EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Precise mass measurements of light and heavy neutron-rich noble-gas isotopes for nuclear structure studies

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Abstract:

We propose to measure the masses of the neutron-rich noble-gas isotopes ^{49,50}Ar and ^{230,231}Rn using the online mass spectrometer ISOLTRAP. The extracted binding-energy trends will be used to study the evolution of nuclear structure in two regions of the nuclear chart towards higher neutron-proton asymmetry. The mass determination of ^{49,50}Ar will allow for the first time to experimentally determine the neutron separation energies up to the proposed N=32 sub-shell closure in argon. Furthermore, the extracted binding energies of argon can be used to probe the average proton-neutron interaction in the calcium chain at the N=32 sub-shell closure. The measurements of ^{230,231}Rn will allow expanding the emerging trend of the two-neutron separation energies above the closed-shell lead isotopes, which exhibit contrasting behaviour, in particular more binding at N=143 compared to N=142.

Requested shifts: 20 shifts

1 Motivation

Mass measurements of radioactive nuclei are needed to determine their binding energies, reflecting the interactions between all their constituents and being sensitive to nuclear-structure features, such as shell effects, collectivity and deformation. Thus, the direct determination of the nuclear binding energy $B(Z,N)$ provides a model-independent study of nuclear structure and shell evolution.
Of the so-called mass filters, the two-neutron separation energy $S_{2n}(Z,N) = B(Z,N) - B(Z,N-2)$ is an important parameter for understanding nuclear stability and shell structure, particularly in neutron-rich isotopes, and an essential input for shell model and mean-field theories.
Furthermore, the double difference of binding energies of isotopes of surrounding elements gives insight in the proton-neutron interaction, described as the δV_{pn} value [1]. It represents the average interaction between the last two protons and the last two neutrons in a nucleus. This parameter is particularly important when discussing the configuration mixing, the onset of collectivity and deformation in nuclei [2]. Enhanced δV_{pn} values often indicate increased collectivity and shape changes in the nucleus.
The δV_{pn}^{ee} value for even-Z and even-N nuclei and δV_{pn}^{oe} for odd-even nuclei are determined by:

$$\delta V_{pn}^{ee}(Z,N) = 1/4\left[(B_{Z,N} - B_{Z,N-2}) - (B_{Z-2,N} - B_{Z-2,N-2})\right], \tag{1}$$

$$\delta V_{pn}^{oe}(Z,N) = 1/2\left[(B_{Z,N} - B_{Z,N-2}) - (B_{Z-1,N} - B_{Z-1,N-2})\right]. \tag{2}$$

Complementarily, an indication for a strong degree of collectivity and deformation can additionally be provided by parameters from γ-ray spectroscopy measurements, like the first excited state $E(2_1^+)$ for even-even nuclei, or its transition probability $B(E2; 2_1^+ \rightarrow 0_1^+)$, referenced as $B(E2)$ in the following. Together with precise mass values a comprehensive picture of the nuclear structure can be formed.
This makes masses an attractive property for studying nuclear structure evolution, challenge model predictions as well as refining predictions of experimentally unknown masses which are necessary for the simulation of nucleosynthesis processes [3]. The purpose of this proposal is to extend the mass values for neutron-rich noble-gas isotopes, whose physical motivation is outlined in more detail in the following.

1.1 Motivation for neutron-rich Ar

ISOLTRAP's precision mass measurements of $^{51-54}$Ca in 2013 established a shell closure at the neutron number N=32 [4], indicated beforehand by a high 2^+ energy $E(2_1^+)$ in ^{52}Ca from γ-ray spectroscopy [5]. This sub-shell effect for Ca can be seen with a drop of S_{2n} from N=32 to 34 in Figure 1(left). As displayed in Figure 1(middle), the experimental $E(2_1^+)$ of ^{48}Ca is very high due to a shell gap at N=28, as expected. After a sudden decrease at N=30, a sharp increase at N=32 is observed. In addition, the transition probability $B(E2)$ for ^{48}Ca [6, 7] confirms the shell closure at N=28, while there is no experimental $B(E2)$ for ^{52}Ca at N=32. Regarding charge radii, the ^{52}Ca charge radius was unexpectedly large in contrast to the expected decrease at N=32 [8].
Looking at spectroscopic results for argon, a 1178(18) keV level is reported and assigned as $E(2_1^+)$ for ^{50}Ar (N=32) [9]. Not only N=32 $E(2_1^+)$ energies but also N=34 are known experimentally for both Ar and Ca. With the $E(2_1^+)$ of ^{52}Ar (N=34) measured for the first time by H.N. Liu *et al.* [10] in 2019, N=34 is stated as a new sub-shell closure confirming the original proposition made for ^{54}Ca [11]. In addition to that, the $E(2_1^+)$ energies of ^{56}Ca and ^{58}Ca are tentatively assigned [12], which reinforced this view. Complementarily, a low $B(E2)$ value was measured for ^{46}Ar (N=28) [6, 13], as expected at the magic number, while slightly increasing for ^{48}Ar [14]. This is also confirmed by an in-beam γ-ray spectroscopy study with high $E(2_1^+)$ for ^{46}Ar (N=28) [6] and ^{52}Ar (N=34) [10], while being relatively

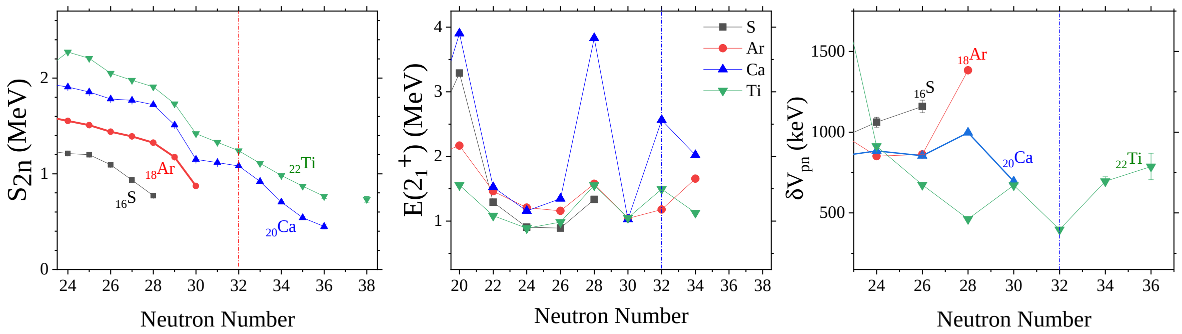

Figure 1: Different observables reflecting nuclear structure are presented for the nuclei of S, Ar, Ca and Ti around the proposed sub-shell closure N=32, indicated with a dashed line in each panel.
(Left): Two-neutron separation energy as a function of neutron number.
(Middle): Similar as (Left) but for the electric quadrupole transition energy from the first 2_1^+ energy to the ground state.
(Right): Average proton-neutron interaction, δV_{pn}, as a function of neutron number.

low at N=32, ^{50}Ar [9, 15] (see Figure 1(middle)). Similar measurement results are given for $Z > 20$ nuclei in the N=28, 32 region with higher $E(2_1^+)$ and lower $B(E2)$ at N=32 for ^{54}Ti and ^{56}Cr [16–18]. One remarks the interesting feature showing a larger $E(2_1^+)$ at N=34 than N=32 for Ar (two protons below Z=20), while the opposite is observed for Ti (two protons above). This pattern hints at significant structural changes going from above to below the calcium isotopic chain.
Regarding mass studies for nuclei below the magic proton number Z=20, the N=32 sub-shell closure has been probed at ISOLTRAP in 2015 for $_{19}$K [19] in addition to the N=32 sub-shell closure of Ca with Z=20. In contrast, $_{18}$Ar is less studied in the region of N=32 more likely due to more neutron-to-proton asymmetry. Here, ISOLTRAP has measured the masses of Ar isotopes up to ^{48}Ar (N=30) probing the N=28 shell closure [20], leaving masses for Ar with $N > 28$ only with extrapolations from systematic trends in the AME2020 [21].
Considering all the spectroscopic results at N=32 and even N=34 for Ar discussed above, extending the chain of S_{2n} by measuring Ar masses up to N=32 will better constrain the shell evolution. Furthermore, the mass measurements of ^{49,50}Ar will be used to determine the average proton-neutron interaction strength reflected by the δV_{pn} value, in particular the $\delta V_{pn}^{ee}(^{52}\mathrm{Ca})$ as seen in Figure 1(right). These will provide insights of shell effects for Ca at N=32 and will contribute to a comprehensive understanding of nuclear structure and behavior together with the known $E(2_1^+)$ values. In addition, the $\delta V_{pn}^{oe}(^{51}\mathrm{K})$ can be deduced with the precise mass measurement of ^{50}Ar. This additionally enables to characterize the shell effects in an odd-Z nuclei below the magic proton number Z=20 at N=32.

1.2 Motivation for neutron-rich Rn

With the discovery of ^{229}Rn at ISOLTRAP in 2009 [22], we would like to take the opportunity using the same target and already optimized technical requirements to continue the survey to detect ^{230,231}Rn and determine the nuclear binding energy for the first time. As shown in Figure 2, the last experimentally derived S_{2n} at N=143 is 20 keV higher compared to N=142, which points towards a sudden change in nuclear structure, reflected in a stronger binding of the last two neutrons. The radon isotopes appear to be a potential boundary between regions of spherical and deformed nuclei. Thus, measuring the Rn isotopes with N=144, 145 will extend the S_{2n} trend and give further insights in shell evolution and its opposite inflection compared to heavier chains.

A further important application of the investigation of masses in this region is the development and refinement of microscopic mass models for nucleosynthesis [23–25], since r-process nuclei in this heavy mass region are currently out of reach to be directly measured. Outside the range of the measured masses, the predictions of different mass models diverge rapidly, especially for increasing neutron excess [26]. Therefore, constraining mass models, which can deviate in the order of several MeV in this region on total binding energies, is of utmost importance. Here, the proposed masses help to adjust nuclear mass models needed for nuclear astrophysical applications, for which a mass precision on the order of 50 keV is needed [3, 27].

2 Experimental apparatus and techniques

Currently, the high-precision mass spectrometer ISOLTRAP [28] consists of four ion traps realizing the capture, accumulation and cooling of the exotic beams provided by ISOLDE in the radiofrequency-quadrupol cooler-buncher (RFQ-CB) and performing cleaning and mass measurements using the multi-reflection time-of-flight mass spectrometer (MR-ToF MS) and a tandem Penning-trap system. An overview of the ISOLTRAP setup is given in Figure 3.

The semi-continuous ISOLDE ion beam with a kinetic energy of 30 to 60 keV is accumulated, bunched and cooled in the linear RFQ-CB with helium buffer gas [29]. Cooled ion bunches are ejected and pulsed down to 3.2 keV before being injected in the MR-ToF MS [30]. The latter is used to mass-dependently separate the isobars in the ToF-domain, due to all ions having the same kinetic energy, with a mass

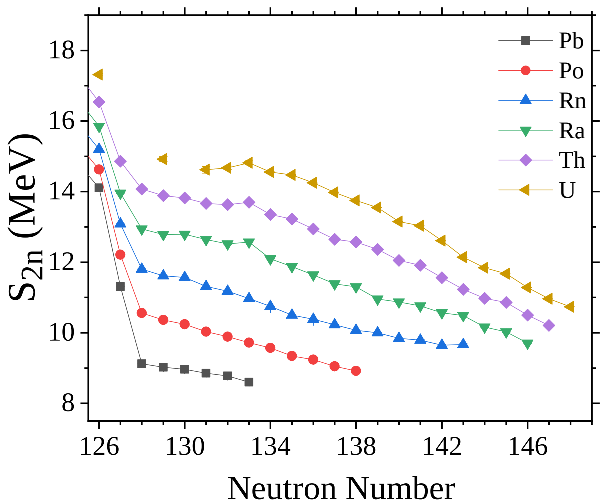

Figure 2: Two-neutron separation energies for Pb, Po, Rn, Ra, Th and U against neutron number.

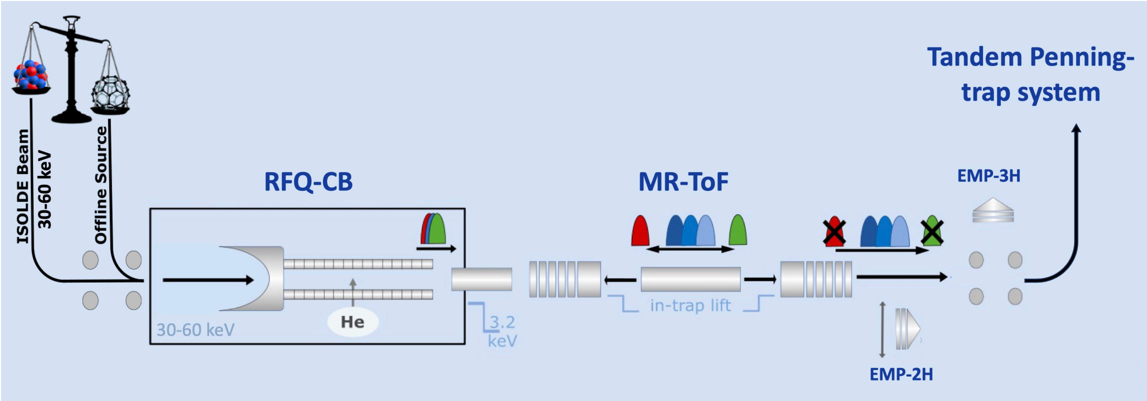

Figure 3: Schematic overview of the horizontal beamline section of the high-precision online mass spectrometer ISOLTRAP, including its offline ion source, its two ion traps and its electron multiplier detectors (EMPs) to perform time-of-flight measurements. For further information see text.

resolving power routinely reaching $R \approx 3 \cdot 10^5$ with the implementation of a mirror-voltage stabilization [31]. The ToF-separated isobaric bunches can be mass-selectively ejected by fast pulsing the in-trap lift of the MR-ToF MS [32] and send to the tandem Penning-trap system for very high-precision mass measurements performed with the PI-ICR technique [33].
However, for cases with low production rates or very short-lived exotic nuclei, precision mass measurements can be directly carried out with the MR-ToF MS, reaching precisions in the order of around 50 keV, sufficient for the above outlined physics motivation. Here, an electron multiplier detector behind the MR-ToF MS allows the measurement of a ToF-spectrum of the separated isobars.
The measured ToF t_{ToF} is related to the ion's mass m and charge q by $t_{\text{ToF}} = a \cdot \sqrt{m/q} + b$, where the calibration parameters a and b are determined by the ToF of reference masses with precisely known mass [4].

The proposed exotic nuclei to be measured are characterized by a large first ionization potential being noble gases, thus susceptible to perform charge-exchange reactions with neutral impurities contained in the helium buffer gas [34]. Therefore, as already demonstrated in the past neutron-rich argon mass measurements, cooling-time optimizations will be performed with the stable isotopes to maximize the charge-exchange half-life inside the RFQ-CB. Furthermore, the He injection gas line will be fed through a bath of liquid nitrogen to freeze out the impurities, which improved the charge-exchange half-life from $23(2)$ ms up to $50(13)$ ms in the experiment in 2017 [20].
The charge exchange of the noble gases in the RFQ-CB is used to its advantage for direct identification of the isotope of interest by enlarging the cooling time in addition to the identification without protons on the target.
As the charge-exchange half-life reveals to be the stronger limitation compared to the nuclear decay half-life (see Table 1) and release [35] for all proposed isotopes to be studied, not only the cooling-time setting has to be optimized, but in addition the accumulation duration in the RFQ-CB and therewith the measurement cycle itself.
As recently successfully investigated at ISOLTRAP with ions from the offline source, the MR-ToF mass measurement can be run parallel to the accumulation and cooling in the RFQ-CB. This allows to probe as much as possible from the release of the target when executing the measurement cycle multiple times per proton-impact and triggering on the latter.

For $^{49,50}Ar$ the stable molecular isobaric contamination $^{33,34}S^{16}O$ is expected to gain a factor 4 and 10, respectively, in in-target production compared to the $^{32}S^{16}O$ observed and identified in the past measurement (see Figure 6 in [20]). Therefore, even shorter accumulation times have to be applied to ensure a low count-rate per experimental cycle. This avoids systematic shifts of the ToF induced by space-charge effects [36]. Taking the expected contamination, as well as charge-exchange half-life and experimental techniques into account leads to the estimated shift numbers given in Table 1 to complete the proposed experiment.

All detailed experimental techniques and optimizations illustrated for the neutron-rich argon measurement apply to the measurement of other proposed noble-gas isotopes in the same manner: Since the first ionization energy of argon is higher compared to higher Z noble gases, the optimized cooling time for the Ar measurement should be applicable to the measurement of $^{230,231}Rn$ as the charge-exchange half-life increases. Still, the charge-exchange half-life remains the limiting factor compared to the half-life of the proposed isotopes. Furthermore, the beam supplied from the separator for Rn are reported as free from isobaric contamination [22, 35, 37].

3 Yield extrapolation

For the mass measurements of the proposed noble-gas isotopes we request an UC_x target equipped with a low-temperature, water-cooled transfer line to a FEBIAD-type plasma ion source. Conditioning the transfer line at low temperatures allows only species that are gaseous at room temperature to be ionized, reducing isobaric contamination [35].
The measured yields for singly-charged noble-gas isotopes from the ISOLDE yield database [38] are listed in Table 1. Only the yields for Rn are extrapolated, for which the release parameters stated in [38] have been used. Since no half-life for $^{230,231}Rn$ is given, upper and lower limit calculations have been performed. Furthermore, the linear trend in the logarithmic yield was continued for a cross-check.
Shift-estimation calculations including the stated yield and half-life in Table 1, the release parameters from [35, 38] and determined charge-exchange half-life [20] have been performed and added to Table 1. Here, the timings for the outlined experimental techniques have been optimized towards the identified limitation. Especially for Ar, this is the stable isobaric contamination SO [20], whose in-target-production yield was scaled according to given values in FLUKA [38, 39] and ABRABLA [38].

4 Shift request

The proposed beam-time request is based on the yields stated in the ISOLDE yield database [38] as well as extrapolations (described in section 3) and the experience gained in past experiments with the ISOLTRAP mass spectrometer for these elements and especially appearing isobaric contamination.
We request **18 shifts for the precision mass measurements of neutron-rich noble-gas isotopes**. In more detail, 15 shifts are allocated for the crucial mass determination of ^{49,50}Ar to study the proposed N=32 sub-shell closure in argon and give insights in the average proton-neutron interaction of calcium and potassium isotopes at the verified N=32 sub-shell closure. As the requested technical requirements (see section 2) are already set up and optimized, these conditions allow the further study of the nuclear structure evolution in ^{230,231}Rn in the same run, for which we request additional 3 shifts. This allows the collection for a minimum of 300 counts of each proposed isotope together with clear identification by comparison ToF spectra without protons or enlarged cooling-time (higher charge-exchange probability).
Additionally, **1 shift** without protons is requested with stable argon isotopes from the plasma ion-source to optimize the target and line conditions, tune from the ISOLDE separator towards the ISOLTRAP setup and optimize the charge-exchange half-life.
Furthermore, **1 shift** is requested for the fine-tuning of cooling time and buffer-gas pressure with exotic argon isotopes to allow for the most efficient measurement cycle to be set up as well as optimization of target-ion source conditions.
Therefore, **in total 20 shifts are requested** to complete the proposed mass measurements of neutron-rich noble-gas isotopes including ^{49,50}Ar and ^{230,231}Rn to extend the nuclear structure evolution towards more neutron-proton asymmetry.

A	N	El.	δm (keV) [21]	Half-life [40]	Yield (ions/µC)	shifts
49	**31**	**Ar**	400*	170(50) ms	$1.1 \cdot 10^1$ [38, 41]	**3**
50	**32**	**Ar**	500*	106(6) ms	$2.0 \cdot 10^0$ [38, 41]	**12**
229	143	Rn	13	$12.0^{+1.2}_{-1.3}$ s	$1.8 \cdot 10^2$ [22, 38]	-
230	**144**	**Rn**	200*	-	$\approx 5.3 \cdot 10^1$ **	**1**
231	**145**	**Rn**	300*	-	$\approx 1.6 \cdot 10^1$ **	**2**

Table 1: Neutron-rich noble-gas isotopes with their current mass uncertainty, half-life, expected yield and allocated shifts.
* represents the uncertainty in the estimation from systematic trends given in AME2020 [21].
** indicates extrapolated yields.

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5 Details for the Technical Advisory Committee

5.1 General information

Description of the setup which will be used for the measurement:

- ☒ Permanent ISOLDE setup: ISOLTRAP
  - ☒ To be used without any modification
  - ☐ To be modified: -
- ☐ Travelling setup
  - ☐ Existing setup, used previously at ISOLDE: -
  - ☐ Existing setup, not yet used at ISOLDE: -
  - ☐ New setup: -

5.2 Beam production

- Requested beams: ^{49,50}Ar and ^{230,231}Rn
- Target - ion source combination: UC_x cold FEBIAD-type plasma ion-source
- RILIS? No
  - ☐ Special requirements: No
- Additional features?
  - ☐ Neutron converter: No
  - ☒ Other: water-cooled transfer line
- Expected contaminants: We expect the stable $^{33,34}S^{16}O$ to be the limiting contamination for the argon measurement. No further strong contamination is stated for Rn, whereas Fr might be expected.
- Acceptable level of contaminants: With the extrapolated yields of the ions of interest as well as expected contamination, the mass resolving power of ISOLTRAP's MR-ToF MS is sufficient to separate the 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, upcoming IDS proposal for At and Rn (different target), and IS759 of CRIS for Ar with same target type. Nonetheless, we request a new target, since the target conditions are known to vary significantly as the target ages [22].

5.3 Shift breakdown

The measurement of the proposed noble-gas isotopes will be performed in one run as follows:

Summary of requested shifts:

With protons	Requested shifts
Optimization of experimental setup using $^{47,48}Ar$	1
Data taking, ^{49}Ar	3
Data taking, ^{50}Ar	12
Data taking, ^{230}Rn	1
Data taking, ^{231}Rn	2
Without protons	Requested shifts
Target/Line conditioning, beam tuning from separator to setup, charge-exchange half-life optimization	1
Total	20

Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

The experimental setup comprises:

Part of the	Availability	Design and manufacturing
ISOLTRAP setup	☒ Existing	☒ To be used without any modification

HAZARDS GENERATED BY THE EXPERIMENT

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