

EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH
Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Mass measurements of neutron-rich Ag and In isotopes
for r-process nucleosynthesis studies

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Abstract:

We propose to measure the masses of neutron-rich silver and indium isotopes – ^{124–129}Ag and ^{124–135}In – with the online mass spectrometer ISOLTRAP. The determination of these masses around $A=130$, and close to the magic $N=82$ shell closure would provide important missing data for understanding the nucleosynthesis of elements produced in the r-process. In addition to the masses of the ground states, we aim to measure the masses of their long-lived isomers, which play a key role in setting the timescale for the release of nuclear energy during the radioactive decay that follows r-process nucleosynthesis in neutron star mergers. These measurements will also provide the isomeric-to-ground state ratios of the exotic silver and indium isotopes that will contribute to the identification of the production site of actinides in the universe.

Requested shifts: 32 shifts, (split into 2 runs)



1 Motivation

Heavy elements are known to be produced through the rapid neutron capture process (r-process), but the exact synthesis sites remain debated and efforts are ongoing to achieve a quantitative description of elemental abundances. The only experimentally confirmed site is neutron star mergers (NSM). Extreme neutron-rich nuclei are synthesised in the r-process and during their decay, energy is produced and radiated away as a kilonova. This was observed following the detection of gravitational waves from the merger of two neutron stars, GW170817 [1]. Moreover, the synthesis of Sr was detected in the spectrum of this kilonova [2]. In addition, observations of elements in the atmospheres of the oldest stars (low metallicity stars) suggest different possible contributions to the production of heavy elements by the r-process [3]. While many stars have a robust pattern (same relative abundances among stars and the solar r-process) between the second ($A=130$) and third ($A=195$) r-process peak, there are clear variations for the region below the second peak [4]. This points to an additional weak r-process contributing to these elements between Sr and Ag. Moreover, U and Th also show variations that can be extreme as in the Ac-boost stars [5]. Thus, there are still many open questions about the origin and detailed conditions for the production of r-process elements. We know that atomic masses impact r-process calculations at different levels, but especially through neutron separation energies used to determine photo-dissociation rates from calculated neutron capture rates via the detailed balance [6]. Therefore, in this proposal, we aim to measure atomic masses of neutron-rich nuclides, including ground states and long-lived isomeric states, around the $N=82$ shell closure. This region is critical to understand not only the second r-process but also its robustness, variations beyond the third peak, and potential fingerprints on the kilonova due to the presence of isomers.

1.1 Mass measurements for the second peak of the r-process

This proposal aims to measure nuclear masses crucial for understanding the nucleosynthesis of elements produced in the r-process, in particular those around $A=130$, which form the second most abundant peak after strontium.

Current studies are still limited by uncertainties in nuclear data, particularly nuclear masses near $N=82$. We therefore propose to measure the masses of neutron-rich isotopes of indium (In, $Z=49$) and silver (Ag, $Z=47$) below and above the $N=82$ shell closure using the ISOLTRAP mass spectrometer. These isotopes significantly influence the determination of the r-process path. Sensitivity studies of the r-process reveal that, regardless of the mass model or astrophysical scenario used [7, 6], the region around $N=82$ is the most critical as shown in Figure 1. In particular, the masses of neutron-rich isotopes of Rh, Pd, Ag, Cd, In, Sn, and Sb have the greatest impact on final abundances. Since many of these nuclear masses are unknown, their measurement is essential for understanding the flow of matter across the $N=82$ shell closure and for determining the astrophysical conditions responsible for the $A=130$ peak in solar abundance. The masses of interest are highlighted with red rectangles in Figure 1 which shows that they are directly on the path of the r-process. Additionally, these measurements will serve to benchmark theoretical mass models, enhancing the accuracy of mass predictions for isotopes that are out of reach experimentally. Silver isotopes beyond $A = 125$ have never been measured before, however, the masses of indium isotopes have been measured up to ^{134}In using

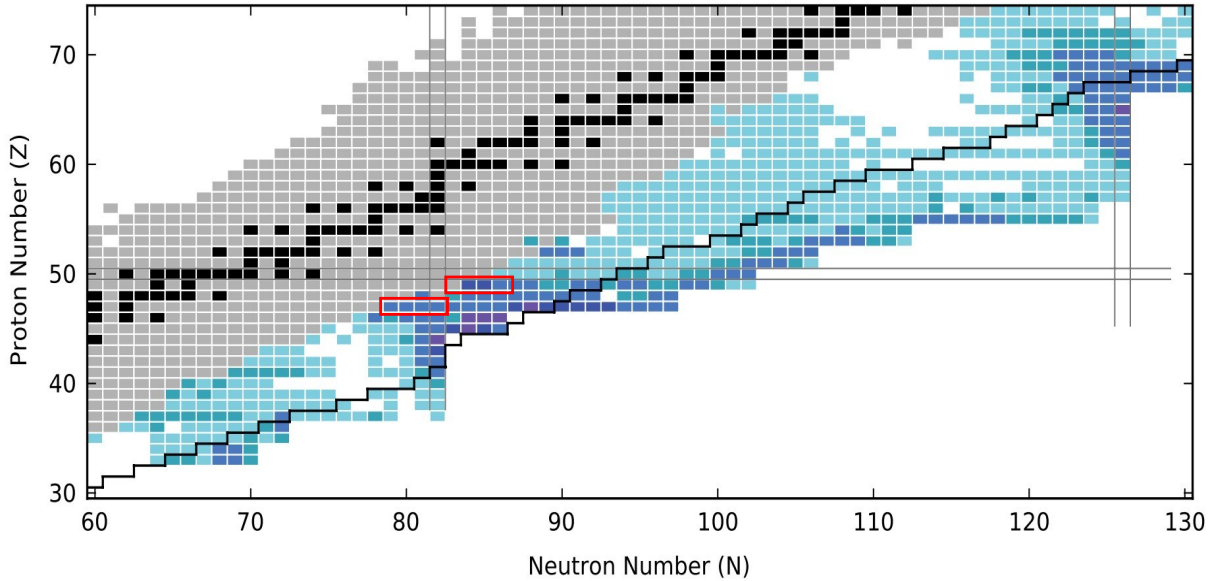


Figure 1: Figure taken from Ref. [6] showing nuclei that significantly impact final r-process abundances in neutron star merger event, with most influential masses in darker color. The masses that will be measured are highlighted by red boxes, which are $^{126-129}\text{Ag}$ and $^{132-135}\text{In}$.

the MRTOF MS of the TITAN/TRIUMF facility [8]. We intend to use the phase-imaging ion-cyclotron resonance (PI-ICR) technique, which will allow to improve the precision by at least an order of magnitude. The lighter silver isotopes $^{124,125}\text{Ag}$ were recently measured using the PI-ICR technique at the JYFLTRAP Penning trap [9].

In addition, several of the proposed isotopes of silver and indium have long-lived isomeric states that could be separated and their excitation energies measured using the PI-ICR method of the ISOLTRAP's Penning trap. Isomers have recently been suggested to play a key role in adjusting the timescale for nuclear energy release during the radioactive decay that follows r-process nucleosynthesis in neutron star mergers, thus affecting the kilonovae light curve [10, 11]. As recently demonstrated [12], high-precision measurements of the excitation energies of β -decaying isomers provide valuable insights into the dynamics of the r-process. Many of the isotopes of interest have at least one long-lived isomeric state, namely $^{125m,126m,127m,127n,129m}\text{Ag}$ and ^{133m}In [13].

1.2 Isomeric yield-ratio measurements for actinides production site

During the mass measurement using the PI-ICR technique, the isomeric-to ground-state ratio produced in the uranium fission can be determined in the same measurement¹. This ratio could be valuable in revealing the production site of actinides, whose presence has not yet been confirmed through direct observation in neutron star merger (NSM) events. Recently,

¹when long-lived isomeric states exist

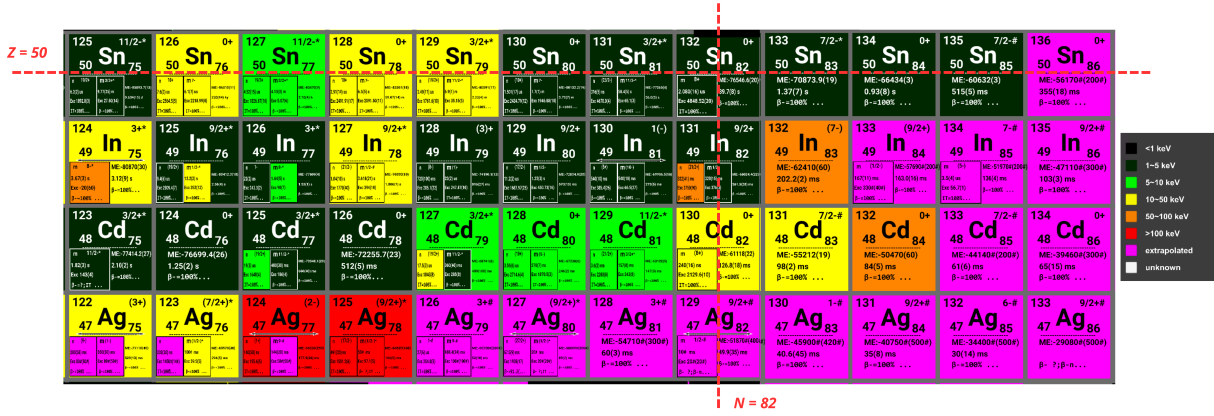


Figure 2: Extract from the nuclear chart of the new NucleusPlus software (2021) [13] showing the Ag and In isotopes of interest.

a method has been proposed to detect MeV γ -ray signals associated with fission in neutron-rich ejecta from NSM events [14]. As it was shown, fission of actinide species in NSMs can produce a unique signature of photons above 3.5 MeV, which, although initially a minor contributor compared to beta decays, may dominate the gamma-ray signal at later times (around 10 days post-merger) when the ejecta becomes optically thin [15]. However, the flux of γ -rays above 3.5 MeV is low, potentially limiting their detection to nearby events. In this context, the isomeric yield ratios (IYR) from fission fragments could play a crucial role during observations, as the γ -ray intensities following the β -decay of these states often depend on these ratios. If such states are predominantly populated in a fissioning system, the resulting γ -ray signal below 1 MeV could be significantly enhanced, due to the higher intensity of γ -ray lines emitted during the β -decay of isomers. For instance, according to the ENSDF, γ -rays emitted during the β -decay of the ground state of ^{126}In are at 969.61 keV and 1141.11 keV, with intensities of 14.9% and 56.0%, respectively. In contrast, γ -rays emitted during the β -decay of its isomeric state include 111.79, 908.58, 1141.1, 1377.99, and 1636.5 keV, with intensities of 88.0%, 99%, 100%, 23.2%, and 29.6%, respectively. This difference might be detectable in more distant, extra-Galactic NSMs, as γ -ray radiation below 1-MeV has a much higher flux compared to that above 3.5-MeV and could be a complementary or alternative method to the one suggested in [14].

Some isomeric yield ratios of indium isotopes, namely $^{119,121,123,125,127}\text{In}$, were measured in the proton-induced fission of uranium at IGISOL with proton energies at 25 MeV [16]. The excitation energy of the compound nucleus impacts the isomeric yield ratio by influencing both the initial spin distribution of fission fragments and their de-excitation process [17]. Higher excitation energy leads to fragments with higher spin, increasing the likelihood of high spin isomer production. However, this effect is balanced out by de-excitation mechanisms, such as γ -ray and neutron emission. Such investigations of the population of high-spin isomers have not yet been conducted at rare isotope facilities. To correct for the target release time we will measure release curves in the shifts also dedicated for yield measurements.

2 Experimental apparatus and techniques

The proposed measurements will be conducted with the ISOLTRAP mass spectrometer apparatus [18], see Figure 3. The system is composed of four ion traps operated for beam preparation, purification, and mass determination. The quasi-continuous ISOLDE ion beam is accumulated, cooled and bunched in a linear radio-frequency quadrupole cooler and buncher (RFQ-CB) with helium buffer gas [19]. The resulting ion bunches are transported into the multi-reflection time-of-flight (MR-ToF) mass spectrometer. This device is used both for beam purification and precision mass measurements of short-lived nuclei in combination with a ToF detector [20]. The last two devices are two Penning traps used for higher precision measurements, where the first Penning trap is used for further cleaning of the ion bunches, and the second Penning trap is used for measuring with either the time-of-flight ion-cyclotron-resonance (TOF-ICR) [21] or the phase-imaging ion-cyclotron-resonance (PI-ICR) techniques [22]. Mass calibration of the ISOLTRAP setup is conducted with an offline alkali ion source situated upstream of the RFQ-CB. It can provide stable ^{133}Cs and $^{85,87}\text{Rb}$ ions.

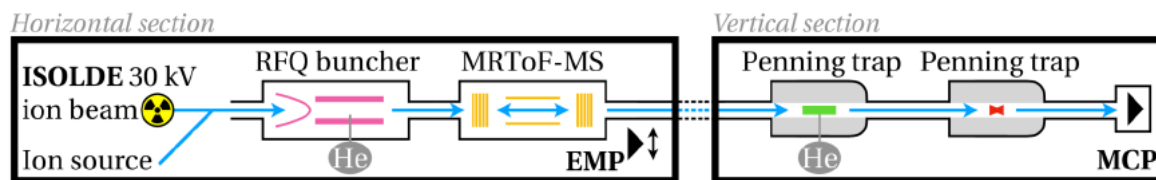


Figure 3: Schematic of the ISOLTRAP setup. The horizontal section encompasses an RFQ-CB and an MR-ToF mass spectrometer. The vertical section consists of two Penning traps.

3 Yields and beamtime planing

We propose to split the measurements into two runs. One for $^{124-135}\text{In}$, and the other one for $^{124-129}\text{Ag}$. An isotopic breakdown of yields, ion properties, and the requested shifts can be found in Table 1 and 2:

Indium : We start the measurements by cross-checking the known masses $^{124-125}\text{In}$ during **1 shift**. Then, we will measure the isomeric yield ratios of indium 126 to 131 with the Penning traps for another **3 shifts**. We also plan to dedicate **2 shifts** for yield checks and release curve measurements, as well as laser on/off comparison spectra for the identification of the most neutron-rich indium isotopes with the MR-ToF mass spectrometer. For high-precision mass measurements and the isomeric yield ratios for more exotic indium isotopes, we ask for **2 shifts** for ^{132}In , **2 shifts** for ^{133}In , and **3 shifts** for ^{134}In . The last isotope, ^{135}In , will be a first direct mass measurement ever, so we ask for **4 shifts**. With an addition of **1 shift** for beam tuning to ISOLTRAP the first run will need **17 shifts of radioactive beam**.

Table 1: Indium isotopes of interest. Half-life, spin, mass, and excitation-energy data from NUBASE 2020 [13]. Except for $^{131}\text{In}^n$ and $^{132-134}\text{In}$ mass uncertainty and excitation energy which are from [8]. Values marked with # are not measured but estimated from trends in neighboring nuclei. The ^{128}In yield as achieved using LIST was communicated by the target group, while the remaining yields (marked with *) are extrapolated from this value taking into account the release curve and FLUKA-simulated in-target production [23, 24].

Nuclide	$T_{1/2}$ (ms)	J^π	yield/ μC	δm (keV)	E_{exc} (keV)	$R = E_{\text{exc}}/mc^2$	shifts
^{124}In	3120	3^+	$8 \times 10^5*$	30			0.5
$^{124}\text{In}^m$	3670	8^-		50	-20(60)	5.8E6	
^{125}In	2360	$9/2^+$	$7 \times 10^5*$	1.8			0.5
$^{125}\text{In}^m$	12200	$1/2^-$		12	352	3.3E5	
^{126}In	1530	3^+	$3 \times 10^5*$	4			0.5
$^{126}\text{In}^m$	1640	8^-		5	90	1.3E6	
^{127}In	1086	$9/2^+$	$2 \times 10^5*$	10			0.5
$^{127}\text{In}^m$	3618	$1/2^-$		15	394	3.0E5	
$^{127}\text{In}^n$	1040	$21/2^-$		40	1770	6.7E4	
^{128}In	816	3^+	1×10^5	1			0.5
$^{128}\text{In}^n$	720	8^-		2	285	4.2E5	
$^{128}\text{In}^p$	>300	16^+		1	1798	6.6E4	
^{129}In	570	$9/2^+$	$6 \times 10^4*$	2			0.5
$^{129}\text{In}^m$	1230	$1/2^-$		2	450	2.7E5	
$^{129}\text{In}^p$	670	$23/2^-$		50	1650	7.3E4	
$^{129}\text{In}^q$	110	$29/2^+$		50	1941	6.2E4	
^{130}In	273	1^-	$2 \times 10^4*$	2			0.5
$^{130}\text{In}^m$	540	10^-		2	67	1.8E6	
$^{130}\text{In}^n$	540	5^+		2	385	3.1E5	
^{131}In	262	$9/2^+$	$7 \times 10^3*$	2			0.5
$^{131}\text{In}^m$	328	$1/2^-$		3	376	3.2E5	
$^{131}\text{In}^n$	322	$21/2^+$		38	3771	3.3E4	
^{132}In	202	7^-	$5 \times 10^3*$	38			2
^{133}In	163	$9/2^+$	500*	41			2
$^{133}\text{In}^m$	167	$1/2^-$		69	642	1.9E5	
^{134}In	136	7^-	40*	44			3
^{135}In	103	$9/2^+$	2*	300#			4

Silver : We will again start with cross-check measurements for $^{124,125}\text{Ag}$ for **2 shifts**, and yield and release checks for **2 more shifts**. This will be followed by mass measurements of ground and isomeric states of the neutron rich silver isotopes $^{126-129}\text{Ag}$ using the PI-ICR technique with the Penning traps. Isomeric yield ratios will be performed in parallel as we need to have both the ground and isomeric states. Here, we will need **2 shifts** for ^{126}Ag and another **2 shifts** for ^{127}Ag . Because of its low yield, and the expected long-lived cesium and barium contaminations, **3 shifts** are required for ^{128}Ag . For the most challenging silver isotope – ^{129}Ag –, **4 shifts** are requested. An additional **1 shift** is requested for beam tuning to ISOLTRAP. In total, the second run will need **15 shifts of radioactive beam**.

Table 2: Silver isotopes of interest. All half-life, spin, mass-uncertainty, and excitation-energy data from NUBASE 2020 [13], except for the $^{124,125}\text{Ag}$ mass uncertainty and ^{124}Ag excitation energy from [9]. Values marked with # are not measured but estimated from trends in neighboring nuclei. The measured ^{129}Ag yield is from [25]. The other yields marked with * are extrapolated yields taken from the INTC proposal P551 [26].

Nuclide	$T_{1/2}$ (ms)	J^π	yield/ μC	δm (keV)	E_{exc} (keV)	$R = E_{\text{exc}}/mc^2$	shifts
^{124}Ag	178	2^-	$1.4 \times 10^5*$	7			1
$^{124}\text{Ag}^m$	144	9^-		7	188	6.1E5	
^{125}Ag	160	$9/2^+$	$1.0 \times 10^5*$	4			1
$^{125}\text{Ag}^m$	50#	$1/2^-$		8	97	1.2E6	
^{126}Ag	52	$3^+\#$	640*	200#			2
$^{126}\text{Ag}^m$	108	$9^-\#$		220#	100#	1.2E6	
^{127}Ag	89	$9/2^+$	1400*	200#			2
$^{127}\text{Ag}^m$	20#	$1/2^-$		200#	20#	5.9E6	
$^{127}\text{Ag}^n$	68	$27/2^+$		200#	1938	6.1E4	
^{128}Ag	60	$3^+\#$	62*	300#			3
^{129}Ag	50	$9/2^+\#$	1	400#			4
$^{129}\text{Ag}^m$	10#	$1/2^-\#$		400#	20#	6.0E6	

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4 Details for the Technical Advisory Committee

4.1 General information

The experimental setup comprises:

	Availability	Design and manufacturing
ISOLTRAP	<input checked="" type="checkbox"/> Existing	<input checked="" type="checkbox"/> To be used without any modification

4.2 Beam production

Isotopes near stability and neutron-rich isotopes are best produced with an UCx target. [Table 1](#) and [2](#) summarize the experimental and extrapolated yields.

- Requested beams:
 - $^{124-135}\text{In}$
 - $^{124-129}\text{Ag}$
- Target - UCx: *New target for each run is requested to avoid complications due to target aging.*
- RILIS? *Yes*
 - Special requirements: *LIST for indium*
- Additional features?
 - Neutron converter? *Yes*
 - Other:
- Expected contaminants: *barium and cesium isobars, especially around stable ^{133}Cs*
- Acceptable level of contaminants: *For the indium measurements LIST will suppress the Cs and Ba contamination. For silver the relative contamination yield will be reduced by using the neutron converter. The mass resolving power of ISOLTRAP's MR-ToF MS is sufficient to separate the remaining isobars and enable the proposed measurements.*
- Can the experiment accept molecular beams? *No*
- Are there any potential synergies (same element/isotope) with other proposals and LOIs that you are aware of? *Yes, IS732 from CRIS. They are also planning to measure neutron rich indium ($^{131-134}\text{In}$) in 2025. But they wont use LIST as they require higher yield than ISOLTRAP.*

4.3 Shift breakdown

We propose to split the measurements into two runs. Planning of the experiment is as follows:

Summary of requested shifts for indium run:

With protons	Requested shifts
Yield and release measurement of exotic indium isotopes	2
Cross-check using $^{124,125}\text{In}$	1
Data taking, $^{126-131}\text{In}$	3
Data taking, ^{132}In	2
Data taking, ^{133}In	2
Data taking, ^{134}In	3
Data taking, ^{135}In	4
Total with radioactive beam	17

Summary of requested shifts for silver run:

With protons	Requested shifts
Yield and release measurement of exotic silver isotopes	2
Cross-check using $^{124,125}\text{Ag}$	2
Data taking, ^{126}Ag	2
Data taking, ^{127}Ag	2
Data taking, ^{128}Ag	3
Data taking, ^{129}Ag	4
Total with radioactive beam	15

HAZARDS GENERATED BY THE EXPERIMENT

Hazards named in the document relevant for the fixed ISOLTRAP installation.