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Mass measurements of the shortest-lived nuclides *à la MISTRAL*

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Abstract

At Princeton in the 1960's, L.G. Smith invented an instrument of astonishing accuracy and rapid measurement time, derived from his so-called mass synchrometer. Using the same principle, a radiofrequency spectrometer was constructed in Orsay to measure masses of the shortest-lived nuclides at CERN's ISOLDE facility. Smith's spectrometer is now a museum piece, making the Orsay version (since baptized, MISTRAL) the sole example of such an instrument and the only one ever to be used on-line. Here we report on a measurement of the 65 ms half-life, $N = Z$ nuclide ^{74}Rb performed with MISTRAL. The measured mass excess of −51944 (117) keV is compared with that obtained by ISOLTRAP, since independent measurements using different techniques assure a healthy gene pool for the recommended masses of the atomic mass evaluation. The nuclide ^{74}Rb is the heaviest for which a precise mass is of importance for the so-called Wigner energy. A discussion is presented concerning this Wigner energy, perhaps the last component of nuclear mass formulas resisting microscopic treatment.

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Keywords: Atomic masses; Nuclear binding energy; RF mass spectrometer; N=Z nuclides; Wigner energy

1. Introduction

Just as the observation of a distant star gives us detailed information about its properties, the weighing of an atomic nucleus reveals a surprising amount of information concerning its structure. This is due to the nuclear binding energy, the difference of its mass and the sum of its constituent masses, a manifestation *par excellence* of Einstein's famous relation: $E = mc^2$.

Mass measurements have a rich history, starting from the work of Francis Aston at Cambridge in the 1920's [1]. His systematic measurements of unprecedented experimental accuracy revealed the nuclear binding energy and resolved the apparent problem of non-integer atomic weights leading to the idea of isotopes. Aston's (more well known) contemporary, Arthur Eddington, developed a stellar model [2] in which the nuclear binding energy (as opposed to the chemical or gravitational energy sources propounded at the time) powered the heat output of the sun, solving the problem of the Sun's (and the Earth's) age. Thus the link between nuclear masses and astrophysics is not simply metaphorical and indeed, goes back to the beginning of the field.

Determination of the binding energy of an exotic nuclide is one of the most demanding experimental challenges of research in nuclear structure, given the sensitivity and, most important, the high precision required. The use of mass spectrometry for this task at accelerator facilities was initiated in the early 1970's by the CSNSM-Orsay group [4] and has since proved to be an extremely powerful probe of nuclear structure, spawning a rich heritage. Of the many experimental techniques now dedicated to mass measurements of radioactive nuclides, the Penning trap has emerged as the superlative tool, exemplary for its resolving power, accuracy, sensitivity and versatility (for which other papers in this volume will testify).

While ISOLTRAP at CERN continues in its role as the pioneering on-line Penning trap spectrometer (since the mid 1980's), several new Penning-trap-based installations have since come into operation: SMILETRAP in Stockholm (for stable, highly charged species), CPT in Argonne, SHIPTRAP in Darmstadt, JYFLTRAP in Jyväskylä, LEBIT in East Lansing, and soon, MLLTRAP in Munich, HITRAP in Darmstadt, and TITAN in Vancouver.

The small trapping volume that minimizes field inhomogeneities and associated ion cooling techniques that allow well-defined initial conditions, both contribute to the high accuracy of the Penning trap technique but the key ingredient is the

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long observation time that allows the resolution of even isomeric states. However, since nuclear half-lives only grow shorter as we approach the extremities of the nuclear chart, it is fair to wonder at what point the act of storing a radioactive ion is incompatible with its decay half-life. This paper describes an alternate technique – that used by the radiofrequency, transmission spectrometer MISTRAL – especially adapted for short-lived nuclides.

A review of mass measurement programs and their associated techniques is found in [5]. These many programs are essential for the mass evaluation, the necessary process for creating the mass table ([3] and G. Audi, this volume). Different techniques with independent systematic errors and different ways of obtaining mass data to form over-determined systems, ensure a mass table of the best possible accuracy.

We also report a MISTRAL mass measurement of the $N = Z$ dripline nuclide ^{74}Rb, which has a half-life of only 65 ms. It is always important to verify any technique and compare the results. In this case we profit from the spectacular feat of ISOLTRAP having also measured the mass of this nuclide (thought by many to be impossible with a trap). $N = Z$ nuclides exhibit an extra binding energy compared to the general tendency of the mass surface, the so-called Wigner effect. The result is discussed in light of systematics for such nuclides. The family of superallowed β-emitters, of which ^{74}Rb is one, is of particular interest for tests of fundamental interactions (see, for example [6]) but not discussed here.

2. MISTRAL description

MISTRAL has been described at length in other publications: [7–15] and dissertations: [17–20]. Here, only the salient points are discussed, stressing the complementarity with ISOLTRAP, when appropriate.

MISTRAL is based on the radiofrequency (RF) transmission spectrometer developed by Smith at Princeton in the early 1960's [21,22]. The idea is to determine the mass m from the cyclotron frequency f of a charged ion q in a homogeneous magnetic field B from $f = qB/2\pi m$. As illustrated in Fig. 1 (top left), the ion trajectory in the magnetic field is mechanically defined by four small slits (of 0.4-mm width and 5-mm height). As such, a mass resolving power of 2500 is obtained. In order to reach resolving powers necessary for precision mass measurements, a longitudinal acceleration is performed after one half turn in the magnetic field (Fig. 1 top right). After a further turn (one cyclotron orbit) another acceleration occurs (Fig. 1 middle left) and if the net kinetic energy remains unchanged, the ion is transmitted through the final slit (Fig. 1 middle right). The acceleration is performed by a narrow-gap (0.5 mm), RF modulator and whether the net change in kinetic energy between the two modulations is zero or not, will depend on the phase relation between the RF and integer-plus-one-half multiples of the cyclotron frequency: $f_{\mathrm{RF}} = (n + 1/2)f$. Fig. 1 (bottom) shows the transmitted ion signal versus a scan of the RF over two multiples of f (with $n \approx 1000$). The resolving power is inversely proportional to the slit width and proportional to n and the amplitude of the modulation. Therefore, low RF-power modulation results in modest resolving power (about 20,000 in Fig. 1 for the higher signal) and higher resolving power requires higher RF power. The operating bandwidth of the MISTRAL modulator is 250–500 MHz and its matching is not trivial (see e.g., [13,20]).

Like ISOLTRAP, MISTRAL requires the moderate transport energy and high-quality beam optics provided by an ISOL facility (both experiments are in fact located at CERN's ISOLDE facility — see [11]). The layout of MISTRAL is shown in Fig. 2. Inset shows the front and side views of the electromagnet and support structure.

The ISOLDE beam or reference ion beam is electrostatically transported to the entrance slit. After the modulation gymnastics described above, the beam is extracted and electrostatically transported to a secondary-electron multiplier for counting. Measurements are performed for a fixed magnetic field, set for the ISOLDE ion at (nominally) 60 keV. Reference ions are injected every 30 s or so, by switching all the electrostatic transport voltages in inverse proportion to their mass with respect to that being measured. This fast switching helps reduced uncertainties brought by long-term field drift. The measurement error will therefore depend on short-term field fluctuations, statistics and resolving power, and systematic effects related to calibration. A detailed account of these uncertainties is given in [15].

Often with direct mass determinations, there is a systematic shift that increases with the difference between the mass being measured and the reference mass, usually due to their differing velocities in the magnetic field. In ISOLTRAP's case, this shift has been evaluated at about 10^{-9} per mass unit [29]. In MISTRAL's case, the same shift is about 100 times larger (and has varied from one experiment to another [15]). The main reasons for the superior trap performance are (1) the fact that ions are cooled, establishing the same conditions for both species and (2) the higher and more homogeneous superconducting field, of smaller volume (roughly 1 cm^3). The magnetic field sampled by ions traversing MISTRAL, though delimited by four 0.4 mm-by 5 mm-slits, is roughly 100 times larger.

In an off-line measurement, a mass peak of 32 frequency channels (e.g., at 10 ms accumulation per channel) would take simply 320 ms with MISTRAL. The equivalent scan with ISOLTRAP, with equivalent resolving power (e.g., 80,000) would take approximately 20 times longer, due to the preparation, cooling, and excitation required for each step. (The time of flight is negligable for both instruments.) This gain in measurement time does not apply to on-line measurements, however, since one frequency step is measured for each PS-Booster proton pulse, at a maximum of every 1.2 s.

In its commissioning phase, MISTRAL measured several masses of light nuclides of fairly short half lives and in many cases greatly reduced the uncertainty e.g., ^{30}Na [12], ^{33}Mg [14,15], and more recently ^{11}Li [16]. Here we present a measurement performed on ^{74}Rb [19].

3. The ^{74}Rb measurement

Proton-rich Rb isotopes were produced by the ISOLDE facility (see [23]) from 1.4 GeV proton spallation of a niobium-foil target. The surface-ionized 60-keV beam was then mass-separated using the HRS separator and transported to MIS-

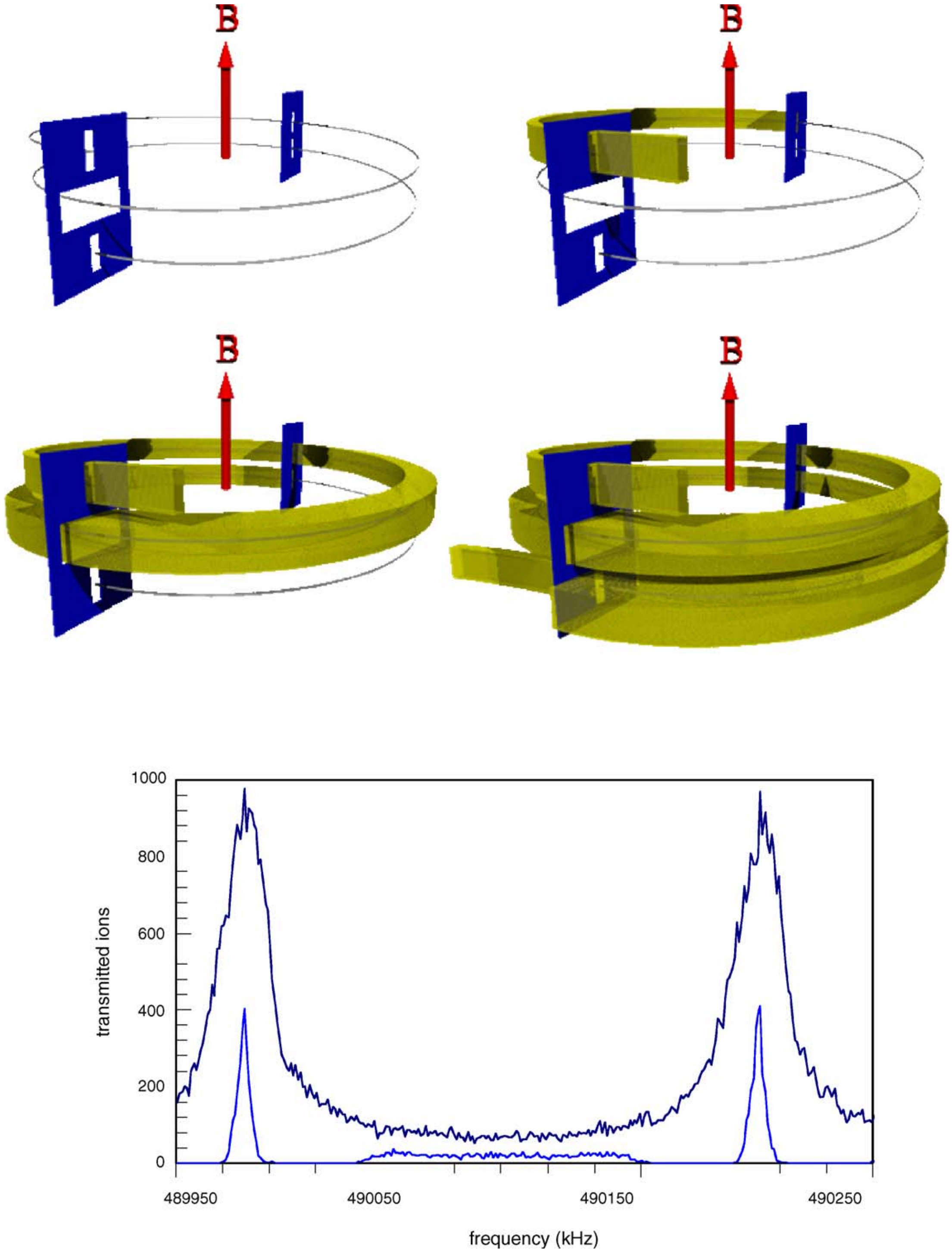

Fig. 1. (top left) an isometric view of the trajectory envelope with the 0.4 mm injection slit followed by the first modulator after one-half turn (top right), an opening to accommodate the modulated ion trajectories after one turn (middle left), the second modulator after three half turns, and the exit slit (middle right). (bottom) The transmission as a function of frequency over two cyclotron harmonics (for a 60-keV ^{24}Mg beam) for two radiofrequency amplitudes. The higher curve shows a modest resolving power of 20,000 while the lower curve reaches over 70,000 with the signal reduced since the modulated trajectory envelope is cut by the 5-mm wide phase-definition slit [7], located between the two modulations.

TRAL. Foil targets are favored for short-lived nuclides since the diffusion time is reduced. The side effect is that thermal expansion of such targets under proton impact causes the foils to scinter together (turning surface into bulk volume) slowing the target as well as destroying it mechanically. Our case was no exception and the short target lifetime made for lack of statistics which limited the ultimate precision.

In previous experiments, we uncovered a systematic shift of our measured masses with respect to their difference with the reference mass used (see analysis in [15]). In order to avoid

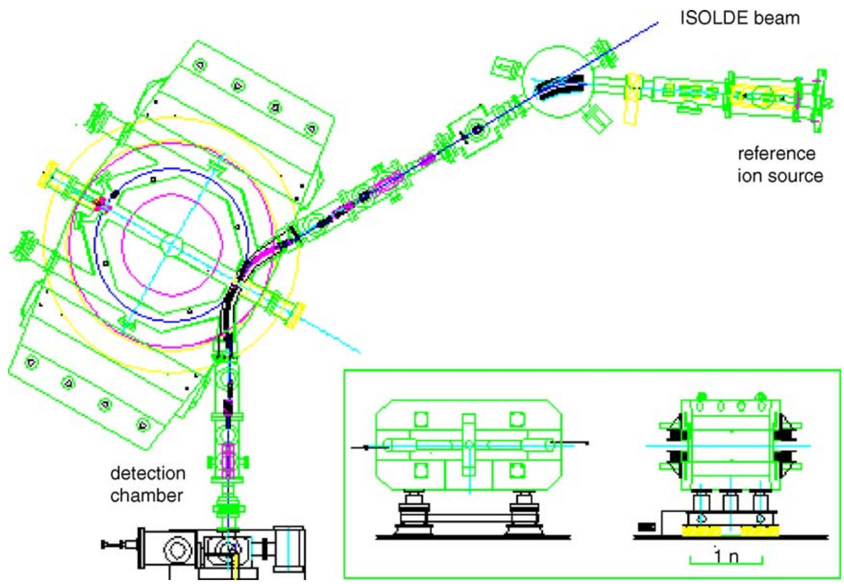

Fig. 2. Overhead view of the MISTRAL spectrometer at CERN's ISOLDE facility. The ion beams (coming from the right) are injected either from the ISOLDE beamline (at 60 keV) or from a reference ion source (variable energy). Inset shows front and side views of the electromagnet and support structure.

such error for ^{74}Rb, a special protocol was established whereby reference beams of stable ^{74}Ge and ^{76}Ge were used. The offset between the ISOLDE beam and reference beam was determined by comparing the well-known ^{76}Rb mass with the (stable) isobar ^{76}Ge. Even in case of isobars, where the mass difference is very small, the two beams still may not have exactly the same trajectory because they originate from two different sources.

As described in [15], comparisons of measured values m_x are made with tabulated AME values $m_x^{\rm ame}$ using the quantity $\Delta_x^{\rm meas} = (m_x - m_x^{\rm ame})/m_x^{\rm ame}$. In this experiment, the values of $\Delta_x^{\rm meas}$ were corrected using the mass measurement of ^{76}Rb compared to that of ^{76}Ge ($\Delta_{76}^{\rm meas}$) according to: $\Delta_x^{\rm corr} = \Delta_x^{\rm meas} - \Delta_{76}^{\rm meas}$. We assume the difference between the $A = 76$ beams is the same for the $A = 74$ beams. Since there is no change in the beam transport system (except for the separator magnet) and the masses are very close, this assumption is reasonable within the conservative error bars. The quantity $\Delta_x^{\rm corr}$ ranged from -12 to -2×10^{-7} over the whole experiment and the average relative uncertainty for the (five) ^{76}Rb measurements is 2×10^{-7}.

A recorded mass peak for ^{74}Rb is shown in Fig. 3(top). The statistics are quite low and dominate the overall uncertainty. The lines in the figure correspond to the triangular fit that is used to determine the peak center (see [7] for detailed derivation of this lineshape). Three measurements of the mass of ^{74}Rb were performed. The differences between the three measured values and the value tabulated in AME'95 [24] are, in keV, -170 ± 237, -71 ± 192 and -387 ± 187. They result in a weighted average of -218 ± 117, considered to be the final value, with $\chi^2 = 0.7$ for the average of the three measurements [19]. This corresponds to a mass excess of

$$\mathrm{ME}(^{74}\mathrm{Rb}) = -51944 \pm 117\,\mathrm{keV} \quad (1)$$

and a total mass of

$$M(^{74}\mathrm{Rb}) = 73.944235(125)\,\mathrm{u}. \quad (2)$$

The statistical error contributes 100 keV to the overall uncertainty with the rest due to the correction brought from the ^{76}Rb–^{76}Ge calibration.

Also shown in Fig. 3(bottom) is the characteristic release curve (see [25] for explanation) of ^{74}Rb, reconstructed from the same data. The decay time of the release curve is seen to correspond to the 65-ms half-life of ^{74}Rb — a method of independant verification that is original to MISTRAL since the transmitted ion signal is measured directly.

On the release curve, a rather high background is visible. This was due to an abnormally high background signal in the secondary-electron multiplier. The effect of this background on the peak position was studied exhaustively in the analysis and found not to affect the final result within the conservative error (see [19]).

Next we examine the results of all experiments in which the mass of ^{74}Rb has been determined. The AME'95 recommended mass [24] was obtained from a Mattauch-Herzog mass spectrometer on-line at the CERN PS facility [26]. At about the same time as the MISTRAL measurement, ^{74}Rb was also measured by the ISOLTRAP spectrometer at ISOLDE. That result is very impressive for the fact that the short half-life does not leave much time for the collection, preparation and actual measurement of the mass. Indeed, ^{74}Rb is the shortest-lived species to ever be investigated by Penning trap mass spectrometry [27]. In Fig. 4, the results of these measurements are compared with those of this experiment. The agreement of all values, despite their widely differing uncertainties (i.e., 720, 117 and 4.0 keV) is impressive.

Although the MISTRAL measurement is not as precise as the ISOLTRAP result, it is nevertheless valuable. In the atomic mass evaluation, the evaluators encourage parallel measurements using different techniques to avoid systematic errors. Precise results, in their words: "... should not remain unchallenged: checks by another group ... are highly desirable to

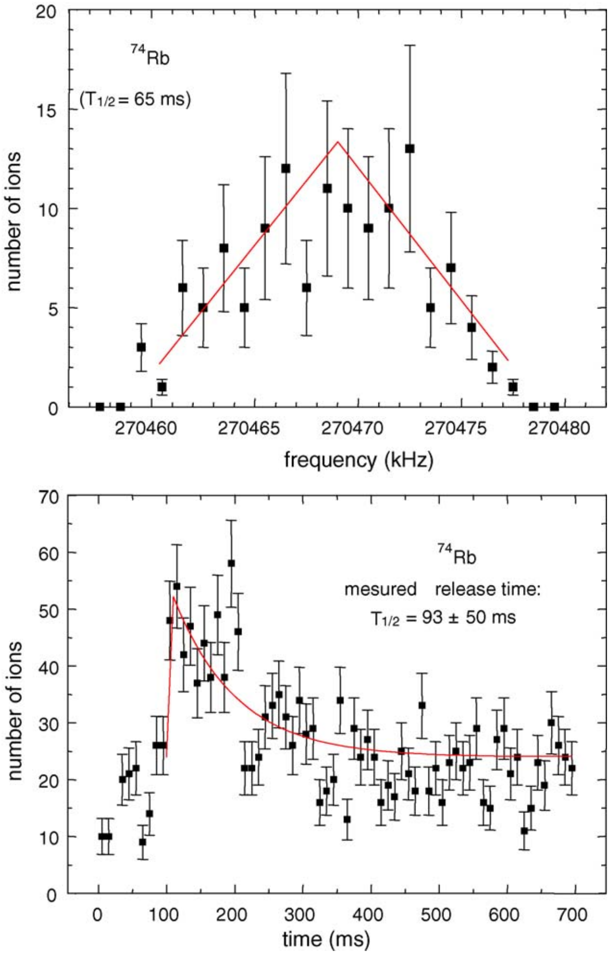

Fig. 3. (top) Transmission peak for ^{74}Rb after summing 67 scans of the radiofrequency and grouping of two channels. Also shown is the (triangular) fit to the theoretically expected lineshape. Due to the weak production rate, a modest mass resolving power of only about 20,000 was used to favor transmission. (bottom) The time dependence of the same recorded data after impact of the proton beam on the target (at 100 ms), showing the characteristic release curve, the decay of which is dominated by the 65-ms half-life.

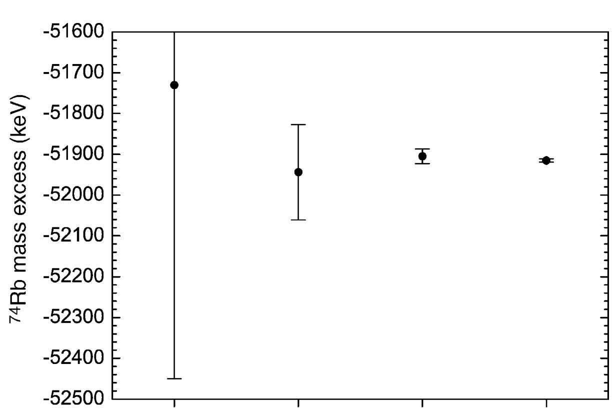

Fig. 4. Comparison of the present measurement of ^{74}Rb with the previously adopted mass value from AME'95 [24] and with the recent results from ISOLTRAP [27,28].

strengthen the validity ... and transform these precise measurements into very accurate ones." [24] (p. 412). In the case of the Penning trap, such short-lived species present a new challenge with perhaps yet un-encountered systematic effects. At the same time, it is an extremely important validation for the MISTRAL technique.

4. Discussion in light of $N = Z$ nuclides

4.1. The n–p interaction

Proton–neutron interactions are fundamentally important to nuclear structure, of particular importance for configuration mixing and questions regarding collectivity and deformation in nuclei. The single particle energies, though quite small, are decisive for determining the ground state configuration and hence, binding energy B. (For a discussion and recent study of the mass surface see [32]). The average interaction of the last proton(s) with the last neutron(s) can be evaluated from binding energy differences via the parameter ΔV_{np}. For the ^{74}Rb case, it is given by

$$\Delta V_{\mathrm{np}}(^{74}\mathrm{Rb}) = [B(^{74}\mathrm{Rb}) - B(^{73}\mathrm{Kr})] - [B(^{73}\mathrm{Rb}) - B(^{72}\mathrm{Kr})]. \quad (3)$$

As the binding energy of ^{73}Rb is unknown, it is deduced from the binding energy of its mirror nuclide ^{73}Kr, to which a Coulomb correction (here, [36]) is applied. This series of differences varies according to whether the nuclei are even–even, odd–odd or even–odd (see [19]).

Fig. 5 shows that the parameter ΔV_{np} is enhanced for $N = Z$ nuclei as compared to the $N \neq Z$ ones. This effect is due to the Wigner energy. It is also a stronger effect for $N = Z$ odd–odd nuclei than the even–even ones. However, the effect decreases when A increases and is thought to progressively vanish. The result obtained for ^{74}Rb contradicts this tendency. This was already suggested by the previous mass measurement, but it is very clearly confirmed here by the more accurate MISTRAL and ISOLTRAP measurements. This feature is expected in the model developed in the frame of Wigner's SU(4) symmetry by Van Isacker et al. [30,31]: the SU(4) symmetry, which produces an enhancement of ΔV_{np} for light $N = Z$ nuclei, is progressively broken when A increases. However, for $N, Z > 28$, a pseudo-SU(4) symmetry could exist, producing an enhancement as observed for ^{74}Rb.

4.2. The Wigner energy[1]

One of the interests of $N = Z$ nuclei is the Wigner energy, which can be represented by an additional term in the semi-

[1] This term is used due to a similarity between binding energy discontinuities at $N = Z$ and a sharp cusp produced by Wigner's supermultiplet theory, based on SU(4) spin-isospin symmetry — see [30].

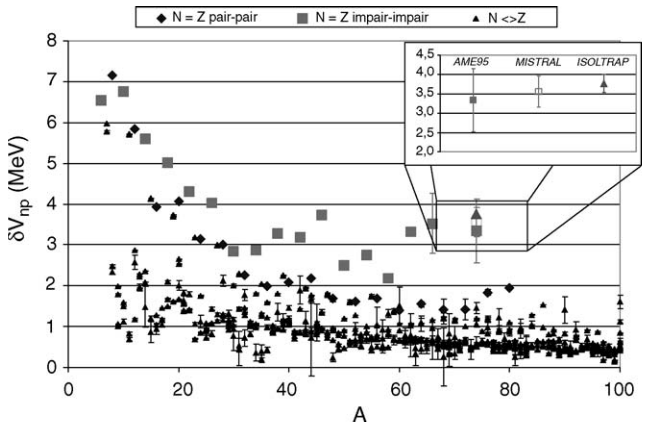

Fig. 5. n–p Interaction, ΔV_{np}, plotted vs. the atomic number A. Diamonds correspond to $N = Z$ even–even nuclides, squares to odd–odd nuclides and triangles to $N \neq Z$. Inset shows that the MISTRAL (empty squares) and ISOLTRAP (triangles) results are clearly larger than the values obtained for $N \neq Z$ nuclei.

empirical formula of Bethe-Weizsäcker:

$$B = a_v A - a_s A^{2/3} - a_c \frac{Z(Z-1)}{A^{1/3}} - a_a \frac{(N-Z)^2}{A} - E_W|N-Z| + \Delta. \quad (4)$$

(In many studies of the Wigner energy, the term E_W has a dependance on A of the form $1/A$ [33–35].) It is interesting to note that the most modern (microscopic) mass formulas, based on the mean-field Skyrme force, systematically underbind the $N = Z$ nuclides and still include such a phenomenological term. In fact, one of the most important factors in improving the first Hartree–Fock–Bogolyubov mass table (HFB-1) [37], was an improvement of this very term in HFB-2 [38] (see also description and discussion in [5]). In summary, no global mass formula has yet been constructed that will include the more fundamental $T = 0$ pairing explicitly since it is still quite unclear how the various correlations are entangled. The more contributions there are to untangle, the more data that is required to untangle them. Mass measurements of (heavier) $N = Z$ nuclides will therefore be of great importance, also to elaborate as yet unseen trends.

5. Outlook

One may well ask, how it is possible to compete with the Penning trap? The "Achilles heel" of the ion storage technique is the very milliseconds of cooling, preparation, and observation that are used up while the exotic ions of interest radioactively decay in vain. While the limit would seem to be of the order of 50 ms – for the moment – new developments, in terms of trap performance, the use of high-charge states, and higher production rates, will certainly lower it.

Though having no such limitation in accessible half-life, MISTRAL's weakness is its poor transmission that keep most of the shortest-lived nuclides out of reach. The addition of a new beam cooling system [39,40] will help in this regard. This system has now been integrated and was successfully used in September 2005 on a test measurement of ^{12}Be, whose half-live is only 21 ms. This measurement, as well as a full description of the new beam cooler, will be subjects of a future publication.

MISTRAL, at over 20 t, is a rather weighty piece of apparatus. Admitting to a vulnerability to systematic error, MISTRAL is somewhat like "a millstone around the neck". However, with the mass table in need of results from different techniques, we can agree that "all is grist for the mill".

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