EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Measurement of the excitation energy of the 5^+ isomeric state in ^{128}Sb for *r*-process nucleosynthesis

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Abstract:

We propose to measure the excitation energy of the 5^+ isomer in ^{128}Sb by directly measuring the masses of the isomer and the ground state to better than 1 keV with the Phase-Imaging Ion Cyclotron Resonance technique. The relative contributions of decay from the isomer and the ground-state of ^{128}Sb impact heating and photon emission following rapid neutron capture nucleosynthesis [1, 2]. The current estimate of ≤20 keV is based on the lifetime of the isomer, not a measurement. By determining the unknown excitation energy of the isomer, we will constrain the astrophysical energies at which the isomer and ground state must be treated independently, which significantly delays energy emission. This will also further constrain the energy limit, narrowing the range of possible decay paths.

Requested shifts: 8 shifts in one run with a UC_x target, neutron converter, and RILIS ionization in narrowband mode.

1 Motivation

One of the driving open questions in nuclear physics is "What is the origin of the heavy elements?" Observations of GW170817—first as gravitational waves by LIGO and followed by optical detection—have re-energized research in the rapid neutron capture process (*r*-process), which is believed to occur in explosive astrophysical events such as supernovae or merging neutron stars (NS mergers) [3, 4].

Interpreting these observations and understanding the timescales of the detected emission require improved fidelity in the nuclear physics that drives the nucleosynthesis and the decay to stability. Of particular interest are observational signatures that point uniquely to a particular isotope, reaction pathway, or astrophysical environment. Several such cases have already been hypothesized. Zhu *et al.* noted characteristic timescales in kilonova light curves that match the lifetime of ^{254}Cf [5]. Korobkin *et al.* looked at nuclear gamma rays that might be able to be directly observed as part of motivating future γ-ray astronomy missions [6]. Nuclear theory efforts have expanded the treatment of nuclear isomers, including the concept of what has been dubbed "astromers", nuclear isomers where it is necessary to treat the ground state and isomeric states as independent species in the nucleosynthesis network [7]. Extending this work, Misch *et al.* have estimated the contributions of individual astromers in *r*-process nucleosynthesis to highlight which decays are playing a significant role in the abundances and observational signatures over time [1]. In the past, many studies have relied on nuclear masses to investigate the neutron capture path for the *r*-process. In contrast, we are focused in this case on the role nuclear structure plays in the decay back to stability and how we can connect astronomical observations to individual nuclei in this process.

Nuclear structure information is necessary for this isomer treatment to be accurate. Level energies, spins, parities, and lifetimes of long-lived states are needed. Beta-decay feeding ratios of the ground state and isomeric states are required as well. Gamma-ray transition strengths between levels—particularly between ground and isomeric bands—have to be known. Together, this information is used to calculate transition rates between different states and isomers, whether those transitions are electromagnetic or weak in origin. Electromagnetic transitions play a particularly important role in a hot astrophysical environment as they provide a possible mechanism for the population of ground and isomeric states to come into equilibrium. When in equilibrium, the states are said to have thermalized. In many cases, the importance of an isomer changes as the temperature changes.

The energy differences between states are a key quantity in determining what states will thermalize. If the necessary transition energies are easily provided by the thermal photon bath, the ground state and isomer thermalize. If neutron capture or β-decay depletes one species, it will be replenished through thermalization, and the reaction flow is determined by the fastest reaction path. However, if the thermal photons do not provide the necessary transition energies, the ground state and isomer state are not in equilibrium and must be treated as independent species in the nucleosynthetic calculations.

Antimony-128 stands out in *r*-process isomer sensitivity studies at time scales of hours to single days [1, 2]. Figure 1 illustrates this, showing the "Astromer Importance Rating" (AIR), as developed in Misch *et al.*; it is a metric to identify the cases where an isomeric state significantly changes the flow in the network [1]. It is the product of the activity (A) of the isomer

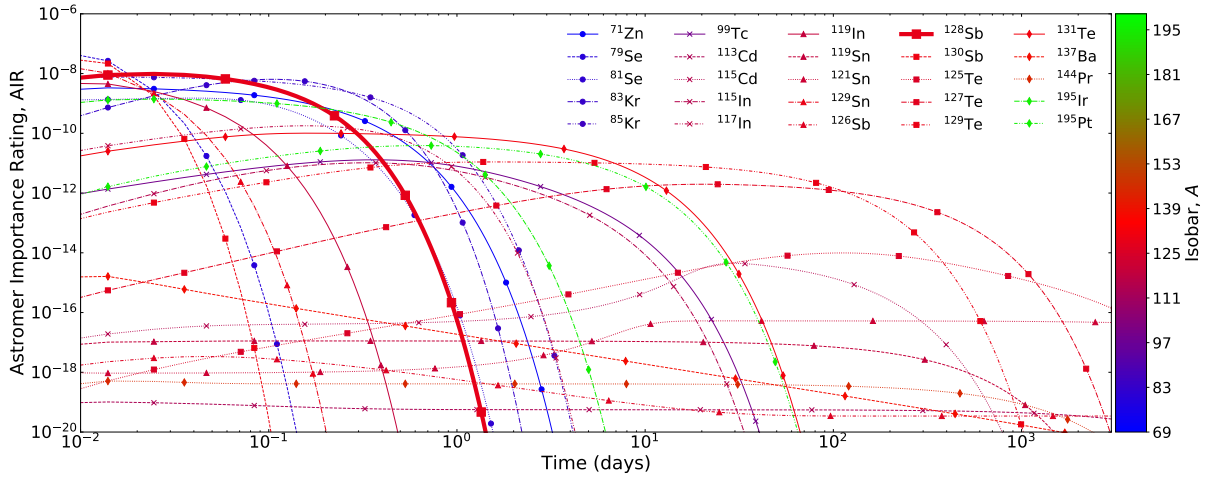

Figure 1: The figure above is adapted from Misch *et al.* and shows the role of individual astromers as a function of time since the *r*-process event [1]. The astromer importance rating (AIR), the vertical scale, is discussed in the main text in more detail. It is a metric for determining which isomers play an important role in *r*-process nucleosynthesis and the observational signatures. Between 0.5-2 hours, ^{128}Sb is the highest impact astromer in the network, and in truth, its effects extend much longer because it effectively accelerates ^{128}Sn decay toward ^{128}Te from ~ 9 hours (^{128g}Sb half-life) to ~ 1 hour (^{128}Sn half-life). The temperature at the 0.5-2 hour time scale depends strongly on the *r*-process event but is often in the range of $kT \lesssim 10$ keV. The different colors indicate whether the isotope participates in the abundances of the first, second, or third *r*-process abundance peaks.

(the rate of decays, reactions, and de-excitations in the isomer); the imbalance (I), the extent to which the isomer and ground state are out of thermal equilibrium; and the population ratio (R), the extent to which the isomer is significantly populated. A nucleus with a high AIR value includes an isomer that has high activity, is significantly decoupled from the ground state, and is significantly populated (either through direct feeding or freeze-out from prior thermal equilibrium). In the case of ^{128}Sb, it is significantly produced, and the factor of ~ 50 difference between the lifetime of the ground state and isomer impacts the timescale of the decay towards stability in the A = 128 mass chain. For ^{128}Sb, the high AIR value indicates that the isomer changes the heating rate, and since the isomer is shorter-lived, it drastically increases heating from ^{128}Sb decay at short times. The relative populations of the ground state and isomer are critical for interpreting the data. However, key data are missing for even developing the astromer network for ^{128}Sb.

The known low-lying structure of ^{128}Sb is shown in Figure 2. The 5^+ isomer is primarily fed by β-decay of the ^{128}Sn parent, which does not feed the ground state. The isomer β-decays both to ^{128}Te (96.4%) and via an $E3$ internal transition to the ^{128}Sb 8^- ground state (3.6%). This feeding of the ground state is relatively small. However, because of the large difference in lifetime between the isomer and the ground state, even a small fraction of decays to the ground state delays the decay of the A=128 chain and delays energy release from β-decay.

The excitation energy of the 5^+ isomer above the 8^- ground state is not known. This is the key quantity to be determined in this work. It is necessary to determine at what temperatures ^{128}Sb will thermalize. Once this is known, the decay rate of the A =128 mass chain

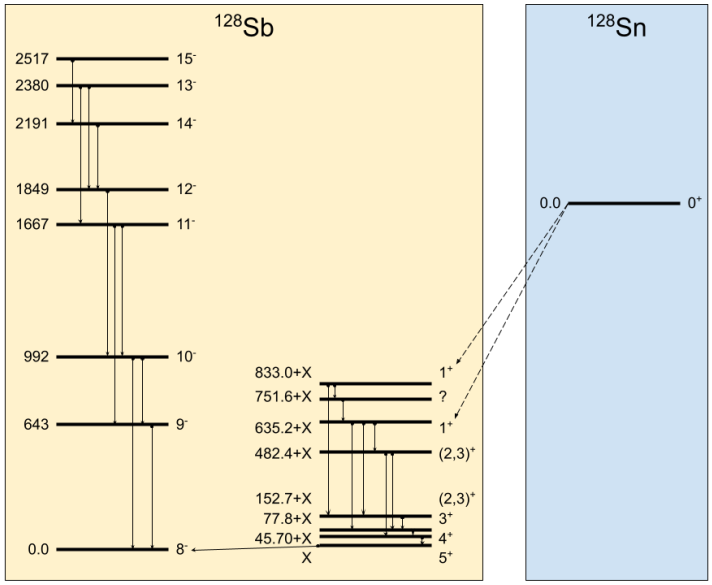

Figure 2: The present state of knowledge about ^{128}Sb level structure and branching ratios together with the β-decay of ^{128}Sn [8, 9]. The excitation energies of all of the levels built on the isomer are uncertain up to the isomer excitation energy.

can be calculated.

As of now, the excitation energy is estimated to be $\leq$ 20 keV [10], while the value 10(6) keV reported in the NUBASE2020 evaluation is based on the assumption that excitation energy is uniformly distributed between 0 and 20 keV. [11]. Currently, any estimation of equilibrium temperatures is completely based on theoretical assessments. Additionally, transition rates between levels built on the ground state and the isomeric state are not measured, further limiting the ability to predict the observational signature from this astromer. Thus, we propose to perform a first direct measurement of the excitation energy of the 5^+ isomer in ^{128}Sb. We will

- determine the excitation energy of the 5^+ isomer in ^{128}Sb by precisely measuring the isomer and ground state masses,
- use this energy to determine the range of energies for which ^{128g,m}Sb are in equilibrium astrophysically, and
- use this information to determine the timescale for the decay of $A = 128$ mass chain in r-process nucleosynthesis, which directly impacts the timescale of emission from this mass chain.

To achieve this, we need to measure the excitation energy to <1 keV precision, e.g. a relative mass precision of $< 10^{-8}$. We note that there is a chance that the excitation energy is less than ~5 keV, in which case it is unlikely that it will be possible to be distinguishable from the ground state. Such an experimental upper limit of the energy is still a powerful constraint on the isomer energy and will substantially advance our understanding of the A=128 decay chain.

2 Experimental techniques

The ISOLTRAP high-precision mass spectrometer [12] will be used for the determination of the excitation energy of ^{128}Sb by employing the well-established Phase-Imaging Ion-Cyclotron Resonance (PI-ICR) technique [13]. Currently, the apparatus consists of four ion traps optimized for different purposes: accumulation/bunching, separation, cleaning, and mass determination. A schematic view of the experimental setup is shown in Figure 3. The quasi-continuous radioactive ion beam (RIB) provided by ISOLDE first enters the radiofrequency quadrupole (RFQ) cooler/buncher [14] for accumulation, bunching, and emittance improvement. The bunched beam is then injected into the Multi-Reflection Time-of-Flight (MR-ToF) ion trap [15], which can be operated either as a beam purification/identification device or, in combination with a ToF detector, as a mass spectrometer (MS). In case the mass measurement is not performed in the MR-ToF mass spectrometer, the ion beam is then guided into the first of two separate Penning traps, the so-called preparation Penning trap. Here, the ion beam is further cooled and purified by employing the mass-selective buffer gas cooling technique [16]. Finally, the ion beam is injected into the precision Penning trap, in which the ion's mass is determined either via the Time-of-Flight Ion-Cyclotron-Resonance technique (ToF-ICR) [17] or via PI-ICR MS. The latter is based on imaging the accumulated phase φ_{acc} of a trapped ion for a given phase accumulation time t_{acc} using a position-sensitive detector. The extracted frequency ω can be used to calculate the cyclotron frequency $\omega_c = \frac{qB}{m}$ of the trapped ion.

The novel PI-ICR technique offers the unique opportunity to precisely measure isomer energies [18, 19]. This is a particularly important capability given that measurements of isomer energies from internal transitions via gamma spectroscopy can be severely limited for low gamma energies as the transitions are often highly converted. A direct mass measurement suffers from none of these limitations. Furthermore, high-precision Penning trap mass measurements in combination with resonant laser ionization have shown the capability of unambiguous identification and spin assignment of isomeric states [18, 20].

For measuring the excitation energy of the long-lived 5^+ we intend to employ the PI-ICR technique owing to the very low-lying excitation state, which necessitates high mass-resolving powers ($\sim 10^7$). As stated above, the exact excitation energy is not known and is expected to be ≤ 20 keV. Using resonant laser ionization with RILIS to selectively ionize either the isomeric state or the ground state, we will individually measure the masses of both states to then extract the excitation energy as a mass difference. As mass calibrants, ISOLTRAP's alkali ion source can provide stable ^{133}Cs ions, while the laser-ablation ion source (currently under construction) will provide $^{12}C_{10}$ and $^{12}C_{11}$ carbon clusters. With this technique, a 170 eV precision on a Q-value measurement at A=131 has been achieved recently

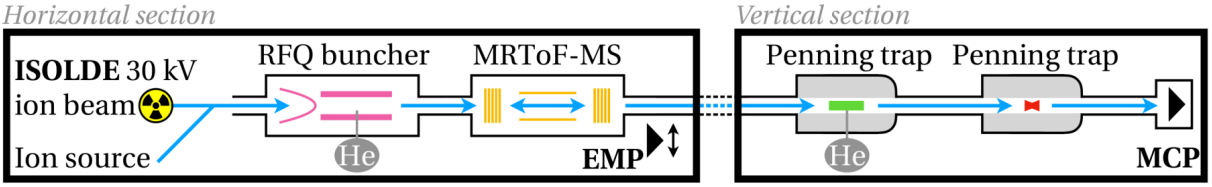

Figure 3: Schematic of the ISOLTRAP setup with its four ion traps and offline source.

at ISOLTRAP [21].

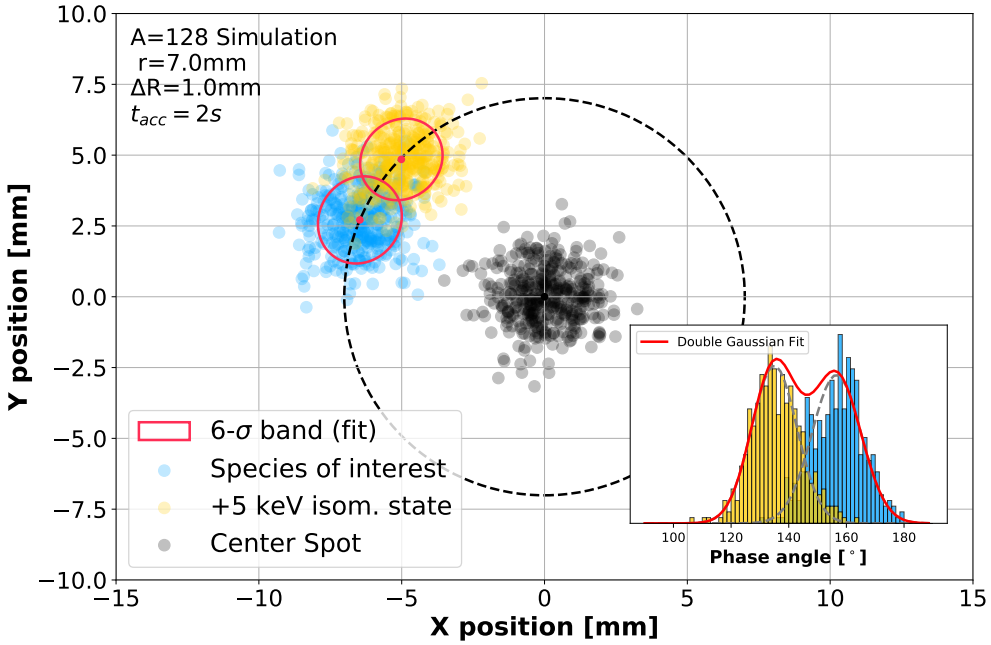

Figure 4: Monte-Carlo simulation to determine the necessary phase accumulation time. The main plot shows ground state (black), a possible 5 keV isomeric state (orange) after a phase accumulation time of 2 s, and a multivariate Gaussian fit of both spots (red). The insert shows a histogram of the projected phase angle of both states with a double-Gaussian fit. In this simulation, rest-gas collisions and ion-ion interactions have not been considered.

Good spatial separation of both ground state and isomeric state on the position sensitive detector is paramount for extracting the excitation energy as a mass difference. Based on recent experimental conditions at a similar atomic mass (A= 129) [18], Monte-Carlo simulations have been performed to estimate the necessary phase accumulation time in order to separate the two states for a given excitation energy. The results are shown in Figure 4. Assuming a possible 5 keV isomeric state, a phase accumulation time of 2 s is thus necessary to reach sufficient spatial separation. It can be seen that long accumulation times[1] are needed to resolve such low lying states, corresponding to a mass resolving power of 2.4×10^7 at A= 128. In this time regime, rest-gas collisions can cause major spot distortion that may lead to extended overlapping and blurring of the image. The main contribution to the rest-gas is helium leaking from the preparation Penning trap into the precision Penning trap. To mitigate this leakage, the cooling cycle in the preparation Penning trap shall be extended with reduced helium pressure[2]. If the highest achievable accumulation time turns out not to be sufficient to resolve both species, an upper limit on the transition energy can be extracted from the data.

[1] A clean PI-ICR image with a two second phase accumulation time has been demonstrated at ISOLTRAP during preparation measurements using the offline ion source.

[2] An arbitrary long cooling cycle is, in this study, possible due to the long half-lifes of the ground and isomeric states.

3 Beam time request

During the experimental run for IS635 conducted at COLLAPS (UC_x target with neutron converter), a whole variety of neutron-rich antimony isotopes was produced. Comprehensive yield estimations were performed, and the yield for ^{128}Sb was found to be ~ 10 pA (~ 10^8 ions/second), including about 10% contamination [22]. No further information about the nature of the contaminant(s) during the experiment is available. However — based on the ISOLDE Yield Database [23, 24] — possible surface ionized contaminants are ^{128}Cs and ^{128}Te. With mass differences of 1.3 MeV and 4.4 MeV, a mass-resolving power of at least 200,000[3] is needed to resolve and separate the species. The MR-ToF mass separator — using active and passive voltage stabilization [25] in combination with mass-selective ion ejection [26] — is fully capable of cleaning the radioactive ion beam from this contamination. Furthermore, no loss of counts caused by in-flight or in-trap decay is expected due to the long half-lifes of ^{128g}Sb and ^{128m}Sb. This enables not only a high number of revolutions in the MR-ToF MS, increasing the separation capability if needed, but also a long cooling cycle in the preparation Penning trap with lower helium buffer gas pressure, reducing rest gas load in the precision Penning trap.

From IS635 it is known that the production for the 8^- ground state is favored over the 5^+ isomeric state. Due to the large spins involved, from the transition strengths it can be estimated that the production ratio will not be lower than 4/1 and not be higher than 10/1 [27]. For an unambiguous production of ground and isomeric state, resonant laser ionization will be used. To this end, isomer selective ionization through RILIS in narrowband mode can be employed [28] to promote the production of either the 8^- state or the 5^+ state. Based on the COLLAPS measurements, it is expected that the 5^+ isomer can be produced while fully suppressing the 8^- state. Due to overlapping transitions, the 8^- state will be produced with a very small contribution of 5^+ in a ratio of about ten to one.

The shift count — detailed in table 1 — is based on different considerations. First, the yields of the ground state and isomeric state with ~ 10^8 ions/second do not pose a limit for the measurement. Second, the global transport and detection efficiency from CA0 to the position-sensitive detector behind the precision Penning trap is assumed to be ~ 0.1%[4], while ionization losses through selective ionization are not smaller than 30% for the ground

[3] At mass $A = 128$, a resolving power of 2×10^5 corresponds to a mass difference of about 600 keV or a time-of-flight of 70 ns.

[4] This value includes losses from CA0 to the MR-ToF, losses from the MR-ToF to the detector and the detection efficiency

Table 1: Isotope properties and detailed summary of the shift request. Spins and parities are assigned based on structure arguments .

Isotope	Half-Life [h]	J^π	E(level) [keV]	Yield in CA0 [ions/s]	Target / Ion source	Method	Shifts (8h)
^{128g}Sb ^{128m}Sb	~ 9 h ~ 10 min	8^- 5^+	0 ≤ 20	10^8	UC_x / RILIS	PI-ICR and MR-ToF MS	4
Beam optimization							4

state and 80% for the isomeric state. Third, the phase accumulation time t_{acc} is assumed to be ~ 2 s which is a major contribution to the overall experimental cycle (~ 3 s). All in all — aiming for the target precision of < 1 keV and including reference measurements with an offline calibrant — two shifts per state are required. Additionally, four shifts are needed to study the ISOLDE beam composition and to optimize beam transport to and throughout the setup, as well as for setting up and tuning the RILIS lasers for the proposed isomer splitting (two shifts per isomer).

Summary of requested protons: 8 shifts in one run with a UC_x target, neutron conversion, and RILIS ionization in narrowband mode.

References

[1] G. Wendell Misch, T. M. Sprouse, and M. R. Mumpower. Astromers in the radioactive decay of r-process nuclei. *Astrophys. J. Lett.(Accepted for Publication)*, 2021. Available on `https://arxiv.org/abs/2011.11889v2`.

[2] Shin-ichiro Fujimoto and Masa-aki Hashimoto. The impact of isomers on a kilonova associated with neutron star mergers. *Mon. Not. R. Astron. Soc.*, 493(1):L103–L107, March 2020.

[3] Abbott et al. Gw170817: Observation of gravitational waves from a binary neutron star inspiral. *Phys. Rev. Lett.*, 119:161101, Oct 2017.

[4] Kasliwal et al. Illuminating gravitational waves: A concordant picture of photons from a neutron star merger. *Science*, 358(6370):1559–1565, 2017.

[5] Yu Zhu et al. Californium-254 and Kilonova Light Curves. *Astrophys. J. Lett.*, 863(2):L23, August 2018.

[6] O. Korobkin et al. Gamma Rays from Kilonova: A Potential Probe of r-process Nucleosynthesis. *Astrophys. J.*, 889(2):168, February 2020.

[7] G. Wendell Misch, Surja K. Ghorui, Projjwal Banerjee, Yang Sun, and Matthew R. Mumpower. Astromers: Nuclear Isomers in Astrophysics. *Astrophys. J. Suppl. Ser.*, 252(1):2, January 2021.

[8] Zoltan Elekes and Janos Timar. Nuclear data sheets for A = 128. *Nuclear Data Sheets*, 129:191–436, 2015.

[9] S. Biswas et al. Effects of one valence proton on seniority and angular momentum of neutrons in neutron-rich $^{122--131}_{51}$Sb isotopes. *Phys. Rev. C*, 99:064302, Jun 2019.

[10] N. Imanishi, I. Fujiwara, and T. Nishi. Decay of 59.3 min ^{128}Sn to levels of ^{128}Sb. *Nuclear Physics A*, 238(2):325–332, 1975.

[11] W. Huang. Private communication with NUBASE team.

[12] M. Mukherjee et al. *The European Physical Journal A*, 35(1):1, 2008.

[13] S. Eliseev et al. A phase-imaging technique for cyclotron-frequency measurements. *Applied Physics B*, 114(1-2):107, 2014.

[14] F. Herfurth et al. A linear radiofrequency ion trap for accumulation, bunching, and emittance improvement of radioactive ion beams. *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, 469(2):254–275, 2001.

[15] R.N. Wolf et al. *International Journal of Mass Spectrometry*, 349-350:123, 2013.

[16] G. Savard, St. Becker, G. Bollen, H.-J. Kluge, R.B. Moore, Th. Otto, L. Schweikhard, H. Stolzenberg, and U. Wiess. A new cooling technique for heavy ions in a penning trap. *Physics Letters A*, 158(5):247–252, 1991.

[17] M. König et al. Quadrupole excitation of stored ion motion at the true cyclotron frequency. *International Journal of Mass Spectrometry and Ion Processes*, 142(1):95–116, 1995.

[18] V. Manea and Karthein et al. First glimpse of the N = 82 shell closure below Z = 50 from masses of neutron-rich cadmium isotopes and isomers. *Physical Review Letters*, 124(9):092502, 3 2020.

[19] M. Mougeot et al. Challenging new ab initio nuclear theory of the iconic nuclide ^{100}Sn through mass measurements of $^{99-101}$In, Submitted to Nature Physics (2021).

[20] J. Van Roosbroeck et al. Unambiguous identification of three β-decaying isomers in ^{70}Cu. *Phys. Rev. Lett.*, 92:112501, Mar 2004.

[21] J. Karthein et al. Direct decay-energy measurement as a route to the neutrino mass. *Hyperfine Interactions*, 240(1):61, Jun 2019.

[22] S. Lechner. Private communication with COLLAPS team.

[23] ISOLDE Yield Database. `https://isoyields2.web.cern.ch/`.

[24] S. Rothe. Private communication with target team.

[25] F. Wienholtz et al. *Nuclear Instruments and Methods B*, 463:348, 2020.

[26] F. Wienholtz et al. *International Journal of Mass Spectrometry*, 421:285, 2017.

[27] M. Bissell. Private communication with COLLAPS team based on prelim. results from IS635.

[28] K. Chrysalidis. Private communication with RILIS team.

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	☒ Existing	☒ To be used without any modification

HAZARDS GENERATED BY THE EXPERIMENT

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