

EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Precision Mass Measurements with ISOLTRAP to Study the  
Evolution of the  $N = 82$  Shell Gap far from Stability

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**Abstract:** Shell effects and their evolution across the nuclear chart pose important constraints on the modelling of the nucleon-nucleon interaction. The strength of shell closures in neutron-rich nuclei also influences the path of the  $r$ -process of nucleosynthesis and the predicted elemental abundances. We propose to measure the masses of the isotopes  $^{132,133}\text{In}$ ,  $^{129-132}\text{Cd}$ ,  $^{125-129}\text{Ag}$  with the Penning-trap mass spectrometer ISOLTRAP. The recently developed multi-reflection time-of-flight mass separator of ISOLTRAP will allow, as a beam purifier, to handle higher contamination ratios as before and, for the more exotic cases, to directly determine the mass of the nuclides of interest. The masses of the proposed isotopes will allow the investigation of a possible weakening of the  $N = 82$  shell gap for  $Z < 50$  and corresponding  $r$ -process waiting point. This in turn enables an exploration of the impact on the  $A = 130$   $r$ -process abundances.

**Requested shifts:** 50 shifts (split in 3 runs over 2 years)



# 1 Introduction

Through precision mass measurements, Penning-trap experiments allow the investigation of various physics topics. The goals of this proposal are nuclear-structure studies and questions related to nuclear astrophysics. From masses one can calculate differential quantities (mass filters), such as separation energies and empirical energy gaps, which are sensitive to different nuclear-structure effects. For example, crossing a magic proton or neutron number produces a sharp drop in the corresponding one-particle or two-particle separation energy, an effect well established as a signature for magicity. The evolution of the properties of magic nuclei towards the proton and neutron drip lines is crucial for the understanding of nuclear structure far from stability. The mass of  $^{54}\text{Ca}$ , for example, led to the firm establishment of a new magic number at  $N = 32$  and provided a crucial test of modern nuclear theories [1]. Similarly, the strength of shell effects in very heavy systems was investigated through mass measurements of nobelium and lawrencium isotopes with SHIPTRAP (Darmstadt, Germany) revealing an enhancement of the two-neutron shell gap at  $N = 152$  [2].

The synthesis of over half of the heavy elements has its origin in the rapid neutron-capture process ( $r$ -process) [3]. For the modeling of the  $r$ -process, reliable masses of exotic nuclei are an indispensable nuclear physics input [4]. Particularly important in a classical  $r$ -process model are nuclei in the vicinity of so-called waiting points. The indium, cadmium and silver isotopes targeted in the proposal lie close to the  $N = 82$  waiting point and are affecting the  $A = 130$  abundance peak of  $r$ -process nuclei [5, 6]. How much the abundances build up depends on the strength of neutron shell closures in these exotic nuclei.

Masses of exotic nuclides are employed to constrain nuclear models, which in turn provide experimentally inaccessible masses and the equation of state of nuclear matter. As such nuclear theory is used to model nucleosynthesis processes and the structure of astrophysical objects such as neutron stars [7]. The Penning-trap mass spectrometer ISOLTRAP has recently achieved a breakthrough measuring the mass of  $^{82}\text{Zn}$ , which allowed constraining neutron-star crust composition to deeper layers [8]. Furthermore, a dominant role of the  $N = 50$  and  $N = 82$  closed neutron shells for the crustal composition was confirmed [9].

The implementation of the multi-reflection time-of-flight mass separator/spectrometer (MR-TOF MS) has enabled mass measurements of nuclei more exotic than previously possible. Used as a separator for beam purification, a resolving power on the order of  $10^5$  together with a suppression of the unwanted species of close to  $10^4$  can be reached within a few tens of milliseconds. This is about an order of magnitude faster than the conventional technique employed so far with any Penning-trap mass spectrometer worldwide. This achievement opens the door for the study of nuclei even farther from stability with shorter half-lives and lower production yields or beams with a higher degree of contamination.

The main aim of this proposal is to investigate the evolution of the neutron magic number  $N = 82$  for highly asymmetric systems (i. e. nuclides with a much larger number of neutrons than protons) for which we propose mass measurements of  $^{132,133}\text{In}$ ,  $^{129-132}\text{Cd}$ , and  $^{125-129}\text{Ag}$ . In the case of Cd, we aim at clarifying the long standing question whether  $^{130}\text{Cd}$  is a strong  $r$ -process waiting point. It is interesting to recall that  $^{129}\text{Ag}$  was the object of the  $r$ -process-motivated IS 333 experiment that also initiated the RILIS technique [10, 11]. The success of RILIS was a major step forward in providing beams farther from stability.



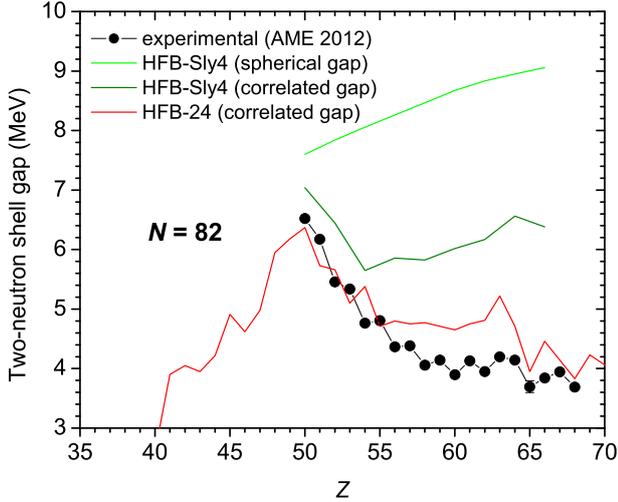


Figure 2: Two-neutron shell gap at  $N = 82$  as a function of proton number. Experimental values [12] as well as values from Hartree-Fock-Bogoliubov mass models are shown [14, 15].

delivered with a large contamination from Cs isotopes, as are the proposed Cd and Ag isotopes. State-of-the-art purification techniques at ISOLDE as well as ISOLTRAP are expected to cope well with the large contamination ratios. For further details, see section 4.

**Neutron-rich Cd isotopes:** Conflicting conclusions on the shell strength at  $N = 82$ ,  $Z = 48$  were drawn from mass measurements through the determination of the  $\beta$ -endpoint energy [6] and  $\gamma$ -ray spectroscopy [16]. Taken into account that systematic deviations were observed in  $\beta$ -endpoint measurements [17], an indispensable information will be obtained by determining the two-neutron separation energy via masses of  $^{130}\text{Cd}$  and  $^{132}\text{Cd}$  isotopes. Furthermore,  $^{130}\text{Cd}$  is considered to be an  $r$ -process waiting point, which is of special importance since the strength of the  $N = 82$  shell gap (established by an ISOLTRAP mass measurement of  $^{134}\text{Sn}$  [18]) has been shown to have a strong influence on the number of neutrons available for fission – and subsequent re-cycling of the  $r$ -process [19]. This question is particularly interesting since the hypothesis of a quenched  $N = 82$  shell has long been cited as a possible cure for the heavy-element-abundance deficits of models [20]. Neutron-separation energy in  $^{131}\text{Cd}$ , given by the difference of the masses of  $^{130}\text{Cd}$  and  $^{131}\text{Cd}$  isotopes, is the key nuclear physics input in the corresponding calculations. Already an accurate mass measurement of only the  $^{130}\text{Cd}$  nuclide will allow to draw conclusions although in a model-dependent way. The mass values of  $^{130-132}\text{Cd}$  will further allow conclusions on the role of the nuclide  $^{133}\text{Cd}$ . The present mass value of  $^{130}\text{Cd}$  exhibits an uncertainty of over 100 keV, while  $^{131,132}\text{Cd}$  are only extrapolated. In addition, laser spectroscopy on neutron-rich Cd isotopes has been conducted [21]. A new isomer of  $^{129}\text{Cd}$  was identified but its excitation energy remains unknown. Its characterization will be done with ISOLTRAP.

**Neutron-rich Ag isotopes:** Neutron-rich silver isotopes have been measured with ISOLTRAP [22] reaching  $^{124}\text{Ag}$  at  $N = 78$ . The masses beyond are experimentally unknown. We intend to push the known masses for the isotopic chain of silver to the  $N = 82$  shell gap. Together with the information on the shell gap for  $Z = 49, 48$ , it becomes possible to draw conclusions of the persistence or possible quenching of the  $N = 82$  shell towards more exotic species. Regarding the crustal composition of neutron stars, nuclides at higher densities than probed with the mass of  $^{82}\text{Zn}$  are believed to cluster around the magic neutron number  $N = 82$  [9]. As the corresponding calculations are based on mass models which infer the existence of magic numbers, a possible quench of the  $N = 82$  shell might lead to a changed picture of the

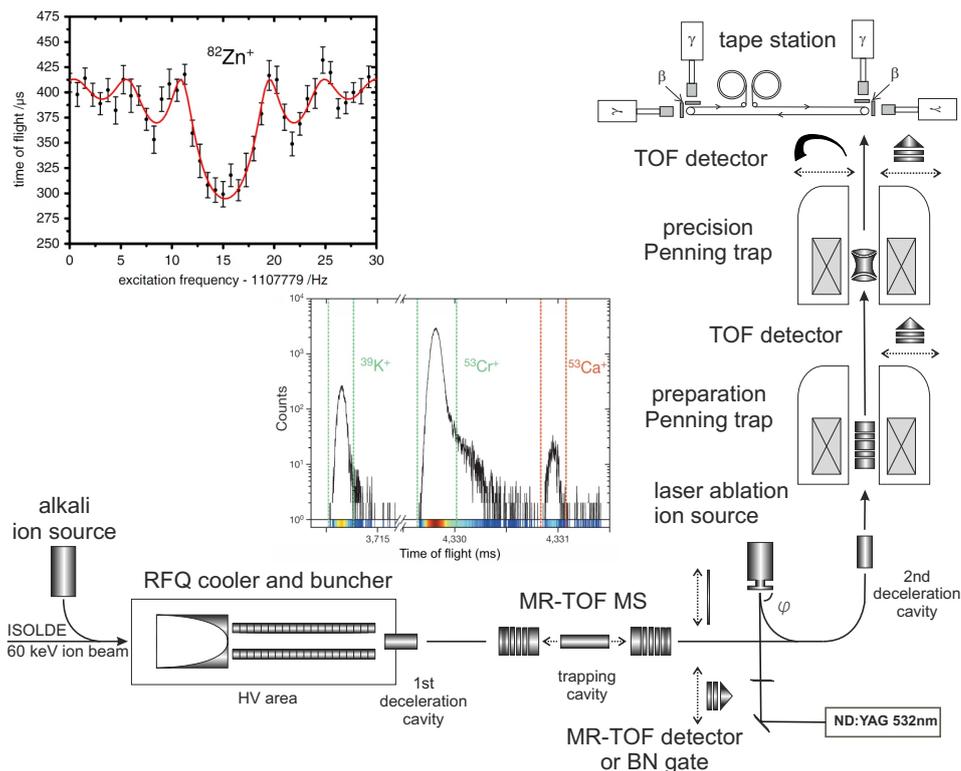


Figure 3: Schematic view of the ISOLTRAP setup. The inset on the left shows a time-of-flight resonance of  $^{82}\text{Zn}^+$  (TOF detector above precision Penning trap), the inset on the right shows a time-of-flight spectrum of  $A = 53$  (MR-TOF detector). For details, see text.

neutron-star crust composition. Silver isotopes are of added importance for unveiling the nature of a second type of  $r$  process [23].

### 3 Mass Measurements with ISOLTRAP

The Penning-trap mass spectrometer ISOLTRAP has been performing mass measurements routinely reaching relative uncertainties of  $10^{-8}$  for many years [24] giving input to various physical topics reaching from nuclear structure studies to astrophysical applications and fundamental tests, which has been documented in many publications [25]. Fig. 3 shows the current experimental setup, which consists of four traps: a linear segmented radio-frequency quadrupole (RFQ) for ion beam cooling and bunching, a multi-reflection time-of-flight mass separator (MR-TOF MS), and two Penning traps (preparation and precision Penning trap) [26].

The ISOLDE ion beam first enters the linear, gas-filled RFQ ion trap where buffer-gas cooling is applied for 10 – 20 ms. After passing the first deceleration cavity, isobaric contaminants are separated due to mass-over-charge dependent flight times after repeated oscillations between the electrostatic mirrors of the MR-TOF MS. The MR-TOF MS can thus act as an auxiliary device for isobaric purification reaching resolving powers on the order of  $R = m/\Delta m \approx 10^5$  within a few ten milliseconds [27]. An example of such a time-of-flight spectrum of  $A = 53$  is shown in the right inset of Fig. 3 for roughly 4 ms flight time. A Bradbury-Nielsen gate (BN gate) installed

directly behind the MR-TOF mass separator reaches a suppression factor for contaminating ions of four orders of magnitude, thus considerably enhancing the performance of ISOLTRAP when dealing with large contamination ratios. Alternatively, the MR-TOF MS can be used for mass measurements reaching uncertainties on a low  $10^{-7}$  level [1].

Furthermore, by gating on the time-of-flight of the ion of interest in the isobaric spectrum (see right inset of Fig. 3), one can scan the wavelength of one of the excitation steps of the RILIS lasers and thus perform in-source laser spectroscopy either for nuclear-structure studies or for selective ionization of different nuclear states. Proof of principle measurements with the MR-TOF MS, showing the capability of isomeric selectivity by exploiting the hyperfine structure, have been performed in 2012 and recently reported in [26, 28].

In the subsequent preparation Penning trap ions can be removed by mass-selective resonant buffer-gas centering with a resolving power of up to 100,000. The selected ions are transferred to the second, precision Penning trap for the mass measurement. The time-of-flight ion-cyclotron resonance technique is employed to determine the cyclotron frequency of an ion stored in a Penning trap (see left inset of Fig. 3), from which the mass can be extracted in conjunction with a reference mass measurement [29]. The overall efficiency from the merging switchyard of ISOLDE to the detector is on the order of  $10^{-3}$  to  $10^{-2}$ , depending on the half-life of the nuclide of interest and possible decay losses.

## 4 Experimental Program

The beam-time request is detailed in the Tab. 1. The feasibility of producing the desired radioactive beams is as follows.

**Neutron-rich In isotopes:** Large contamination is expected from surface-ionized cesium. Tests with the new neutron-converter geometry are planned for early next year from which an enhancement of the contamination ratio between 10 and 100 is expected [30]. In the case of  $^{132}\text{In}$  a contamination ratio around 1 is expected. In the case of  $^{133}\text{In}$ , the stable (not radiogenic) Cs contamination is expected to lie four orders of magnitude higher. This contamination ratio coincides with ISOLTRAP's best performance using the MR-TOF MS in combination with the BN gate. We thus ask for one extra shift for purification of the beam.

**Neutron-rich Cd isotopes:** Test measurements involving a neutron converter and a cold quartz line ( $300^\circ\text{C}$ ) yielded a value of  $1.3 \cdot 10^4 \text{ ions}/\mu\text{C}$  for  $^{128}\text{Cd}$  [31] and two orders of magnitude less for the only contamination in the beam,  $^{128}\text{In}$ . Comparing this yield to that of protons on target [32], one observes a factor 3-10 decrease. Based on this information, the values for  $^{129-132}\text{Cd}$  have been extrapolated. In the meantime the RILIS lasers have been upgraded such that a slight enhancement of the yield is expected. The contamination arising from  $^{130}\text{Cs}$  was measured to  $9 \cdot 10^5 \text{ ions}/\mu\text{C}$ , which can be purified by the ISOLTRAP setup. The neutron converter with a newly developed geometry should further decrease the Cs contamination. Nevertheless, on mass 132 the contamination of In and Cs might lie three or four orders of magnitude above the Cd yield, which is why we ask for an extra shift.

**Neutron-rich Ag isotopes:** Based on measurements in 2007, the yield of  $^{122}\text{Ag}$  is given as  $9 \cdot 10^6/\mu\text{C}$ , the yield of  $^{123}\text{Ag}$  is given as  $5 \cdot 10^6/\mu\text{C}$  using the laser ion source [30]. An enhancement of these yields through further developments of the RILIS lasers is expected but could not be demonstrated yet. We expect contaminating isotopes of Cs and In as had been already

seen before with ISOLTRAP [22]. A neutron converter with the newly developed geometry should be used to decrease the Cs yield. Purification of the beam can be achieved with the ISOLTRAP setup using the MR-TOF MS in combination with the preparation Penning trap. However, as numbers from PSB for the respective less-exotic In yield are not available we ask for two additional shifts for beam purification. The ratio between the ground and isomeric state in  $^{125-127,129}\text{Ag}$  can be adjusted, due to the shorter half-life of the isomer, by in-trap decay. Isomerically selective ionization can also be attempted with RILIS, using the MR-TOF MS as a diagnostics tool. As the excitation energies of the isomers in Ag are mostly unknown, we do not ask for separate shifts to determine the excitation energies at this point.

**Isomerically pure beams:** For many of the proposed nuclei, isomers have been seen or are predicted to exist. We intend to use the MR-TOF MS for in-source laser spectroscopy tests to provide isomerically pure beams for the mass measurements with ISOLTRAP [26]. One shift each is required for  $^{129}\text{Cd}$ ,  $^{125-127,129}\text{Ag}$ , and  $^{133}\text{In}$ .

Table 1: Isotopes for which shifts are requested are shown with half-lives and mass uncertainties taken from [12], extrapolated values are marked with #. Expected yields were taken from the ISOLDE yield database or communicated by the ISOLDE Target Group, extrapolations are marked with \$. Isomeric states of interest are indicated by *m*. Moreover, the measurement mode of ISOLTRAP as well as the the target-ion-source combination are listed.

Nuclide	Half-life	$\delta m$ keV	yield ion/ $\mu\text{C}$	PTMS or MR-TOF-MS	shifts	target	ion source
$^{132}\text{In}$	207.0 ms	60	$1 \cdot 10^4$	PTMS	3+1	UCx	Ta cavity + n. c. + RILIS
$^{133}\text{In}$	165.0 ms	200#	$1 \cdot 10^3$	PTMS	3	UCx	Ta cavity + n. c. + RILIS
$^{129}\text{Cd}$	242.0 ms	200#	$1.0 \cdot 10^4$ \$	PTMS	3	UCx	Ta cavity + n. c. + RILIS
$^{129m}\text{Cd}$	104.0 ms	200#	$1.0 \cdot 10^4$ \$	PTMS	3	UCx	Ta cavity + n. c. + RILIS
$^{130}\text{Cd}$	162.0 ms	160	$1.0 \cdot 10^3$ \$	PTMS	4	UCx	Ta cavity + n. c. + RILIS
$^{131}\text{Cd}$	68.0 ms	200#	$1.0 \cdot 10^2$ \$	MR-TOF MS	4	UCx	Ta cavity + n. c. + RILIS
$^{132}\text{Cd}$	97.0 ms	200#	$5.0 \cdot 10^0$ \$	MR-TOF MS	4+1	UCx	Ta cavity + n. c. + RILIS
$^{125}\text{Ag}$	166.0 ms	600	$4.0 \cdot 10^5$ \$	PTMS	2+1	UCx	Ta cavity + n. c. + RILIS
$^{126}\text{Ag}$	55.0 ms	200#	$2.0 \cdot 10^4$ \$	PTMS	3	UCx	Ta cavity + n. c. + RILIS
$^{127}\text{Ag}$	79.0 ms	200#	$3.0 \cdot 10^3$ \$	PTMS	3	UCx	Ta cavity + n. c. + RILIS
$^{128}\text{Ag}$	58.0 ms	300#	$5.0 \cdot 10^2$ \$	PTMS	4	UCx	Ta cavity + n. c. + RILIS
$^{129}\text{Ag}$	44.0 ms	300#	$8.0 \cdot 10^1$ \$	MR-TOF MS	4+1	UCx	Ta cavity + n. c. + RILIS

**Summary of requested shifts:** We request 50 shifts, 40 for the actual mass measurements and 4 for beam purification as can be seen from Table 1 as well as 6 shifts for the provision of isomerically pure beams. Shifts for target-and-ion-source development are not included.

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# Appendix

## DESCRIPTION OF THE PROPOSED EXPERIMENT

The experimental setup comprises: ISOLDE central beam line and ISOLTRAP setup. The ISOLTRAP setup has safety clearance, the memorandum document 1242456 ver.1 “Safety clearance for the operation of the ISOLTRAP experiment” by HSE Unit is released and can be found via the following link: <https://edms.cern.ch/document/1242456/1>.

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