EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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High-Precision Mass Measurements in the Rare-Earth Region to Investigate the Proton-Neutron Interaction

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Abstract

We propose precision mass measurements on a series of short-lived nuclides for the investigation of proton-neutron interactions and two neutron separation energies and their role in nuclear structure, especially concerning deformation and collectivity. The nuclides include neutron-rich Nd, Sm, Dy, Er, and Yb isotopes, as well as ¹⁸⁶Hf. The investigations will require the Penning trap mass-spectrometer ISOLTRAP which routinely reaches the necessary mass precision of about 10 keV. The measurements are planned for a period of two years and include a request for the test of the RILIS scheme of Sm.

1. Introduction and motivation

Looking at the number of valence nucleons in a nucleus is a key first and helpful step to get an idea about its structure. For even-even nuclei, the $R_{4/2}$ ratio, which is the energy of the first 4^+ excited level over the energy of the first 2^+ level, is often considered as a critical next step. The quantity $1/E(2_1^+)$ is also a useful observable which reveals the same trend as $R_{4/2}$ as a function of Z and N but which is often available experimentally in exotic nuclei when $R_{4/2}$ is not. In medium- and heavy-mass regions, particularly in deformed nuclei, not only $R_{4/2}$ but also such quantities as the beta-band states, gamma-band states, and certain $B(E2:J_1 \rightarrow J_f)$ values are naturally necessary to fully define the structure of the nucleus.

Mass measurements provide complementary information, especially nowadays, with Penning traps and storage rings, which provide high accuracy mass values for short-lived and stable nuclides [1]. Moreover, often masses can be obtained when spectroscopic data cannot. Therefore, atomic mass measurements can sometimes in fact give us the first information about nuclei. Recently it has been shown that masses are much more sensitively related to structure than previously thought [2], especially in deformed nuclei. Thus mass measurements can complement spectroscopic observables in determining the structure of nuclei and their evolution with N and Z. This will be particularly important in nuclei far from stability where other data are sparse. To prepare for this and to study how to exploit this enhanced mass-structure sensitivity also requires mass measurements in nuclei where some spectroscopic data is already known. Note, though, that, in many cases, one can obtain the mass value for a given nucleus even if $R_{4/2}$ is unknown but $R_{4/2}$ can seldom be obtained when the mass is unknown.

Atomic masses allow extracting nuclear binding energies which reflect all nucleonic interactions inside the nucleus. The binding energy is given by the difference of the summed masses of all protons and neutrons inside the nucleus and the actual mass of the nucleus:

$$B(Z,N) = (Z \cdot m_p + N \cdot m_n - M(Z,N)) c^2$$
 (1)

One nucleon (proton or neutron) or two nucleon separation energies can be extracted from the binding energies. Equation (2) shows one neutron and two neutron separation energies, respectively.

$$S_n(Z,N) = B(Z,N) - B(Z,N-1)$$
; $S_{2n}(Z,N) = B(Z,N) - B(Z,N-2)$ (2)

These two observables reveal considerable information about the structure. There are several areas of the nuclear chart where new information on masses can help understand anomalous or puzzling behavior of either S_{2n} / S_n or δV_{pn} values [3,4], which are empirical measures of proton-neutron interaction strengths and will be discussed below (see Eq. 3). These mass-related quantities can thereby help understand the evolution of structure in these regions. Some proposed masses are presented briefly below:

Flattening of S_{2n} values around Z = 70 and N = 108

In terms of the separation energies, an obvious example (except magic shells) of structural change is given for nuclei at critical points of quantum phase transitions. As is well known, the Nd, Sm, Gd and Dy nuclei at N = 90 undergo a shape transition [5]. The onset of deformation at N = 90 for these nuclei gives increased binding energies which is seen in a flattening in S_{2n} . This flattening is clear for the rare-earth nuclei in Fig. 1 compared to a linear decrease in S_{2n} for neighboring isotopic chains.

Interestingly, a similar flattening in S_{2n} seems to start in the Yb chain at N = 108, which might be connected to the subshell closure or other structural changes mentioned in Ref. [7]. Unfortunately there are no data for Yb beyond this point, and masses for lighter rare-earth

elements beyond N = 108 are also missing. In order to investigate this corner of the nuclear chart, masses of Yb above N = 107, Er with N > 104, Dy with N > 102 will be required. These are summarized in Table 1.

δV_{pn} values for the rare-earth nuclides

Masses can be further exploited by isolating specific interactions using double differences of masses. One of the most important is a specific double difference of binding energies which gives the average interaction between the last two protons and the last two neutrons, δV_{pn} [3,4]. Since the interactions of the protons and neutrons largely determine the character of the nucleus (in competition with pairing), the role of the proton-neutron (p-n) interaction is extremely important in understanding the evolution of both single particle and collective structure: Magic numbers, single particle energies, collectivity, phase/shape transitions [8-10].

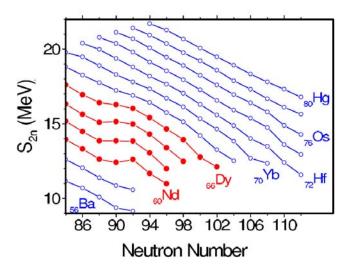


Fig. 1 The S_{2n} values for even-even nuclides as function of neutron number for the $_{56}$ Ba - $_{80}$ Hg nuclei between N = 84 and N = 112 [6]. Isotopic chains showing deformation related to the phase transition around N = 90 are marked with solid circles.

Specifically, for the even-even nuclei, δV_{pn} is the difference of two-neutron (or two proton) separation energies for consecutively numbered even nuclei with Z and Z-2 for the same neutron number (or the difference between two neutron separation between nuclei with N and N-2 for the same proton number):

$$|\delta V_{pn}(Z,N)| = 1/4 \left[\{B(Z,N) - B(Z,N-2)\} - \{B(Z-2,N) - B(Z-2,N-2)\} \right]$$
(3)
= 1/4 \[\(S_{2n}(Z,N) - S_{2n}(Z-2,N) \]
= 1/4 \[\(S_{2p}(Z,N) - S_{2p}(Z,N-2) \]

 δV_{pn} can sometimes be simply interpreted in terms of the orbit occupations of the last two protons and neutrons, by considering of the spatial overlap between the proton and neutron wave functions [3,4]. In other cases one must resort to detailed microscopic calculations such as those provided by Density Functional Theory (DFT) – see below.

The standard interpretation of p-n interactions is that they reflect the overlaps of the wave functions of the outermost protons and neutrons, and that these should be largest when the proton and neutron shells are filled to roughly the same fractional extent. This is verified in many nuclei by large values of δV_{pn} along the diagonal joining nuclides with consecutive magic N and Z (see Fig. 2). Inspection of Fig. 2 show an anomalously low δV_{pn} value for ¹⁵⁸Sm (blue box at N = 96) which seems to contradict the idea of large values along the diagonal [4,11]. However, this unexpected δV_{pn} value has a large uncertainty.

We therefore propose to measure the masses of 158 Sm, 154,156 Nd with higher accuracy to extract the δV_{pn} value at 158 Sm, and to extend the systematics near the diagonal to see if the drops in δV_{pn} seen in Fig. 2 are also present in heavier isotopes. This investigation is critical in order to determine the detailed mechanisms underlying the p-n interactions that drive the development of collectivity in these nuclei, specifically to see if they can indeed be simply understood in terms of spatial overlaps or whether other effects are also important. The nuclides for which the δV_{pn} values should be determined include neutron-rich rare-earth isotopes, as well as Yb isotopes with N > 104, Hf isotopes with N > 108, W above N > 112.

These new δV_{pn} values will also be compared with new DFT predictions which have themselves already been triggered by our recent measurements in the Rn and Xe nuclei. These comparisons will provide sensitive tests of the p-n correlations in these calculations. It has also been argued that one might even be able to help evaluate the merits of different interactions in these calculations by comparison to new masses. For example, the recent Xe masses [12] confirmed DFT calculations carried out with the SKPDMIX interaction [12].

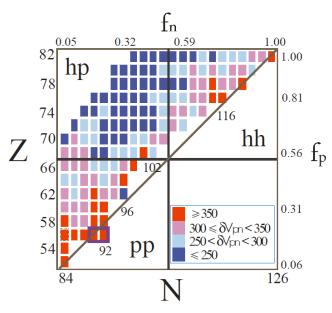


Fig. 2 Empirical δV_{pn} values at the major shells Z=50-82, N=82-126 with proton and neutron fractional filling values (fp and fn, respectively) [4,11]. Indicated δV_{pn} values at N=84, 90 and 92 for the Ba nuclei are based on recent Xe mass measurements at ISOLTRAP [12].

2. Experimental setup

The presented mass measurements, especially for the $\delta V_{\rm pn}$ study, need to be performed with an uncertainty below 10 keV, which corresponds to a relative uncertainty below $\delta m/m=10^{-7}$ in the heavy mass region around A=200. The required precision has been routinely reached by Penning trap mass spectrometers like ISOLTRAP [13].

With ISOLTRAP over 400 exotic nuclides have already been investigated, meanwhile reaching regularly a relative mass precision of the order of $\delta m/m=1\cdot 10^{-8}$ [14]. Nuclides with production rates as low as 100 ions/s and half-lives well below 100 ms can be addressed. Last year ISOLTRAP already reached a number of highlights within the previous proposal devoted to the proton-neutron interaction and δV_{pn} values, P-230 [15]. During a single beamtime neutron-rich $^{136-146}$ Xe and $^{223-229}$ Rn were studied, with 11 out of 18 masses determined directly for the first time. In addition a new nuclide, 229 Rn, was identified [12], a feature which had not been achieved at a Penning trap system before.

ISOLTRAP employs three ion traps for the preparation and purification of radioactive ion bunches delivered by ISOLDE, as well as for the measurement of the atomic mass. The 60-

keV ISOLDE ion beam is first stopped, cooled, and bunched in a linear radio-frequency quadrupole (RFQ). Isobaric contaminants which are still present after mass selection with either the GPS or the HRS separator magnets, can be removed in the first, cylindrically shaped Penning trap, which can reach resolving powers of up to $m/\Delta m=10^5$. The isobarically pure ion bunch is then transferred to the precision Penning trap, where possible isomeric ions can be removed using a resonant dipolar rf-excitation.

The mass measurement is based on the very precise determination of the cyclotron frequency $v_c=qB/(2\pi m)$ of ions with mass m and charge q stored in a strong and homogeneous magnetic field B. With the time-of-flight cyclotron-resonance technique the gain of radial energy in the Penning trap after a quadrupolar rf-excitation is probed by monitoring the change of the time of flight towards an external detector after axial ejection of the ions from the trap. The magnetic field strength is determined by measuring the cyclotron frequency of a reference ion with well-known mass. The mass of the ion of interest is finally deduced from the ratio of the two cyclotron frequencies.

Table 1: Nuclides of interest to investigate both the two neutron separation energy and proton-neutron interaction.

	Nuclide(s) of interest	Nuclide to study	Half life T _{1/2}	Mass uncert. /keV
S_{2n}	¹⁷⁰ Dy	¹⁷⁰ Dy*		200 #
	¹⁷⁴ Er	¹⁷⁴ Er	3.2 min.	300 #
	¹⁷⁸ Yb	¹⁷⁸ Yb	74 min.	10
	¹⁸⁰ Yb	¹⁸⁰ Yb	2.4 min.	400 #
	¹⁸² Yb	¹⁸² Yb *		400 #
	¹⁸⁶ Hf	¹⁸⁶ Hf	2.6 min.	300 #
$\delta m V_{pn}$	158 Sm	^{154}Nd	25.9 sec.	110
	¹⁵⁸ Sm	¹⁵⁶ Nd	5.49 sec.	200
	¹⁵⁸ Sm	¹⁵⁸ Sm	5.30 min.	80
	¹⁷² Er	170Dv *		200 #
	¹⁷⁶ Yb	¹⁷⁴ Er	3.2 min.	300#
	¹⁸² Hf	¹⁸⁰ Yb	2.4 min.	400 #
	$^{188}\mathrm{W}$	¹⁸⁶ Hf	2.6 min.	300 #

^{*} nuclides which have not yet been identified;

ISOLTRAP performance

The overall ISOLTRAP efficiency is in percent range for longer-lived nuclides, and in per mill range for nuclides with half-lives below 1 s, where decay losses during ion manipulation in the system are the limiting factor. Most losses are due to a limited acceptance of the ISOLTRAP buncher, which ranges from 1-10% [13].

The time required for excitations in the traps causes decay losses which influence the shortest half-life of the investigated ions. The measurement cycle lasts usually about 1.5s, but it can be lowered (at cost of lower precision) for more exotic ions. So far, the shortest-lived nuclides investigated at ISOLTRAP were ⁶⁶Mn and ⁷⁴Rb [16] with half-lives of 64 ms and 65 ms, respectively.

ISOLTRAP has single-ion sensitivity, therefore the minimum ISOLDE yield depends only on overall efficiency and decay losses during measurements. Assuming negligible losses and 1% efficiency, ions with production yields as low as 100 ions/s can be and have been addressed.

Precision achievable at ISOLTRAP is presently in most cases limited by systematic uncertainties. Using mass measurements with carbon clusters [14] a residual systematic uncertainty has been determined to be 8x10⁻⁹, to which a relative mass-dependent uncertainty

[#] extrapolated from systematics [6]

has to be added [14] equal to 1.6×10^{-10} *(m-m_{ref}), where m is the mass of the reference ion. This allows to determine atomic masses below 10 keV precision, even for masses around A=200.

3. Overview on proposed beams

The nuclides proposed for mass-determination are presented in Table 1 together with their half-lives. Information on their availability and production at ISOLDE is given below:

Rare earths: Nd, Sm, Dy, Er, Yb

About 10 years ago ISOLTRAP performed first very successful high-precision mass measurements in the rare-earth region, at that time on the neutron-deficient side [17]. These beams have been available at ISOLDE using surface ionization, which guarantees high efficiency. However, this method suffers from large isobaric contamination, due to chemical proximity of neighboring elements. Thus, while neutron-rich rare-earth masses close to stability can be addressed with the standard surface ion source, nuclides far from stability need probably RILIS in order to reduce the large amount of contaminations.

Pure beams of ^{154,156}Nd, and ¹⁵⁸Sm were already requested in a previous ISOLTRAP proposal, P-230 [15]. Following the request the INTC committee recommended Nd and Sm beams for development (with lower priority). The present beam request includes the results from recent ion-source development on Nd beams which took place since that time.

In general, the development comprises a combination of a selective laser ionization and low work-function cavities suppressing surface ionization. RILIS schemes are already known for Nd, Sm, Dy, and Yb. In the case of Nd the ionization scheme was tested in 2009 with a low work-function cavity and first results were obtained. The suppression of surface ionized contamination was not as large as expected, however, suitable for the ISOLTRAP mass spectrometer which is equipped with a purification Penning trap with a high mass resolving power.

ISOLDE production yields for the elements of interest are available in the SC database and include isotopes closer to stability than those requested. The extrapolation is not easy, since we will use PSB. Concerning half-lives the requested nuclides should be easily within reach, because most of them are in second- and even minute-range. For three of the listed nuclides the half-live is not known. However, their half-lives are estimated to be above 500 ms [6], and ISOLTRAP has recently shown the potential to even discover unknown nuclides (²²⁹Rn) [12].

186 Hf

Hafnium yields in the ISOLDE data base come only from a Ta target (SC not PSB), but they show yields as high as $3x10^6$ for 180 Hf. At PSB-ISOLDE $^{177,179-184}$ Hf were investigated by the NICOLE experiment, using a hot plasma Ta/W/Ir target with a CF₄ gas leak in order to release the beams as a tri-fluoride. Previously also 185 Hf was observed. However, no yields were given. For a more neutron-rich 186 Hf a target consisting of Ir foils might be the optimal choice.

4. Beam time request

Table 2 gives a summary of the requested beams, to be investigated over a two year period, including target and ion-source development for a Sm beam.

In the first part we list beams already available at ISOLDE (Hf and Nd) with an estimated number of required shifts, together with a target and ion source combination which was already successfully used to deliver these elements.

The second part includes rare-earth beams, for which target material and surface ionization is expected to give enough yield for mass measurements (about 1000 ions/ μ C needed). Possible isobaric contamination will be removed using the purification Penning trap of ISOLTRAP. The shift estimate includes the time to set-up the cleaning procedure in order to remove the isobars. It is possible to combine the mass measurements for the different elements and to use the same target/ion-source unit.

The third part aims at the test of a Sm RILIS scheme which is planned for 2010. No shifts are requested at this time.

<u>Table 2</u>: Overview of nuclides requested in this proposal, to be investigated over a period of two years.

A	Element	Shifts	Target	Ion source			
Beams available at ISOLDE: 7 shifts requested							
186	Hf	3	Ir +CF ₄	Hot plasma			
154-155	Nd	4	UCx + low work-	RILIS			
			function cavity				
Rare-earth beams with possible contamination which can be separated with							
ISOLTRAP purification trap: 8 shifts							
168, 170	Dy	3	Ta	W (surface)			
174	Er	2	Та	W (surface)			
178, 180	Yb	3	Ta/W mixture	W (surface)			
Rare-earth beams requiring target and ion source tests							
158-161	Sm		UCx + low work-	RILIS*			
			function cavity				

^{*} RILIS scheme to be tested during 2010

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