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

MASSES OF NOBLE GASES

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Abstract

The so-called magic numbers, cornerstones of the quantum nuclear ensemble, are now known to lose their supernatural powers far the protected valley of stability. To complement the well-established (but not yet well-understood) case of $N = 20$, we propose to examine the erstwhile $N = 28$ shell closure via a measurement of the important (but unknown) mass of the nuclide ^{48}Ar. The quenching of a shell closure, a mechanism as mysterious as the reason for magic numbers themselves, also has important consequences in nucleosynthesis. While ^{48}Ar is not part of the region concerned by the canonical rapid neutron-capture (*r*) process, the question of shell strength is of great importance for heavier nuclides. The location of the *r*-process path would benefit from extending the successful ISOLTRAP krypton mass measurements beyond the $N = 58$ sub-shell to $^{96\text{-}98}$Kr. Modeling the complementary rapid proton-capture (*rp*) process, putative source of some proton-rich species, requires the mass of ^{70}Kr, near the established waiting point at ^{68}Se and determined to be a critical input for *rp*-process reaction networks. Finally, extending other earlier ISOLTRAP work, the unknown masses of $^{111\text{-}113}$Xe, located in an interestingly isolated region of alpha activity, would provide missing links to several other exotic nuclides, including the proton-emitting ^{112}Cs.

These measurements will be feasible thanks to the stunning performance of the recently-developed VADIS arc-discharge ion source (with cooled transfer line) for Xe and Rn isotopes. Three different targets would be required, and a total of 42 shifts of beam time.

Summary: Mass measurements of $^{46\text{-}48}$Ar, $^{70\text{-}72}$Kr, $^{96\text{-}98}$Kr, and $^{111\text{-}113}$Xe using ISOLTRAP.

1. Introduction

The mass of a nuclide is arguably its most fundamental property. From the mass, the binding energy reflects the result of all interactions at work in the atom. Thus, many facets of physics can be studied from the measurement of just this one quantity. The most general interest in the mass is nuclear structure since the binding energy is determined by the very configuration of the nuclear constituents that minimize this quantity. We have learned that studying these configurations in extreme cases, where neutrons greatly outnumber protons, reveals surprising features of nuclear structure not occurring closer to the more balanced, valley of stability. Aside from nuclear halos, perhaps one of the most intriguing paradigms is the study of shell structure – and the disappearance thereof. The mass surface is an excellent indicator of shell structure and hence, its disappearance. Since the mass also tells us the amount of energy available for reactions and decays, it is a quantity of capital importance for nucleosynthesis. The different processes that cook the heavy elements inside stars and power observable astrophysical phenomena all involve the binding energies of nuclides that are near the limits of stability. Finally, exotic nuclides have large energy windows for different decay modes. Knowledge of these energy budgets helps constrain studies of these types of decays and establish the correct links between the different daughters.

The tandem Penning trap mass spectrometer ISOLTRAP [Muk08] installed at the on-line isotope separator ISOLDE at CERN/Geneva plays a leading role in the field of mass measurements in general [Lun03]. In particular, ISOLTRAP is the pioneering Penning-trap spectrometer [Bla06] and has made several hundred measurements over the past 20 years, now routinely reaching relative uncertainties of 10^{-8}, highlighting physics in great panoply.

In this proposal we would like to address four different areas of the nuclear chart, each with a particular physics motivation. The unifying feature is the use of the newly-developed VADIS source for noble gases, the remarkable performance of which allowed the very first observation of the nuclide ^{229}Rn [Neid09]. The four areas concerned are:

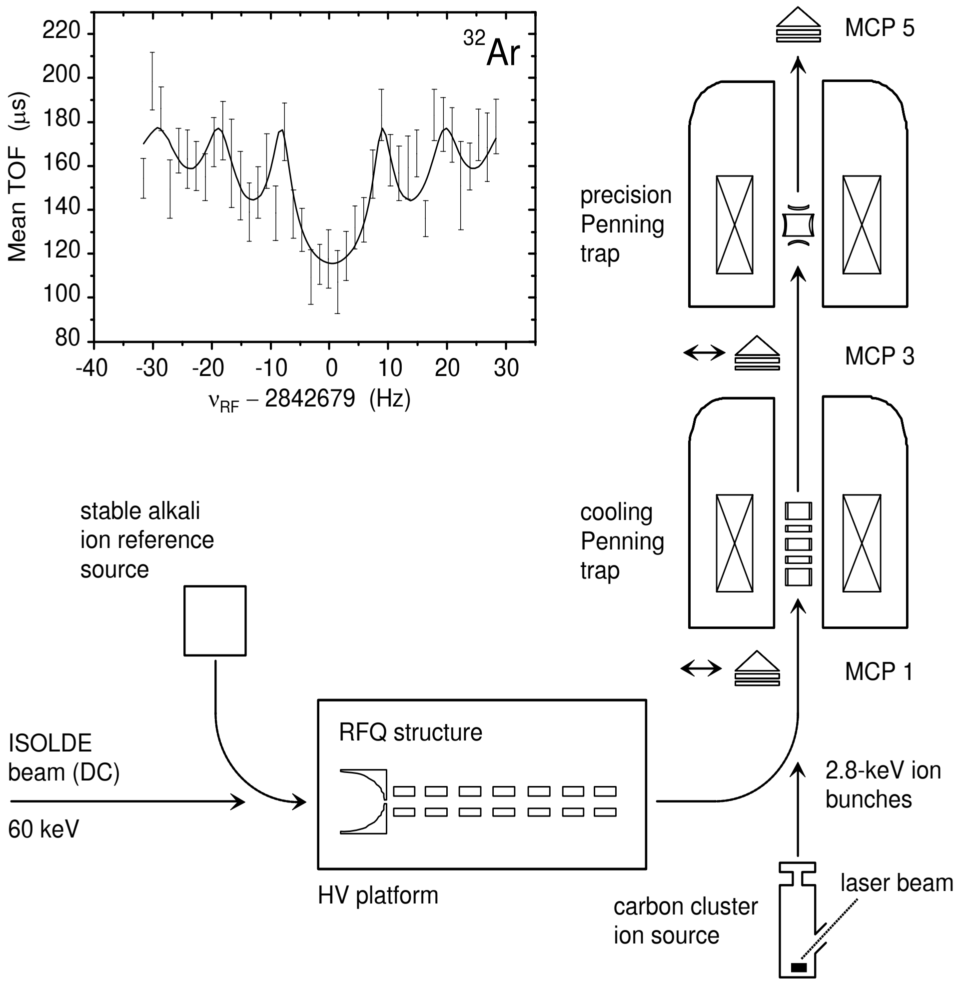
- $^{46-48}$Ar: These nuclides cross the $N = 28$ shell closure, recent studies of which conclude that it is not a shell closure at all. This work would extend previous ISOLTRAP work.
- $^{96-98}$Kr: Another extension of previous ISOLTRAP measurements which would encompass most realistic rapid neutron-capture paths and aid in constraining the closely-associated development of microscopic mass models.
- $^{70-72}$Kr: These are important nuclides involved in the modeling of x-ray bursts and the rapid proton-capture process that powers them. They are also an extension of previous ISOLTRAP work (reaching ^{72}Kr).
- $^{111-113}$Xe: Nuclides close to the proton drip-line linked via proton-emission and other decay Q-values that will anchor floating links and calibrate other measurements. Again, ISOLTRAP has investigated this region in the past.

2. Mass measurements with ISOLTRAP

The triple-trap mass spectrometer ISOLTRAP has now been operated at ISOLDE for many years (see [Muk08] for a recent, detailed description). The essential technique is trapped-ion cyclotron motion excitation with time-of-flight detection, as described in the following.

2.1 Experimental Setup

The present layout of the ISOLTRAP spectrometer is shown in the figure below. It consists of three distinct stages: a radiofrequency quadrupole (RFQ) preparation trap, and a tandem Penning trap setup.



Schematic drawing of the ISOLTRAP mass spectrometer with carbon-cluster and alkali-metal ion sources. Multi-channel plate (MCP) detectors are used to monitor the ion transfer as well as to record the time-of-flight resonance (MCP5) for the determination of the cyclotron frequency. The spectrum shows the cyclotron resonance curve of $^{32}Ar^{+}$ with a fit of the theoretically expected line shape to the data points.

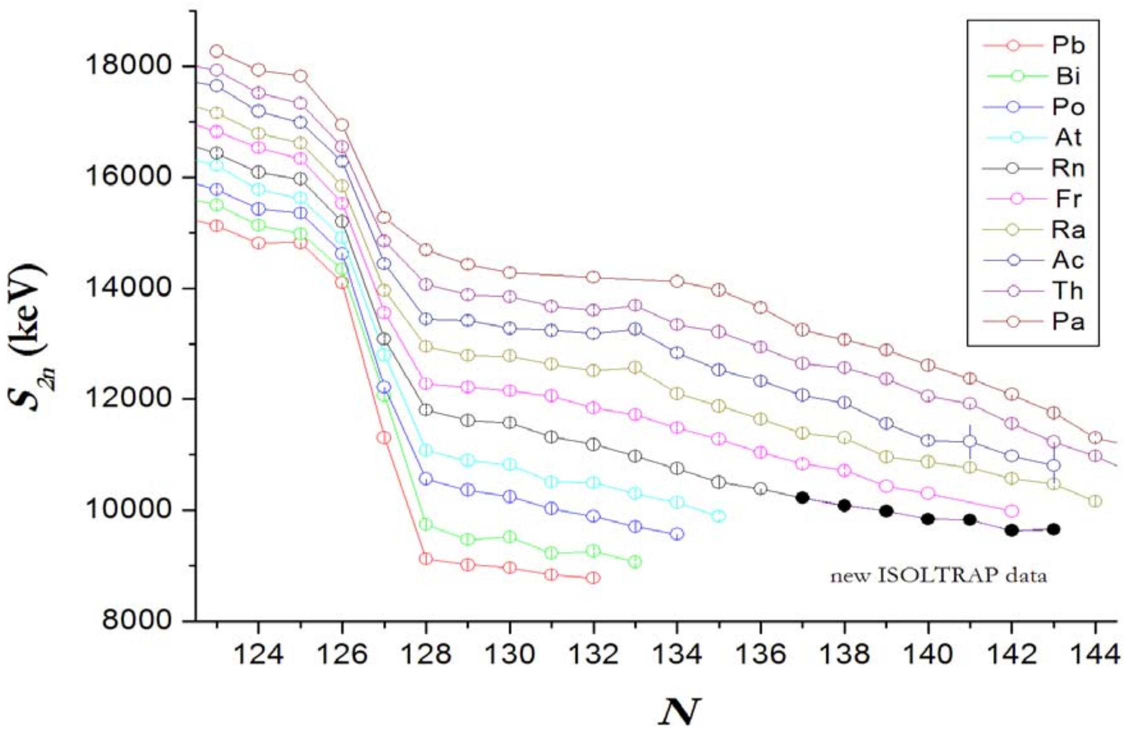
The linear gas-filled RFQ ion trap stops the 60 keV continuous ISOLDE beam to prepare it for efficient transfer into the cooler trap. The ISOLDE beam is electrostatically retarded for entry into the RFQ, where the ions are cooled by energy loss from collisions with buffer gas. After an appropriate accumulation time (typically 10-20 ms) the bunched ions are transferred at low energy into the tandem Penning-trap system. The first Penning trap is of a large-volume, cylindrical geometry, and separates the desired species from contaminants, using a mass-selective, buffer-gas cooling technique [Sav91]. The ions are then transferred to the second, hyperbolical Penning trap, where a precision measurement of the cyclotron frequency is performed. A mass measurement consists of a series of trial excitations of the cyclotron motion, each excitation followed by axial ejection of the ions and time-of-flight analysis of the absorbed energy [Grä80]. The measurement of the time of flight as a function of the frequency ν_{RF} of the applied azimuthal RF-field, results in a characteristic cyclotron resonance curve [Kön95], shown in the inset of Fig. 1. The mass m of an ion with charge q is obtained by the comparison of its cyclotron frequency: $\nu_c = 1/2\pi \cdot q/m \cdot B$, with the cyclotron frequency of a known reference ion mass. Since the magnetic field is subject to a long-term exponential decay as well as to short-term fluctuations, a reference measurement is carried out both before and after the measurement of the cyclotron frequency of the ion of interest.

2.2 Specifications and highlights of ISOLTRAP

2.2.1 Efficiency

The total efficiency of the ISOLTRAP setup is roughly 1% for stable isotopes, including the transfer efficiency of ISOLDE, the transport efficiency of the ISOLTRAP spectrometer and the detection efficiency of the MCP5. For very short-lived isotopes e.g., ^{32}Ar ($T_{1/2}$ = 98 ms), we reached a total efficiency of ~0.1%, lower due to decay losses and larger emittance of the outgassing target. This high efficiency allowed us to measure the mass of ^{32}Ar with an ISOLDE yield of ~100 atoms/proton pulse to a precision of better than $\delta m/m < 1 \cdot 10^{-7}$ within three shifts. The data shown in Figure 1 is an accumulation of 6 hours.

Since that time, other measures have been undertaken to improve efficiency, notably the installation of a channeltron-type detector [Yaz06]. A very successful experiment in 2008 with the UC target and a newly developed arc-discharge (VADIS) ion source enabled ISOLTRAP to extend the chain of neutron-rich Xe isotopes by four masses and in the case of Rn, *seven* new masses were measured, with the highlight of discovering ^{229}Rn, never before reported [Neid09]. Results from this landmark experiment are shown in the following figure. This combination of high efficiency (from both the experiment and the production facility) is what makes proposing these more exotic nuclides possible.



The two-neutron separation energies (S_{2n}, from [AME03]) for nuclides north-east of ^{208}Pb showing the seven new Rn values, including the newly-discovered ^{229}Rn (from [Neid09]).

2.2.2 Accuracy

In a pioneering effort using carbon clusters as etalons, an upper limit of 8×10^{-9} was determined for the relative, mass-dependent systematic error over a wide mass range of 250 u, covering the complete nuclear chart [Kel03]. With the knowledge gained from these studies we were able to determine the mass of ^{34}Ar ($T_{1/2}$ = 844 ms) with a relative uncertainty of only 1.1×10^{-8} [Her02]. This performance has set a new standard for the accuracy of on-line mass measurements.

2.2.3 Resolving power

High accuracy mass measurements with Penning traps require clean beams to avoid systematic errors in the mass determination arising from Coulomb interaction of different ion species in the trap. Therefore, the first Penning trap of ISOLTRAP uses a mass-selective cooling technique capable of a mass-resolving power of about $R = 10^5$, which is sufficient to resolve and separate isobars even close to stability. The resolving power of the precision trap is given by [Bol01]: $R = m / \Delta m = \nu_c / \Delta\nu_c \approx 1.25 \cdot \nu_c \cdot T_{obs}$. Hence, the resolving power can be increased with ion-observation time T_{obs}. For $T_{obs} \approx 1$ s, a resolving power R close to one million is reached with the precision Penning trap for A=100.

Obviously, the maximum resolving power will be limited by the half-life of the observed nuclide. In this case, the accuracy must be recovered by increasing the statistics. On the other hand, the observation time can be increased to obtain sufficient resolving power for the separation of sufficiently long-lived isomeric states, as performed in the case of the Hg isotopes [Sch01]. In fact, the ISOLTRAP results for ^{187,191}Hg were the first in which isomeric energies were directly determined via mass spectrometry. ISOLTRAP was also the first mass spectrometer to *separate* isomeric states – three of them, in the case of ^{70}Cu [Roo04]. These feats may well be necessary in the case of ^{111}Xe if its purported 900-ms isomeric state is produced at ISOLDE.

2.2.4 Half-life

The shortest-lived isotope ever investigated with ISOLTRAP is ^{74}Rb, with a half-life $T_{1/2}$ of only 65 ms [Kel04]. The relative accuracy of its mass as obtained in this measurement, $\delta m/m \approx 2.6 \cdot 10^{-7}$ (i.e. δm = 18 keV), is governed mainly by statistics. Note that the record was set recently at TRIUMF by TITAN with a mass measurement of ^{11}Li, having a half-life of only 8.8 ms [Smi08].

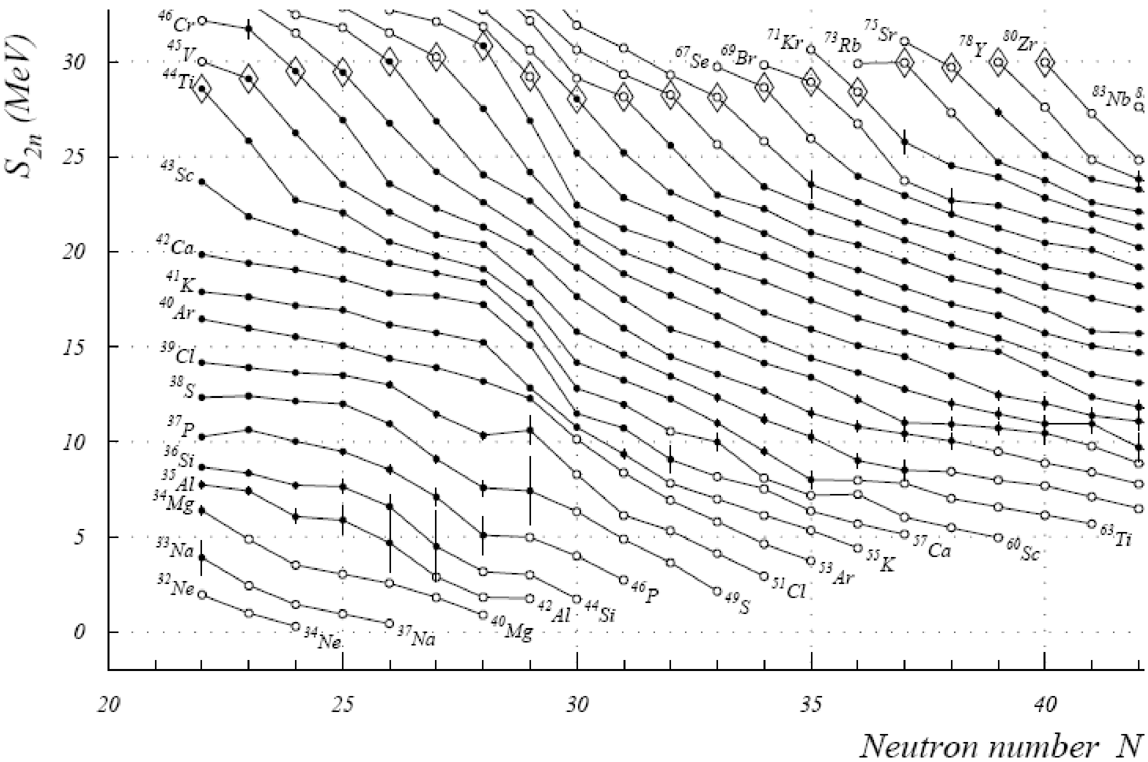
3. Experimental Program

The four different areas and themes are outlined in the following subsections.

3.1 Disappearance of the *N* = 28 closed shell: ^{46,48}Ar

The so-called magic numbers that form exceptionally bound nuclides, are now known to lose their supernatural powers as their isospin loses equilibrium. The most well-established case of an eroded shell closure is the famous island of inversion of Na and Mg nuclides at $N = 20$, discovered by pioneering on-line mass measurements [Thib75] at CERN using the PS beam. This region has also been intensely studied at ISOLDE, by mass measurements with MISTRAL [Lun01,Lun06] and more recently by laser spectroscopy and β-NMR by

COLLAPS [Ney05, Yor07]. The case of $N = 28$ has come into the limelight more recently. These isotones are also now the subject of much scrutiny using a plethora of experimental techniques including reactions and nuclear spectroscopy, for example from gamma spectroscopy, in the case of $Z = 14$ [Bast07]. Again, mass measurements provided the first indication that there may be a weakening of this shell [Sara00]. Unfortunately, those measurements had a very large uncertainty and some of them were excluded from the 2003 Atomic-mass evaluation [AME03]. The mass surface from that work is illustrated in the following figure.



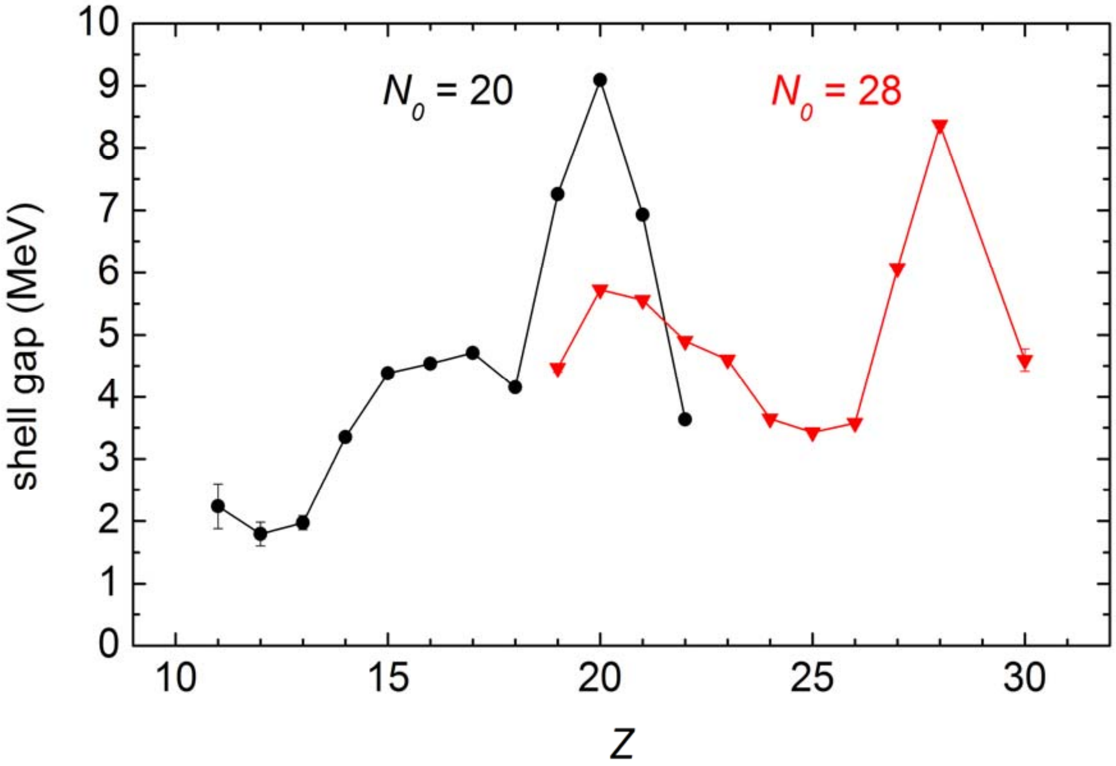
Two-neutron separation energies in the region of ^{48}Ar (from [AME03]). The kink at N = 28 is not as pronounced as for the heavier isotopic chains but since the mass of ^{48}Ar is unknown, it is difficult to say with certainty that the shell closure is really quenched.

One of the most solid indicators for shell strength is the so-called shell gap, defined as the difference in neutron separation energies at and two neutrons after the closed shell: $S_{2n}(Z,N+2) - S_{2n}(Z,N)$. This quantity is illustrated in the following figure for the cases of $N = 20$ and $N = 28$. As the mass of ^{48}Ar is still unknown, there is no value for $Z = 18$. In order to help decide if the shell is still closed, a measurement of this mass is crucial.

Masses are not the only indicator however: recent spectroscopic factor results from a $^{46}Ar(d,p)^{47}Ar$ transfer-reaction study [Gaud06] concluded that the $N = 28$ shell was indeed weakened for $Z = 18$ as compared to $Z = 20$. Moreover, they derived a mass for ^{47}Ar (with a 90-keV uncertainty) over 700 keV less bound than the value in [AME03]. Curiously, the consequence is that the S_{2n} value for ^{47}Ar in the previous figure *decreases* (from 12.28 MeV to 11.57 MeV) which tends rather to resurrect the magic status of $N = 28$. It is therefore extremely important to verify this value with a different type of measurement of comparable (or better) precision.[1]

[1] It is interesting to note that a Comment [Sig07] was written with respect to [Gaud06] specifying the importance of fragmentation of *p*-shell occupation and proposing an alternative formulation using absolute binding energies.

Since the shell gap requires also the ^{46}Ar mass, we would like to measure this quantity with the same precision as the rest of the isotopes in this chain. Note that the masses of $^{44\text{-}45}$Ar were already measured by ISOLTRAP [Bla03].

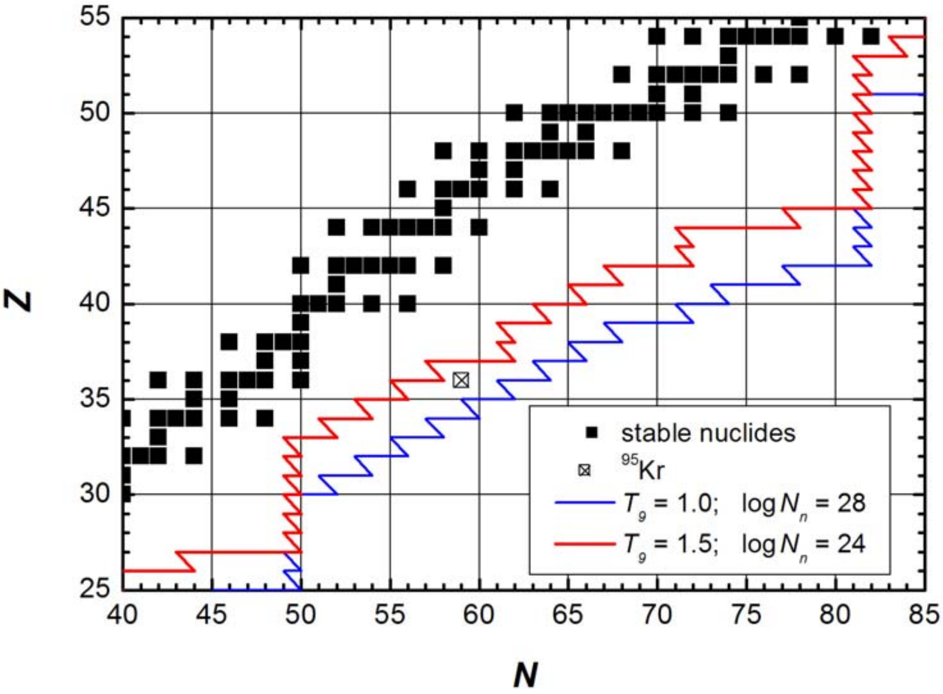


Shell gaps derived for N = 20 and 28 as a function of proton number Z (data from [AME03]. The large peaks correspond to the cases of exceptional binding where N = Z. The question is whether the N = 28 case will drop (i.e. quench) like the N = 20 case does. The proposed measurement of ^{48}Ar will answer this important question.

3.2 Exploring the r-process path: $^{96\text{-}98}$Kr

The rapid neutron-capture (*r*) process, thought to occur in exploding supernova, is responsible for the creation of about half the heavy elements found in nature [Arn07]. Masses of exotic nuclides are only one ingredient, but one of particular importance given the strong dependences of reaction cross-sections on *Q*-values. As the *r* process proceeds very far from stability (see following figure), there is no solution but to use models. However, models are constrained by measurements so pushing farther from stability is very important.

The quenching of a shell closure, an effect not predicted by most nuclear models, has important consequences in nucleosynthesis. Although most *r*-process scenarios do not involve the lighter species around N = 28, the correct modeling of shell structure (or lack thereof) is very important for mass models. This question has been very recently tackled by ISOLTRAP in the cases of N = 50 for ^{81}Zn [Bar08] and N = 82 for ^{134}Sn [Dwo08].

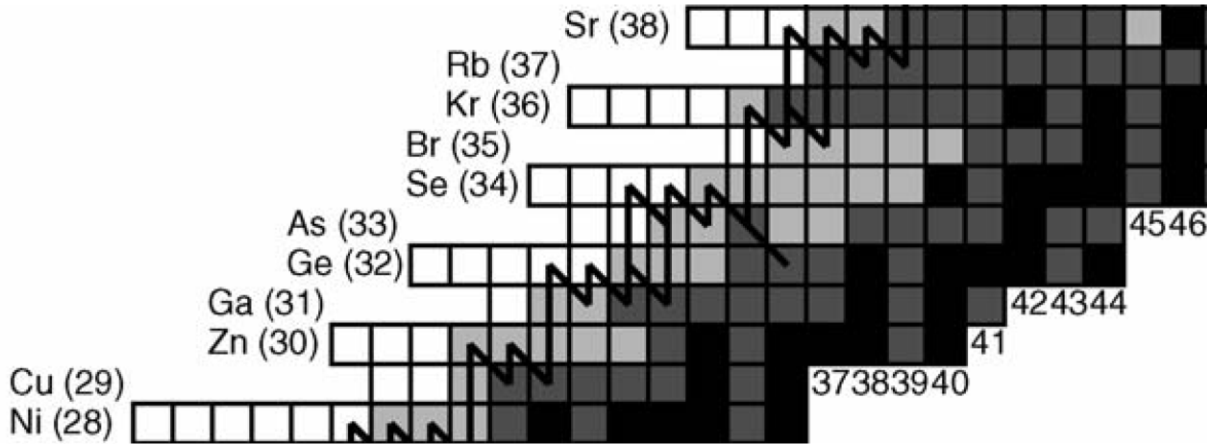


Nuclear chart showing the rapid neutron-capture path for two sets of astrophysical conditions (from the ISOLTRAP paper [Del06]). While certain conditions place with r-process path closer to stability, mass measurements up to ^{98}Kr would encompass a much greater realm of astrophysical possibilities as well as constraining mass models.

Other previous work by ISOLTRAP of relevance for the *r*-process was addressed with measurements of neutron-rich Kr [Del06] (see above figure). As in that work, the question of the *r*-process path would benefit from extending the successful ISOLTRAP Kr mass measurements to $^{96\text{-}98}$Kr, beyond the $N = 58$ sub-shell. Here the importance is to test the prediction of microscopic mass models (see e.g., the latest in the impressive development of Hartree-Fock-Bogoliubov models [Cha08] and references therein).

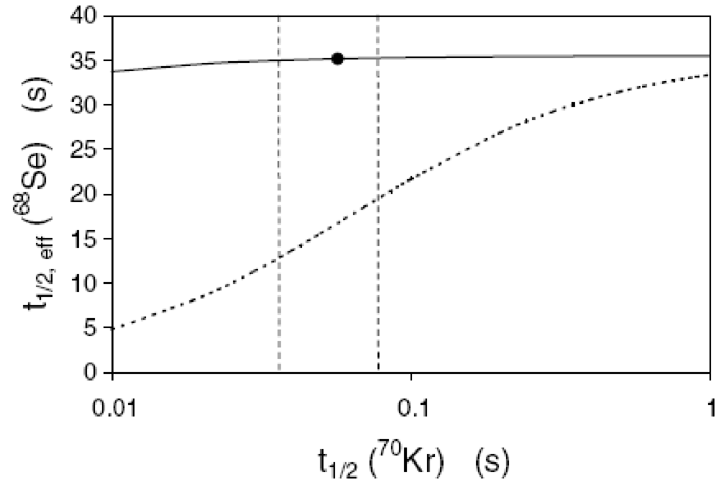
3.3 X-ray bursts and the rp process: ^{70}Kr

The rapid proton-capture (*rp*) process is thought to be responsible for the energy generation of type-I X-ray bursts in accreting neutron stars [Scha98]. As illustrated in the following figure, the *rp* process follows isotonic chains to the point where proton capture is inhibited by photodisintegration (or proton emission). At this, so-called waiting point, the process is delayed by beta-decay, which allows the flow to continue to heavier elements. The nuclides ^{64}Ge, ^{68}Se and ^{72}Kr all have beta-decay half-lives that are long compared to the *rp*-process timescale of 10 – 100 s. Thus, these waiting-point nuclides can generate extended observable tails in X-ray burst light curves [Scha06]. The beta-decay of ^{68}Se offers one escape from this situation, the other being two sequential proton captures: ^{68}Se (p,γ) ^{69}Br (p,γ) ^{70}Kr. Amongst the most critical quantities to determine the 2p-capture rate and the effective lifetime of the ^{68}Se waiting point are: the beta-decay half-lives of ^{68}Se and ^{70}Kr and the proton separation energies of ^{69}Br and ^{70}Kr. The 2p-capture rate has an exponential dependence on the masses of the nuclides involved. Therefore, the uncertainties of these masses need to be below 10 keV [Scha06].



Partial reaction network showing the main flow of the rp-process path in the N=Z=34 region (from [Scha06]). Here, ^{68}Se and ^{72}Kr are considered waiting points.

A better understanding of X-ray burst phenomena would be achieved with an improvement of the effective lifetime of ^{68}Se. Under equilibrium conditions at high temperatures, the effective lifetime of ^{68}Se is proportional to the beta-decay lifetime of ^{70}Kr. However, the relevant temperature range will depend on its proton-separation energy. Therefore a mass measurement of ^{70}Kr would reduce the uncertainty of the ^{68}Se effective lifetime. Estimates indicate that a 30% variation in this quantity results in a factor-of-five variation in the ^{68}Se effective lifetime (see following figure, taken from [Cla04]).

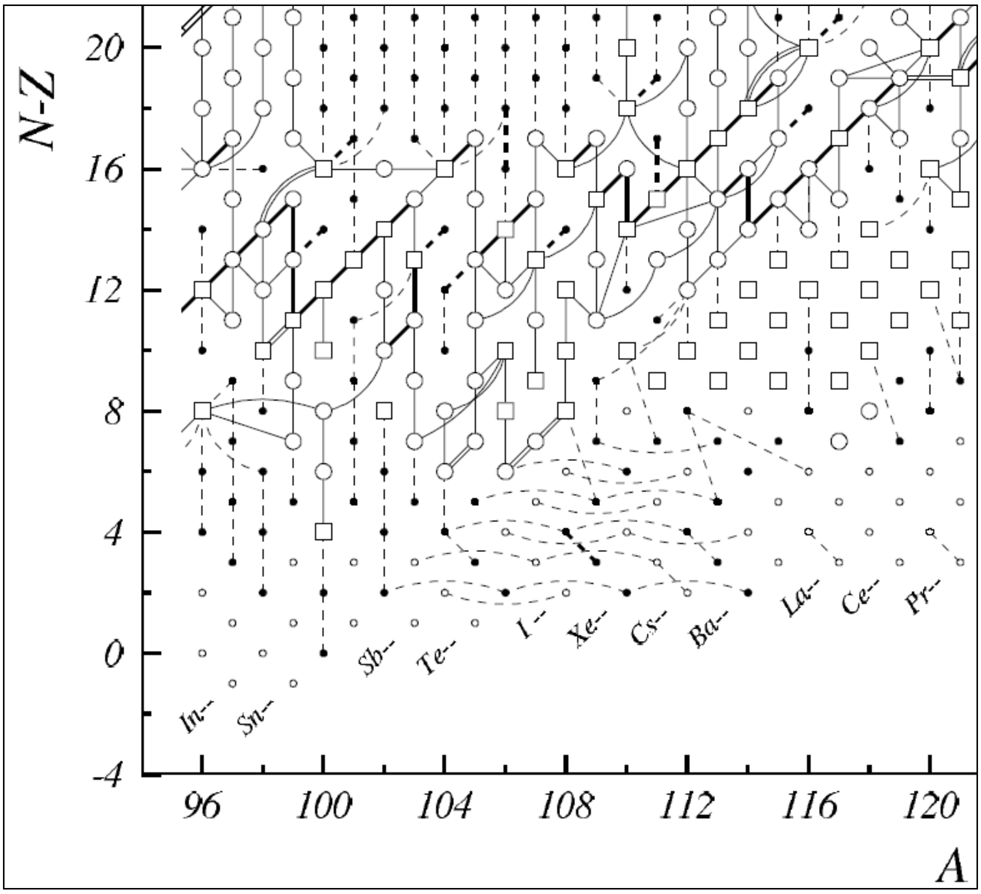


Effective stellar half-life of the ^{68}Se rp-process waiting point versus the half-life of ^{70}Kr (dot with error band shown as vertical dashed lines, from [Oin00]). The two curves correspond to the Q_{2p} values (i.e., mass of ^{70}Kr) 1.95 MeV (dotted) and 1.33 MeV (solid). (Figure taken from [Cla04].)

The masses of ^{70}Br and ^{71}Kr are also relevant for the pathway of the *rp* process. The above figure shows the main flow that passes via the waiting point at ^{72}Kr ($N = 36$). However, depending on the conditions, branching is possible, involving the beta-decay of ^{70}Br or ^{71}Kr. The flow between these paths is determined by the proton-separation energy of ^{70}Br hence, the masses of ^{70}Br and ^{71}Kr. The mass of ^{70}Br has been measured with an accuracy of about 10 keV at the NSCL LEBIT facility [G. Bollen, private communication]. The present uncertainty for the mass of ^{71}Kr is 650 keV [AME03]. Clearly, this needs to be improved.

3.4 Exotic-decay mass links: $^{111\text{-}113}$Xe

These nuclides are interesting due to their location adjacent to an isolated region of alpha-decay. This region is illustrated in the following diagram of decay links (from [AME03]). Specifically, the case of ^{111}Xe shows links to the proton-emitting ^{112}Cs and to ^{109}Te and to ^{103}Sn via alpha decay. However this group is not connected to any nuclide with a known mass. The measurement of ^{111}Xe would therefore provide the masses of all these exotic nuclides.



The link diagram (from [AME03]) for the region of proton-rich Xe.

The case of ^{111}Xe is made more interesting by the existence of a 900-ms isomeric state whose excitation energy is unknown. ISOLTRAP has the proven capability of dealing with isomeric states (see e.g., [Web05]) and would be able to provide important information e.g., its excitation energy, if it were produced.

Direct measurements of the nuclides $^{112\text{-}113}$Xe would also allow the replacement of many links visible in the above diagram and strengthen the continuity of the mass surface in this region. These nuclides would extend those already measured by ISOLTRAP [Dil06].

4. Beam time request

The beam time request of 42 shifts is detailed in the table below. In all cases we request the newly-developed arc-discharge (VADIS) ion source, with cooled transfer line [Pen08]. From on-line measurements made by the target/ion-source group in 2008, the gain factors for the different species are as follows: argon (13); krypton (9); xenon (4). ISOLTRAP has already made forays into all of these areas of the chart. For argon, ISOLTRAP measured the mass of ^{45}Ar [Bla03] however this primary goal of that experiment was the neutron-deficient isotopes and the target was not appropriate for neutron-rich species. The ISOLDE (PSB) book yield for ^{49}Ar (there is no number for ^{48}Ar) is given as 10/μC. Given the recent improvements of the ISOLTRAP efficiency [Yaz06] combined with the factor-of-twenty gains from the VADIS source [Pen09, Neid09], we feel that ^{48}Ar is feasible. The neutron-deficient kryptons were measured down to ^{72}Kr [Rod06] for which a yield of just over 10^3/s was measured, and neutron-deficient xenons, to ^{114}Xe [Dil04] for which 10^4/s was measured. The same ISOLTRAP and VADIS efficiency improvements lead us to believe that ^{70}Kr and ^{111}Xe are within reach. The neutron-rich kryptons were measured out to ^{95}Kr (book yields are about 10^5/μC) in an experiment that was limited in time [Del06]. With the same arguments, we believe that ^{98}Kr is within reach. While short, the half-lives of these nuclides are still close to what has already been achieved with ISOLTRAP (65 ms) and will indeed offer a good opportunity to probe this limit.

nuclides	half-life (s)	$\delta m/m$	yield (s^{-1})	shifts	target
^{46}Ar	8.4(6)	$5\cdot10^{-8}$	lots	1	U carbide
^{47}Ar	0.580(120)	$< 1\cdot10^{-7}$	60,000	2	U carbide
^{48}Ar	0.5 (est.)	$< 5\cdot10^{-7}$	20,000	4	U carbide
^{72}Kr	17.16(18)	$5\cdot10^{-8}$	10,000	3	Nb foil or Y oxide
^{71}Kr	0.100(3)	$< 1\cdot10^{-7}$	1,000	5	Nb foil or Y oxide
^{70}Kr	0.057(21)	$< 5\cdot10^{-7}$	200	6	Nb foil or Y oxide
^{96}Kr	0.080(7)	$< 1\cdot10^{-7}$	lots	1	U carbide
^{97}Kr	0.063(4)	$< 1\cdot10^{-7}$	100,000	2	U carbide
^{98}Kr	0.046(8)	$< 5\cdot10^{-7}$	20,000	4	U carbide
^{113}Xe	2.74(8)	$< 1\cdot10^{-7}$	20,000	3	La_2O_3 or CeO_x fibers
^{112}Xe	2.7(8)	$< 1\cdot10^{-7}$	2,000	5	La_2O_3 or CeO_x fibers
^{111}Xe	0.740(200)	$< 5\cdot10^{-7}$	400	6	La_2O_3 or CeO_x fibers

List of nuclides proposed for mass measurements with their half-lives, relative precision aimed for, (extrapolated) ISOLDE book yields with the increased VADIS efficiency folded in, number of shifts required and corresponding production target.

The above list represents three separate experiments which we would propose to run over the next two years. The first priority is the measurement of ^{48}Ar, followed by the neutron-rich krypton isotopes that would come from the same target. The next priorities are the (more difficult) neutron-deficient krypton and xenon isotopes.

Priority 1 (U target): 14 shifts
Priority 2 (Nb or YO): 14 shifts
Priority 3 (La_2O_3 or CeO): 14 shifts

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