

Letter of Intent to the ISOLDE and Neutron Time-of-Flight Committee

High precision mass measurements of odd-odd $T=1$ nuclides for the study of the
Isobaric Multiplet Mass Equation

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Abstract

An evaluation of isobaric analogue states, for isospin $T = \frac{1}{2}$ to $T = 3$, has recently been completed and the most complete and precise set of isobaric multiplet mass equation coefficients has been extracted. These coefficients, related to the Coulomb energy component of the nuclear force, are nearly complete up to atomic mass $A = 60$ for $T = 1$ multiplets. There are currently three unmeasured ground state masses, ^{52}Co , ^{56}Cu and ^{60}Ga , that hinder the extraction of the remaining coefficients on the approach to the proton dripline. The measurement of the ground state mass of these $T = 1$ odd-odd nuclides would complete the minimal set of experimental observations. This Letter of Intent describes the physics motivation and possibilities for the measurement the relevant masses with the ISOLTRAP mass spectrometer.

Other key measurements, of ^{44}V and ^{48}Mn , would allow to resolve outstanding issues related to ground state isomer contamination and the currently limited mass precision.

Requests: target development for neutron-deficient Cu , Ga , Co and V beams; 10 shifts with a ZrO_2 felt target and RILIS for tests of Cu and Ga beams;



Motivation

Several odd-odd nuclei have recently been highlighted as being key nuclei in the study of Isobaric Analogue States (IAS) and the Isobaric Multiplet Mass Equation (IMME) [1]. A recent detailed evaluation of isospin $T = 1$ to $T = 3$ nuclei up to $A = 60$, as part of the 2012 Atomic Mass Evaluation [2,3] and NUBASE [4], has established the current best set of isoscalar, isovector and isotensor components of the Coulomb Energy, based on first order perturbation theory and the charge independence hypothesis of the nuclear force. In this study several ground state masses are unmeasured, and this adversely affects the determination of observed excited IAS levels, preventing the determination of the complete set of $T = 1$ multiplet member masses.

The mass measurement of five key nuclides, $^{44gs+m}_{23}V$, $^{48}_{25}Mn$, $^{52gs+m}_{27}Co$, $^{56}_{29}Cu$, $^{60}_{31}Ga$, are essential to extract the Coulomb energy components up to the proton dripline

Mass and Isospin

It was in 1932 that Heisenberg [5] applied the Pauli matrices to the labelling of the two charge states of the nucleon, and 1937 when E.P. Wigner [6] pointed to isospin as being a good quantum number to characterize so-called "isotopic", or isospin, multiplets. Two concepts are important in this description. First, the charge symmetry of the nuclear potential, implying $V_{pp} = V_{nn}$, where V_{pp} (V_{nn}) is the proton-proton (neutron – neutron) potential. Secondly, the hypothesis of charge independence of the nuclear potential, implying $V_{pp} = V_{nn} = V_{pn}$. Consequently, within these hypotheses, any nuclear mass can be described as being the resultant of the charge independent strong forces M_0 plus an additional perturbing Coulomb energy E_C between protons. The mass generated through the proton-neutron configuration will depend on the known proton-neutron mass difference, Δ_{nH} , and the relative proton-neutron configuration described through the isospin quantum number T and its projection T_Z .

Isospin is a quantity associated with ground or excited nuclear states, and for any given nuclide can take the values

$$\left| \frac{N - Z}{2} \right| \leq T \leq \frac{N + Z}{2}$$

where N and Z are the neutron and proton number, respectively. The isospin projection T_Z , a unique value associated with all ground state nuclei, is defined to be

$$T_Z = \sum_{i=1}^A t_{z,i} = \frac{N - Z}{2}$$

where A is the atomic mass number. For any nuclide, the possible isospin configurations are such that $\geq T_Z$. Finally, any multiplet T contains $2T + 1$ members, each member having a unique T_Z value.

Using these definitions and hypotheses any nuclear mass may then be written

$$M(A, T, T_Z) = M_0 + E_C + \Delta_{nH} T_Z$$

In this theoretical mass description, the Coulomb energy E_C due to the proton-proton interaction has yet to be described. Several papers published by W.M. MacDonald [7] regroup experimental observation and theory, and amongst other things, he proposes that a first order perturbation theoretical description of the Coulomb energy component should be sufficient if isospin is a good quantum number.

The Coulomb energy is written

$$E_C(A, T, T_Z) = E_C^{(0)}(A, T) - T_Z E_C^{(1)}(A, T) + [3T_Z^2 - T(T + 1)] E_C^{(2)}(A, T)$$

where $E_C^{(0)}$, $E_C^{(1)}$ and $E_C^{(2)}$ are the isoscalar, isovector and isotensor components of the Coulomb energy, respectively [8]. Injecting this back into the isospin dependent mass description yields the quadratic Isobaric Multiplet Mass Equation (IMME)

$$M(T, T_Z) = a + bT_Z + cT_Z^2$$

and the coefficients, a , b and c are related to the Coulomb energy components through the relations

$$a = M_0 + E_c^{(0)} - T(T+1)E_c^{(2)} \quad b = \Delta_{nH} - E_c^{(1)} \quad c = 3E_c^{(2)}$$

Consequently, the experimental observation of these isospin-dependent mass multiplets gives access to the theoretical Coulomb energy components. If the multiplet masses cannot be adequately described by the quadratic IMME form, it implies that other charge dependent forces are at work, and /or, that first order perturbation theory is not sufficient. This is currently the experimental observation for the $A = 32, T = 2$ quintuplet, where higher order IMME terms are required to describe observations [9].

Measurements with the ISOLTRAP mass spectrometer

Penning-trap spectrometers are ideal tools for studying the masses of isobaric multiplets, due to their very high resolving power and precision. In particular, the ISOLTRAP experiment [10] has already produced a number of results testing the accuracy of the IMME [11-13].

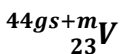
Since the 2008 review of the ISOLTRAP experiment [10], a multi-reflection time-of-flight mass spectrometer (MR-ToF MS) was added to the setup [14]. A recent update of the ISOLTRAP developments, including the MR-ToF MS, is given in [15]. The MR-ToF device can act both as a beam purifier, offering resolving powers of $m/\Delta m \approx 10^5$, and as a mass-measurement tool, first results showing that mass uncertainties lower than 50 keV are possible [16]. Because both modes of operation require trapping times as low as a few tens of milliseconds, the MR-ToF MS allows extending the ISOLTRAP mass-measurement program to more challenging regions of the nuclear chart, bringing within reach nuclides of shorter half-life, higher contamination ratio and lower production rate.

For a Penning-trap mass measurement, a suppression of the contaminants better than three orders of magnitude can be achieved with the MR-ToF MS, in cases where low resolving power is needed. For isotopes in the 100 ms half-life range, a further suppression can be achieved using the standard Penning-trap purification techniques (see [10] and references therein). Considering the typical efficiency of the ISOLTRAP setup ($\approx 1\%$ for light-medium mass nuclides) and a total purification/measurement time of ~ 300 ms (allowing some resolving power also in the Penning traps), a yield of ~ 500 ions per proton pulse gives one count per proton pulse on average for a nuclide of ≈ 100 ms half-life.

For an MR-ToF mass measurement, however, uncertainties lower than 50 keV have been obtained for nuclides with production rates as low as 10 per proton pulse [16]. Unpublished data show that a similar precision is achievable for ~ 30 ms half-life nuclides with similarly low production rate.

Unmeasured ground and isomer masses

In evaluating the complete set of masses for all experimentally observed ground state isobaric analogues using the Atomic Mass Evaluation, the gaps and imperfections of the current experimental situation naturally show up. The long-term goal of this Letter of Intent is to measure the nuclear states summarized in Table 1. The current experimental status for each of these nuclides, and the proposed measurement at ISOLTRAP, is discussed below.



The current difficulty is that the ${}^{44}_{23}V$ ground and ${}^{44m}_{23}V$ isomer states are unresolved. The $T=2$ IAS in ${}^{44}_{23}V$ is well known (± 13 keV), but the excitation energy uncertainty is dominated by the ground state experimental error (± 180 keV).

Table 1

A	E* (keV)	Element	J π	T _{1/2} (ms)	AME2012 Precision (keV)	Estimated Yield (ions per μ C)
44		V	(2+)	111 \pm 7 ^[17]	\pm 180 ^[18]	No estimate
	\sim 270	V ^m	(6+)	150 \pm 3 ^[17]	\pm 180 ^[18]	No estimate
48		Mn	4+	158.1 \pm 2.2	\pm 170 ^[18]	5
52		Co	(6+)	115 \pm 3 ^[17]	-	5
	\sim 380	Co ^m *	(2+)	104 \pm 11	-	5
56		Cu	(4+)	93 \pm 3 ^[19]	-	100
60		Ga	(2+)	70 \pm 10 ^[20]	-	1

The references, in NSR type format, are ^[17] 1997Hagberg04 (measured with TASCC and ISOL at the Chalk River Labs), ^[18] 2004Stadlmann05 (measured at GSI with the FRS-ESR based on TOF measurements), ^[19] 2001Borcea54 and ^[20] 2001Mazzocchi96 (both measured at the GSI On-Line Mass Separator).

The * highlights a hypothetical isomer state, deduced from mirror symmetry, and is discussed further in the text describing the intended $A = 52$ measurement. The estimated yields are discussed in the text.

$^{44}_{23}V$ was previously produced via fragmentation at the GSI FRS-ESR. The aim was to measure the ground $^{44}_{23}V$ and isomer $^{44m}_{23}V$ states. However, with this TOF technique, the mass resolving power of 1.1×10^5 ($FWHM$) was insufficient to clearly separate the ground and isomer state at around 270 keV [18]. The half-lives of both states have been measured [17], based on experimental data taken at the TASCC and ISOL facilities of the Chalk River Laboratories (Ontario, Canada). The determination of the $^{44}_{23}V$ level structure was strongly based on mirror properties, and as such imply (2⁺) and (6⁺) ground and isomer state spin-parities.

In the recent IAS evaluation, the $A=44$, $T=1$ multiplet returns a highly doubtful negative isotensor coefficient. This is an exception, the other comparable ($4n$) multiplets ranging from 130 to 160 keV. This unusual result is most probably the consequence of the isomer contaminated ground state mass measurement which inflates the uncertainty to ± 180 keV. To resolve this case it is necessary to identify and measure the ground and isomer state masses.

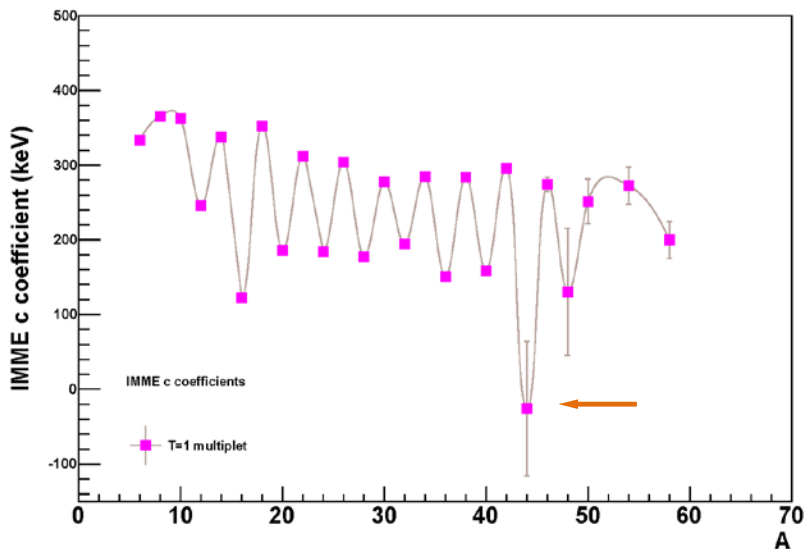
In the $A=44$, $T=2$ multiplet the IAS in $^{44}_{23}V$ has been measured via p-decay to have a mass excess of -21124 ± 13 keV [21]. However, in order to establish the excitation energy in $^{44}_{23}V$ to a similar precision, the ground state mass must be known to a similar, or higher, precision. The current IAS excitation energy is an order of magnitude greater, the level currently being placed at -2990 ± 180 keV.

The $^{44}_{23}V$ (2⁺) ground state has a half-life of 111 \pm 7 ms, and the (6⁺) isomer state 150 \pm 3 ms and so although an MR-ToF type measurement would be a first step forward, a better selectivity is achievable with using in-source laser ionization to separate out the (2⁺) and (6⁺) hyperfine structure.

A Penning-trap mass measurement with a mass resolving power of 10^5 for 100 ms excitation would lead to a ± 5 keV precision, leading to an overall ± 15 keV precision on the excited level.

Vanadium has an extremely low vapor pressure and is not released in thick ISOL targets, even at high temperature. As such, ISOLDE has no yields for any V isotopes. However, research at Oak Ridge has shown that ^{48}V ($T_{1/2} = 16$ days) can be produced by chlorination in a TiO_2 target [22]. A plasma source was used, which also destroyed the molecules, delivering ionic vanadium through the separator. This would

be a promising start for similar work at ISOLDE to produce V beams.



$T = 1$ IMME c -coefficient, related to the isotensor component of the Coulomb energy. The unusual, negative-valued c -coefficient for the current $A = 44$ clearly stands apart. This unusual result would be clarified by a high precision mass measurement allowing the separation of the isomer and ground state masses.

${}^{48}_{25}\text{Mn}$

${}^{48}_{25}\text{Mn}$ was produced via fragmentation at the GSI FRS-ESR, and the TOF mass excess measurement was also published in [18] as -29320 ± 110 keV and adjusted to ± 170 keV in the AME.

In AME2012, series of measurements stemming from the same experimental installation and technique are regrouped and studied independently. A Partial Consistency Factor (PCF), based on a normalized Chi-squared distribution of the experimental precision, is calculated for the series of measurements; a perfect agreement in datasets returns a PCF close to a value of 1, as is generally the case for Penning-Trap measurements [2, p.1311]. The analysis of 67 independent experimental mass measurements carried out at the FRS-ESR highlights 15 cases, ${}^{48}_{25}\text{Mn}$ being one of them, where the experimental precision has not been superseded by higher precision datasets and so remain the sole data references. Today, a multiplicative factor of 1.5, applied to the experimental uncertainties, is required to reconcile the different datasets and consequently the original published error of ± 110 keV has been adjusted to ± 170 keV.

The $T=2$ IAS in ${}^{48}_{25}\text{Mn}$ has been measured via proton-decay, to have a mass excess of -26259 ± 14 keV, which leads to a less precise excitation energy of 3060 ± 170 keV. The half-life of the 4^+ ground state is well known, at 158.1 ± 2.2 ms, and has also been produced previously at ISOLDE/PSB with a RILIS yield of 5 ions per μC using a Nb foil target [23]. This case is presently feasible with the MR-ToF.

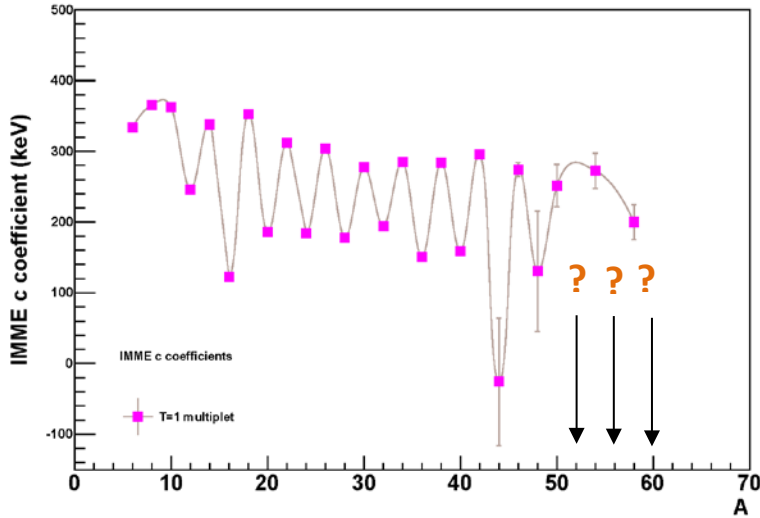
Although the ${}^{44}_{23}\text{V}$ ground and isomer state measurements would have a greater immediate scientific impact, the ${}^{48}_{25}\text{Mn}$ ground state, which presents a similar half-life, would be an excellent candidate for a more straightforward first measurement and a 10-20 keV precision on the IAS level is feasible.

${}^{52}_{27}\text{Co}$, ${}^{56}_{29}\text{Cu}$ and ${}^{60}_{31}\text{Ga}$

These three odd-odd 4-nucleon systems are the remaining nuclides required to complete the IAS $T=1$ multiplets up to $A=60$. The mirror nuclei and excited IAS states are all measured with keV and sub-keV precision. All of them have been previously produced in fusion-evaporation reaction, and half-lives have been measured. A high precision mass measurement of these elusive nuclei would allow the extraction of the IMME coefficients on the approach to the proton dripline.

$^{52}_{27}\text{Co}$

Similar to the previous cases, the $T=2$ IAS mass excess has been measured to ± 13 keV via proton decay, but the missing ground state, currently estimated to ± 200 keV is propagated to the excitation energy of the state. A high precision ground state measurement would simultaneously resolve the remaining $T=1$ issue and allow an improved determination of the $T=2$ IMME coefficients.



The question marks and arrows indicate the position of unmeasured $T = 1$ ground state masses and the rough value for the IMME c -coefficient.

This nuclide was first observed at GANIL, but the first decay information was obtained at the Chalk River Laboratories [17]. The experimental decay intensities suggest a probable isomeric (2^+) state, estimated to lie approximately 380 keV above the (6^+) ground state. This situation is similar to the $^{44}_{23}\text{V}$ case, and can be understood through symmetry argument; $^{44}_{23}\text{V}$ has four particles in the $f_{7/2}$ shell whereas $^{52}_{27}\text{Co}$ has four holes, the isomeric states are also expected to have similar excitation energies and spin-parities through mirror properties.

Co is a difficult ISOL beam but has been produced at ISOLDE using a special ZrO_2 felt target [24], combined with plasma ionization. The reported yield for ^{55}Co ($T_{1/2} = 17.5$ hours) is 57000 ions per μC . The long release time will severely reduce the more exotic cases. We estimate for ^{54}Co ($T_{1/2} = 193$ ms) this could drop to 500 ions per μC ; to 50 ions per μC for ^{53}Co ($T_{1/2} = 242$ ms) to 5 ions per μC for ^{52}Co ($T_{1/2} = 115$ ms). The expected isomer in ^{52}Co ($T_{1/2} = 104$ ms) would have spin 2^+ and might be produced more in comparison with the 6^+ ground state. These cases are on the limit of what is feasible with the MR-ToF. In 2011 neutron-deficient Co was produced using a Y_2O_3 target and a hot-plasma ion source (GPS, target #446), giving good yields of the long-lived isotopes ($\sim 3 \times 10^7$ ions/ μC of ^{57}Co) [25]. The more recent VADIS version of the plasma source [26] could produce a substantial enhancement of the ionization efficiency.

$^{56}_{29}\text{Cu}$

The decay properties of $^{56}_{29}\text{Cu}$ have been measured [19] and the suggested spin-parity of (4^+) is associated with a half-life of 93 ± 3 ms. The ground state mass is as yet unmeasured, even though the $T=2$ IAS has been established via the proton decay of the IAS $^{56}\text{Cu}^i$ to ^{55}Ni to have a mass excess of -35120 ± 30 keV [21]. The excitation energy of the level is currently estimated to ± 200 keV.

A high precision mass measurement is required, not only to complete the $T=1$ multiplet and extract the Coulomb Energy component, but also to pin down the excitation level of the $T=2$ IAS. An MR-ToF measurement would provide a precision of ± 50 keV leading to an overall $T=1$ excitation level precise to ± 58 keV.

The ZrO_2 felt target has been used to produce at ISOLDE 3000 ions per μC of ^{57}Cu ($T_{1/2} = 196$ ms) with RILIS. ^{56}Cu ($T_{1/2} = 93$ ms) may be expected at a rate of 100 per μC , which would be feasible with the MR-ToF and also with the Penning trap. Still, during the 2010 HRS run on the ZrO_2 target #424 (Ta surface), the RILIS yield of ^{58}Cu was estimated at $\sim 6 \times 10^4$ ions/ μC and no ^{57}Cu was observed.

$^{60}_{31}\text{Ga}$

Has been produced and decay properties measured at GSI [20] using the fusion-evaporation reaction $^{28}\text{Si}(^{36}\text{Ar}, p3n)$ and the spectroscopy of β -delayed γ -rays. The half-life of the (2^+) ground state has been measured to be 70 ± 10 ms.

The particularity of this nuclide is that it is most probably on the proton dripline (defined by a proton separation energy $S_p = 0$ keV). The experimental measurement [20] suggests $S_p = 40 \pm 70$ keV, whereas the AME2012 estimation, based on trends in the mass surface, suggests $S_p = -140 \pm 200$ keV.

The ISOLDE yield book value for ^{61}Ga ($T_{1/2} = 170$ ms) is only 10 per μC (^{62}Ga , with $T_{1/2} = 116$ ms, is 200 ions per μC), using W surface ionization and the ZrO_2 felt target. Extrapolating to ^{60}Ga ($T_{1/2} = 70$ ms), we would not expect more than 1 ion per second, which is marginal for a MR-ToF mass measurement. A gain of a factor of four using RILIS would greatly help. Neutron-deficient Ga was produced in 2010 using a ZrO_2 target (HRS, target #426, Ta surface) with RILIS, yielding $\sim 7 \times 10^5$ ions/ μC of ^{63}Ga and $\sim 7 \times 10^2$ ions/ μC of ^{62}Ga . As in the case of Co , the slow release of Ga is a major impediment.

Requested developments and shifts

The beams of neutron-deficient Cu , Ga , Co and V are of long-term interest for the study of the IMME along the chain of $T = 1$ isobaric multiplets and we would like to request their development at ISOLDE. Based on immediate feasibility and physics impact, we would like to prioritize in a first stage the development of the beams of ^{56}Cu and ^{60}Ga and request 10 shifts (5+5) on a ZrO_2 felt target with RILIS to test the yields of neutron-deficient copper and gallium isotopes approaching ^{56}Cu and ^{60}Ga . However, the Letter of Intent is open to alternative target solutions which would provide the required yields of the isotopes of interest. As shown in recent publications [14-16], the MR-ToF MS is a very powerful tool for determining the yields and release properties of rare isotopes, as well as for the on-line optimization of their production parameters (^{54}Ca being a notable highlight), so it would be directly used for the feasibility tests. The MR-ToF MS also being one possible method for determining the masses of the isotopes of interest, the requested shifts would allow to assess not only the yield, but also the isobaric contamination of the beam and its impact on the measurement strategy.

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