, then used to study laser-ionized beams of long-lived actinides. ISOLDE began taking protons on the target for commissioning, facilitating production of radiogenic species for further study. The ISOLTRAP MR-ToF high-resolution mass separator [1] was used to help identify the species through time-of-flight measurements. The developmental beams for the LISA network are also relevant for several Letters of Intent submitted to the ISOLDE and n\_TOF Experiments Committee [2], [3].

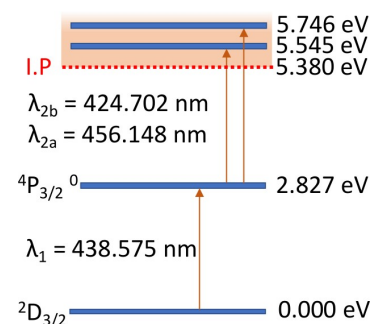
[1] R. N. Wolf et al., *IJMS* 349-350.1: 123-133, (2013).

[2] Wright, T. CERN-INTC-2021-009; INTC-I-224, (2021)

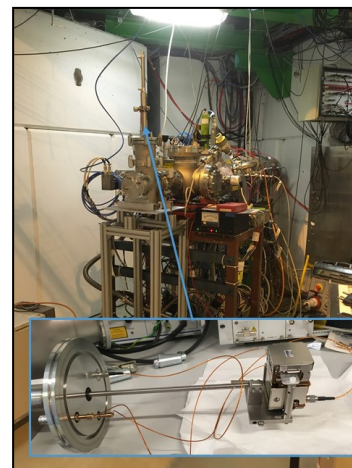
[3] Athanasakis, M. et al. CERN-INTC-2021-017; INTC-I-227, (2021)



Ophaniel: Particle from the Chernobyl exclusion zone (CEZ), glued to a tungsten needle.



The 2-step laser resonance ionization scheme for atomic actinium.



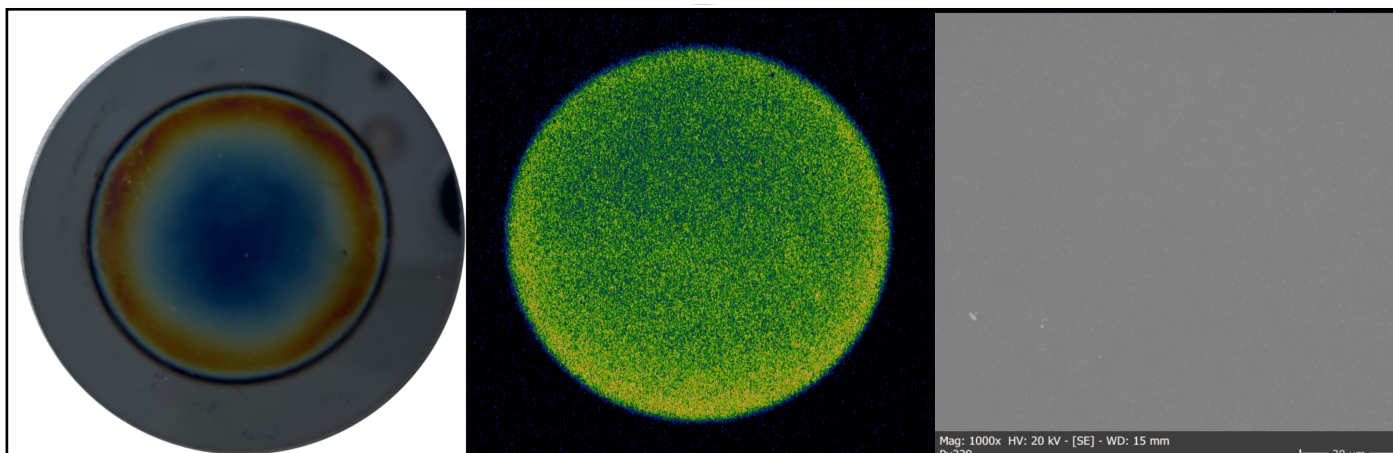
MagneToF detector installed at the GLM beamline in ISOLDE for single ion counting and detection of low intensity beams



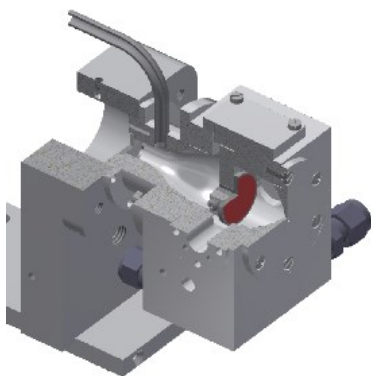
## Developments towards high resolution

### spectroscopy of $^{235m}\text{U}$

Andrea Raggio - ESR 4, Lauren Reed - ESR 11,  
Jessica Warbinek - ESR 10



Picture (left), radiographic image (middle) and SEM x1000 picture (right) of a  $^{239}\text{Pu}$  source produced at JGU.



Cross-sectional view of the actinide gas cell CAD model with the alpha recoil source mounted facing the nozzle.

The measurement of hyperfine structures and isotope shifts with high-resolution laser spectroscopy offers access to the extraction of fundamental nuclear structure properties including spins, mean-square charge radii and electromagnetic moments. The application of this method to the heaviest elements, the actinides, is particularly challenging due to limiting factors including the lack of naturally abundant stable isotopes for prerequisite atomic spectroscopy and challenging online production cross sections. For example, in uranium, only five isotopes have been addressed by laser spectroscopy so far ( $^{233-236,238}\text{U}$ ) [1].

Within the LISA project, a new measurement campaign has been started on uranium by means of collinear laser spectroscopy at the IGISOL facility, University of Jyväskylä. The aim of the current work is to apply this technique to the study of the second lowest-lying isomeric state in the nuclear landscape, that of the 76-eV isomer in  $^{235}\text{U}$ . The production of a  $^{235m}\text{U}$  beam is achieved using a  $^{239}\text{Pu}$  alpha-recoil source, developed at the Johannes Gutenberg University of Mainz. The source will be mounted in the IGISOL actinide gas cell [2, 3] and  $^{235m}\text{U}$  recoils will be extracted for collinear laser spectroscopy studies.

The source development is a critical aspect of this work, since thickness and composition have a large effect on the recoil efficiency [4]. A set of sources with different characteristics (diameter, activity, substrate material) have been produced at Mainz and are under analysis at Jyväskylä using a combination of spectroscopic methods including direct alpha spectroscopy, gamma-ray spectroscopy and Rutherford Backscattering Spectrometry. This aims to support our understanding of how the molecular plating technique [5, 6, 7] affects the recoil efficiency. The optimal source will be used in the foreseen laser spectroscopy experiment.

Meanwhile, in the last week of July, a successful experiment studied the three natural isotopes,  $^{234,235,238}\text{U}$ , produced with an electric discharge source. More than 10 different transitions in the singly-charged ion, in a wavelength range of 288 to 314 nm, were recorded for each isotope. These data include a new set of hyperfine constants of the levels involved (in  $^{235}\text{U}$ ), providing a challenge to atomic theory. The most efficient transition will serve for the study of  $^{235m}\text{U}$  in combination with the new Pu sources in the future.

- [1] M. Block et al., *PPNP*, 116 (2021), 103834
- [2] I. Pohjalainen et al., *NIM B*, 376 (2016): 233-239.
- [3] I. Pohjalainen et al., *NIM B*, 484 (2020): 59-70.
- [4] I. Pohjalainen et al., *NIM B*, 463 (2020): 441-448.
- [5] A. Vascon et al., *NIM A*, 696 (2012): 180{191}.
- [6] N. Trautmann et al., *NIM A*, 282 (1989): 102-106.
- [7] W. Parker et al., *NIM*, 16 (1962): 355-557.

## ***Preparation for the determination of the electron affinity of polonium***

Miranda Nichols—ESR 6, Raphaël Crosa Rossa—ESR 14

Polonium is a near-actinide element of which the electron affinity remains experimentally unknown. The electron affinity determines the chemical reactivity and, indirectly, the compounds the element can form as well as the stability of its chemical bonds. From a theoretical standpoint, electron affinity investigations are of interest because they can serve as a precise probe for electron correlation. Polonium is of particular interest due to the prevalence of relativistic effects in its atomic structure as well as its two-hole electron configuration.

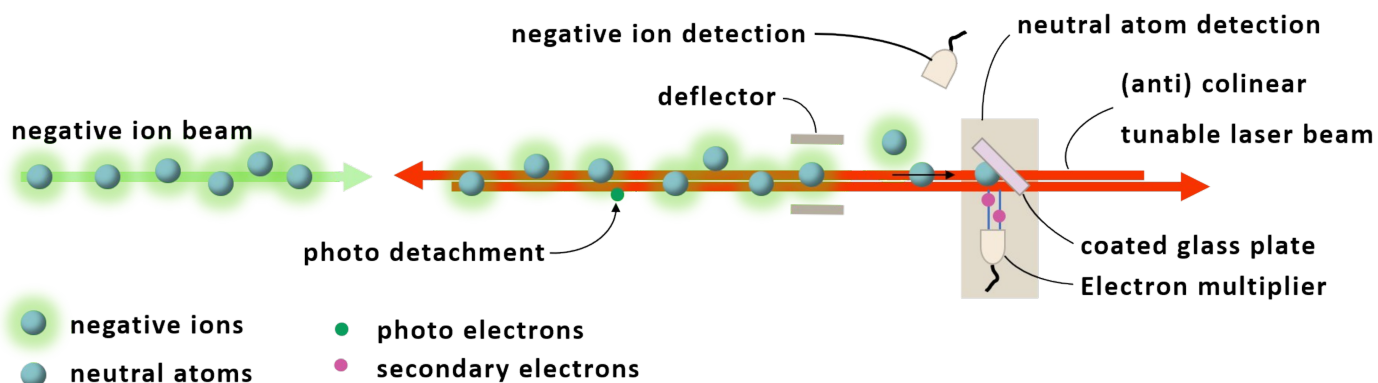
A letter of intent has been accepted to perform negative ion beam preparation work at ISOLDE for the eventual determination of the electron affinity. The experiment will be in collaboration between the University of Gothenburg and the CRIS beamline as well as theoretical work from the University of Groningen.

Theoretically, a main challenge is to consider both electron correlation and relativistic effects, as they cannot simply be neglected for elements with a high atomic number. To do so, one needs to use high accuracy *ab initio* methods as coupled cluster, which, combined with the Dirac-Coulomb hamiltonian within the DIRAC program. This can allow one to correctly predict the fundamental atomic properties such as the electron affinity.

After extensively testing the computational parameters, consistent mainly of the basis set used, a first value of 1.429 eV has been calculated, which is in excellent agreement with the literature. Soon, the final value will be obtained by extrapolating to the complete basis set limit as well as calculating the Breit contribution and the QED effects. This will not only enhance the quality of the results, but also allow to estimate the uncertainty related to this prediction.

This experiment will require a negative ion beam of polonium which we plan to produce via a charge exchange cell. Beginning with a positively charged ion beam, we can produce a negatively charged ion beam by means of interaction with an alkaline vapor where two separate electron capture processes will occur. In preparation for this experiment, we first plan to test the negative ion production off-line at CRIS using any element. After this, we will investigate the charge exchange efficiency using a lighter homologue of polonium, tellurium. After preliminary tests and beam tuning are complete, we will use collinear laser photodetachment threshold spectroscopy to measure the electron affinity of polonium. This involves a tunable Ti:Sa laser which will be used to measure the cross section of electron photodetachment from the Po atom. Thanks to high precision theoretical calculations, we will have a narrow wavelength scanning range for the threshold energy investigation.

Measuring the electron affinity of polonium is an important step towards the LISA program deliverable of measuring the electron affinity of an actinide. With this information, we can improve atomic structure theory for heavy elements as well as gain experimental knowledge on increasingly complex systems. In the process, we plan to build a new beamline at CRIS which will allow us to study any radioactive isotope, and actinides in particular.

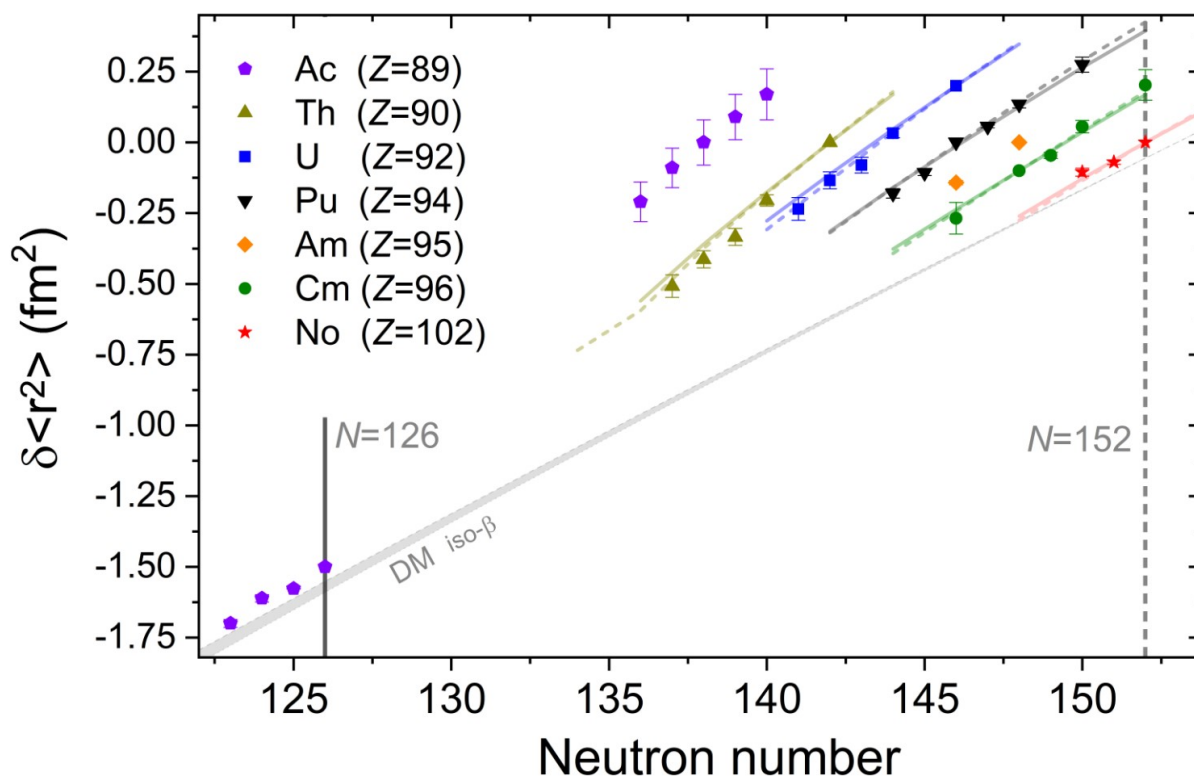


Collinear laser photodetachment spectroscopy will be used to measure the electron affinity. The frequency tunable laser beam is overlapped with the negative ion beam. The photodetachment takes place in the overlap region. Non-detached ions are deflected, and the photo-neutrals are detected via secondary electron generation in a graphene coated glass plate.



## Laser Spectroscopy of the Actinides at GSI

Elli Rickert-GSI, Jessica Warbinek—ESR 10



Changes in the mean square charge radii based on laser spectroscopy data for a wide range of actinide isotopes, taken from [1].

Laser spectroscopy has proven to be a powerful tool for the investigation of the nuclear structure of the actinides and more exotic nuclei. The heavier actinides and super heavy elements are especially interesting to be studied via laser spectroscopy due to relativistic effects, quantum electrodynamics and electron correlations leading to close lying levels, which may result in a variation from the expected ground state.

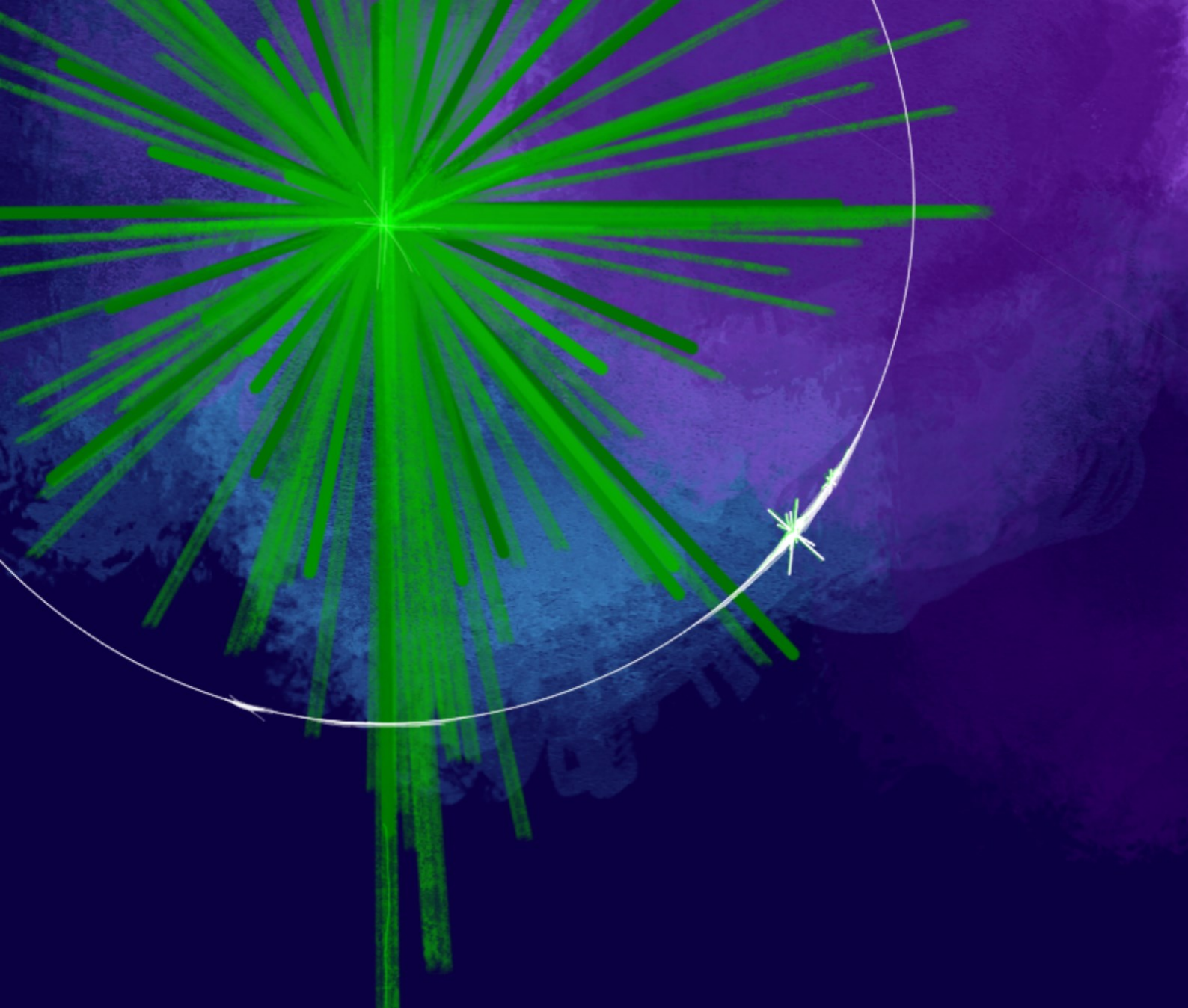
In recent years, the development of the buffer-gas-stopping technique enabled the study of more exotic nuclei in the range of the heaviest actinides far off from stability. Due to low production rates and short lifetimes, the application of laser spectroscopy yields additional challenges. Only recently, RADIATION DETECTED RESONANCE IONIZATION SPECTROSCOPY (RADRIS) enabled the study of the first experimentally observed atomic level in nobelium (Z=102) [1][2].

For the RADRIS technique, the actinide isotope of choice is produced in a fusion-evaporation reaction before the recoil nuclei are separated from the primary beam in the velocity filter SHIP. The transmitted fusion products are stopped in a buffer-gas cell and subsequently collected and neutralized on a catcher filament. After a sufficient accumulation on the filament, collected neutral atoms are evaporated and two-step laser resonance ionization spectroscopy is performed. In this way produced ions are finally detected via their alpha decay.

Recent laser spectroscopy experiments on the actinides helped to cover a range of nuclides between the spherical shell closure at N=126 and the weak deformed shell closure at N=152, as very well described in [3]. As visible in the figure, the respective charge radii increase roughly linearly, but with different slopes. A prolate deformation of the investigated isotopes can be concluded from that.

Upcoming experimental campaigns will extend the laser resonance ionization spectroscopy to the heaviest actinide, lawrencium (Z=103). Prerequisite experiments for the adaption of the RADRIS technique highlighted earlier in this newsletter by Jessica Warbinek. This will help shed light towards the super heavy elements.

[1] S. Raeder et al., *Phys. Rev. Lett.*, **120** (2018) 232503  
[2] A. Borschevsky, et al., *Phys. Rev. A* **75** (2007) 042514  
[3] M. Block, M. Laatiaoui, S. Raeder, *Progr. in Part. and Nuc. Phys.*, **116** (2021) 103834



# LISA WINTER SCHOOL

La Laguna, Tenerife  
15-19 February 2022

*Structure of Complex Atoms*

Payment opening: 1 September 2021  
Abstract deadline: 20 November 2021  
Confirmation of abstract acceptance: 13 December 2021  
Registration deadline: 23 December 2021

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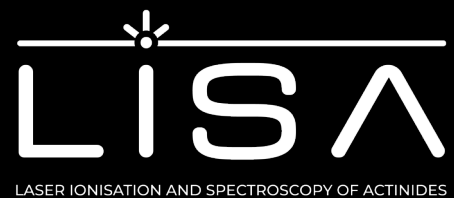


LISA with the help of Hübner foundation  
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**LISA**  
LASER IONISATION AND SPECTROSCOPY OF ACTINIDES



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