

MASS MEASUREMENTS OF SHORT-LIVED NUCLEI USING MISTRAL AT ISOLDE

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The MISTRAL experiment is able to measure masses of very short-lived nuclides with an accuracy of a few 10^{-7} . Masses are determined by measuring the cyclotron frequency of an ion in a homogeneous magnetic field. The ISOLDE separator beam (60 keV) is injected directly into the spectrometer alternately with a stable beam used as a mass reference. Near $N=20$, the masses of $^{26-30}\text{Na}$, $^{25-26}\text{Ne}$ and ^{32}Mg with half-lives as short as 30 ms have already been measured. Very recently, the mass precision of ^{74}Rb ($T_{1/2}=65$ ms) has also been improved by a factor 7. These results are presented, compared to various mass models, and discussed.

1 Motivations

Atomic mass measurements are the key to study the behaviour of nuclear binding energies.

On the first hand, they allow to observe and study nuclear structure such as shell closures, pairing, or deformations which break the general smoothness of the binding energies. However, these effects are relatively tiny, typically about 1 MeV or even less, compared to the total mass of the order of 100 GeV for $A=100$, *i.e.* 10^{-5} only. This implies that atomic mass measurements must be accurate at the level of 10^{-6} .

On the second hand, in order to build models able to reproduce and predict masses, it is important to have available a set of masses as large as possible and more especially for nuclei with large excesses of neutrons or protons. This is particularly important in view of astrophysical calculations of the r-process which call very neutron-rich nuclides into play. Indeed, these nuclei are very difficult to produce, and therefore very rare. Furthermore their

half-lives are eventually quite short, especially in the case of light nuclei where the drip-line is reached. This requires that the mass measurement methods are fast and feasible with the few available atoms.

2 The Mistral experiment

Many different methods have been developed to measure masses. Each one is optimized according a special emphasis either on the precision, or the short half-lives, or the sensitivity.

MISTRAL (Mass measurements at ISOLDE/CERN with a Transmission and Radio-frequency spectrometer on-Line) has been designed to reach a high precision (about 0.5 ppm), and at the same time, to have a very low half-life limitation. It is presently installed on-line with the ISOLDE mass separator facility at CERN ¹. The flight time through MISTRAL is about 50 μ s so that the limitations only come from the ISOLDE yields. They depend on the production cross section of the radioactive species and on their diffusion time out of the thick target bombarded by the 1 GeV proton beam from the PS booster.

2.1 MISTRAL principle

MISTRAL is a radio-frequency, transmission spectrometer based on the principle proposed by L.G. Smith in the sixties ^{2,3}. The mass m of an ion of charge q is determined by measuring its cyclotron frequency while it rotates in a homogeneous magnetic field \vec{B} (Fig. 1) :

$$f_c = \frac{q}{m} \frac{B}{2\pi} \quad (1)$$

The 60 keV radioactive beam of 1^+ ions provided by ISOLDE is directly injected into the MISTRAL spectrometer through a 0.4 mm wide inlet slit. It follows a two-turn helicoidal trajectory before being extracted and transported onto a secondary electron multiplier for counting. At the one-half and three-half turns inside the magnetic field, the beam passes through a radio-frequency acceleration gap which modulates its energy, and hence the diameter of its trajectory. Instead of being focused onto the exit slit, the beam is spread out and only a small fraction is transmitted through the 0.4 mm exit slit, except if the two modulations cancel exactly. This is the case when the radio-frequency is related to the cyclotron frequency by :

$$f_{RF} = (n + \frac{1}{2})f_c \quad (2)$$

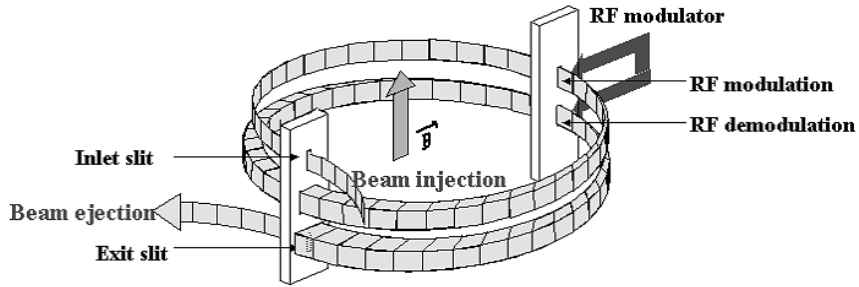


Figure 1. Principle of MISTRAL (transmission spectrometer) : the ions are injected through a defining inlet slit. They rotate twice inside the homogeneous magnetic field along a helicoidal trajectory. Two radio-frequency energy modulations separated by one turn are effected so that the diameter of the trajectory is modulated accordingly. The net effect is to prevent the beam to be focused on the exit slit, except when the radio-frequency is $(n + 1/2)$ times the cyclotron frequency (n integer).

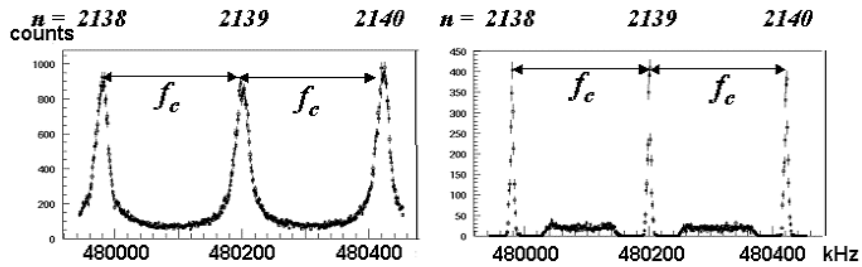


Figure 2. Example of scan of the transmission versus the radio-frequency. Left : regularly spaced maxima are observed for each integer value of n . Right : applying a phase selection half a turn after the first modulation allows to suppress the background around the transmission peaks of interest which can be fitted by a theoretical triangular function.

where n is an integer.

By scanning the transmission versus the radio-frequency, narrow peaks are observed for each integer value of n (Fig. 2, left). Furthermore, an adjustable slit situated half-way between the two modulations, allows to select a phase window of the first modulation, thus suppressing the background around the peak (Fig. 2, right) ⁴. The price to pay is a reduction of the transmission, but the resolving power is improved and the theoretical triangular function

may be used to fit the peak shape (Fig. 3). As the magnetic field cannot be measured with sufficient accuracy, the masses m_x of the radioactive nuclides from ISOLDE are determined relative to a reference one m_{ref} produced by the MISTRAL source. At constant \vec{B} :

$$m_x = \frac{f_{c,ref}}{f_{c,x}} \times m_{ref} = \frac{f_{ref}}{f_x} \times \frac{n_x + 1/2}{n_{ref} + 1/2} m_{ref} \quad (3)$$

The kinetic energies E must at the same time obey the relation :

$$m_x E_x = m_{ref} E_{ref} \quad (4)$$

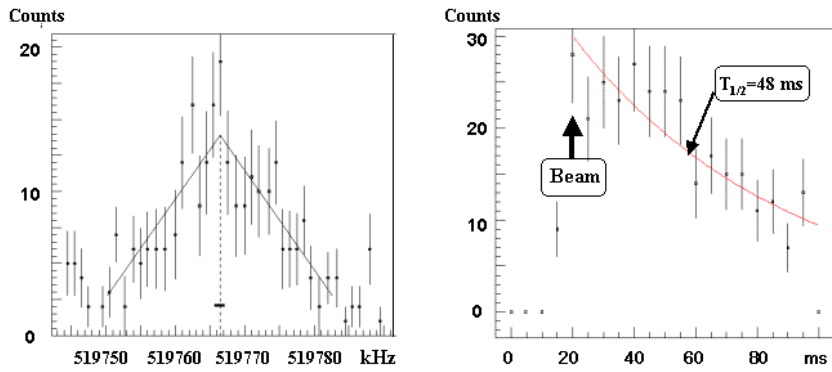


Figure 3. Left : example of a resonance peak obtained for ^{30}Na . The central frequency is determined as $f = 519766.2 \pm 0.7$ kHz. Right : example of time dependence observed for ^{30}Na , with an exponential function drawn for its 48 ms half-life.

The mass jump from one mass to the other, without changing \vec{B} , implies a fast (typically 1-3 s) switch of all the electrostatic potentials of the injecting and ejecting beam transport elements.

2.2 MISTRAL performance

The FWHM resolving power is given by ⁴ :

$$R = 2\pi n \frac{D_m}{w} \quad (5)$$

where D_m is the amplitude of the diameter modulation, and w is the exit slit width. The basic parameters of MISTRAL summarized in Table 1 lead to

Table 1. Main features of the MISTRAL spectrometer

Ion kinetic energy	30-80 keV
Time of Flight	$\sim 50\mu\text{s}$
Frequency band	250-500 MHz
Mass resolving power	10^5
Sensitivity	$\sim 10^3$ ions/s
Precision	$\Delta m/m \sim 5 \cdot 10^{-7}$

$R \sim 10^5$. Using the MISTRAL source, a $5 \cdot 10^{-3}$ transmission is currently obtained while a somewhat lower one is obtained for the ISOLDE beam, due to its larger emittance, and to the optical aberrations related to a long transport line from ISOLDE to MISTRAL. However, the limit of sensitivity was observed to be $\simeq 1000$ ions/pulse. By fitting a triangular function to the resonance peak, and taking into account the magnetic field fluctuations, the resonance frequency may be reproducibly determined with an accuracy of $2\text{-}5 \cdot 10^{-7}$ provided that the statistics are sufficient.

3 Mistral measurements

Measurements were performed on Ne, Na, Mg and Al isotopes near $N=20$, on some K isotopes and on the very neutron-deficient nucleus ${}^{74}\text{Rb}$ with $N = Z$.

The neutron-rich light nuclides were produced by fragmentation of uranium while ${}^{74}\text{Rb}$ was produced by spallation of niobium.

A typical recorded peak, obtained for ${}^{30}\text{Na}$, is shown in Fig. 3 (left). Due to the pulsed structure of the proton beam, the radio-frequency is scanned 1 step/pulse and the time dependence is recorded simultaneously. Fig. 3 (right) shows a good agreement between the observed time dependence and the one expected from the known radioactive half-life.

3.1 Calibration

Masses of ${}^{23\text{-}30}\text{Na}$ and ${}^{27}\text{Al}$ from ISOLDE were measured using ${}^{23}\text{Na}$ and ${}^{39}\text{K}$ from MISTRAL as references, implying very large mass jumps. The very precisely known ${}^{23\text{-}25}\text{Na}$ and ${}^{27}\text{Al}$ masses were used to check and eventually calibrate the measurement procedure. It appeared that they were deviating proportionally to the difference $(m_x - m_{ref})$. Such a linear function can be understood in relation with the voltage switching required to compare the masses m_x and m_{ref} of ions rotating in the same magnetic field. At

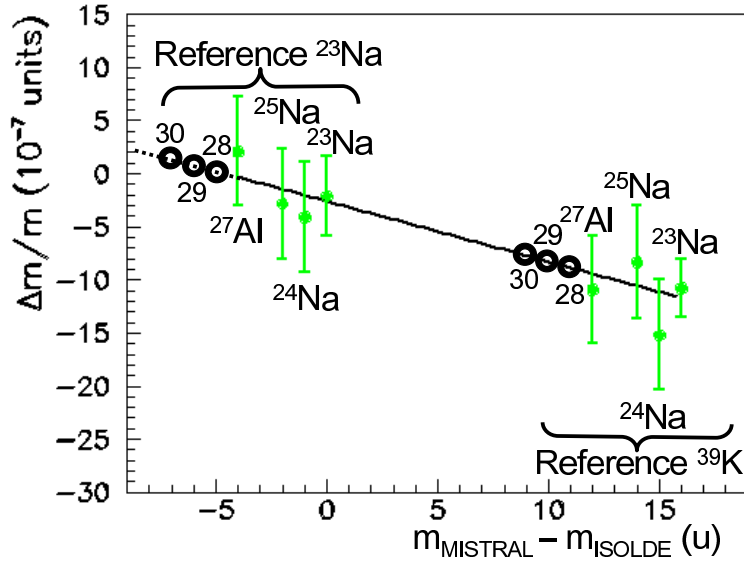


Figure 4. Example of calibration using the well known masses of $^{23,24,25}\text{Na}$ and ^{27}Al as calibrants from ISOLDE and ^{23}Na or ^{39}K as reference masses from MISTRAL. $\Delta m/m$ is the relative difference between the measured value and the one from the AME'95 Mass Table ⁶. The corrections to apply for $^{28,29,30}\text{Na}$ are indicated.

the measurement time, the voltages may be off by a few 10^{-4} so that the trajectories inside the magnet are slightly shifted. If the magnetic field is not homogeneous enough ⁵, the ions do not experience exactly the same field and equation 3 no longer holds exactly. Different calibration functions may be proposed but, at first approximation, the precision of the measurements does not allow to choose between them. Finally, a calibration of the form

$$\frac{\Delta m}{m} = \frac{m_{meas} - m_{table}}{m_{table}} = a(m_{ref} - m_x) + b \quad (6)$$

where $\Delta m/m$ is the relative difference between the measured value and the one from the AME'95 Mass Table ⁶ (calibrant mass) has been used satisfactorily ⁷ (see *e.g.* Fig. 4). The constant term b reflects the ISOLDE and MISTRAL beams shift. Its value was typically a few 10^{-7} . The slope a varied from -1 up to $+7 \cdot 10^{-7}$ according to the beam lines tuning. Moreover, since for a given line tuning the measurements were slightly fluctuating, a systematic uncertainty

was added to each measurement (e.g. 4.10^{-7} for the sodium measurements)

It is obviously highly desirable to minimize the $(m_x - m_{ref})$ difference. In some cases, it was even possible to use isobars as references, e.g. $^{74,76}\text{Ge}$ for $^{74,76}\text{Rb}$, or to compare two isobars coming from ISOLDE e.g. ^{27}Al - ^{27}Na .

3.2 Mass results around $N=20$

The masses of $^{26-30}\text{Na}$ were determined using a selective thermionic ionization source. The results are shown in Fig. 5 as compared to the values from the AME'95 mass table ⁶. It is seen that all four calibrants are in very good agree-

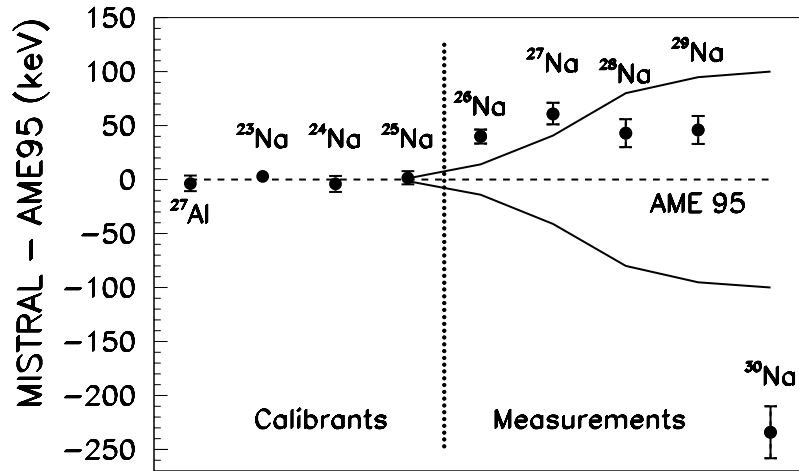


Figure 5. Comparison of the mass values measured by MISTRAL with those from the AME'95 Mass Table. The solid lines indicate the limits of the uncertainties from the Table. It is seen that $^{26,27}\text{Na}$, and especially ^{30}Na are deviating more than 1σ .

ment with the Mass Table. The measured values exhibit large improvements of the precision, up to one order of magnitude, but ^{26}Na and ^{30}Na clearly deviate from AME'95. For ^{26}Na , more details are given in reference 7. For ^{30}Na , Fig. 6 shows that only the TOFI measurement performed at LAMPF by Zhou *et al.* ⁸ is really deviating. This measurement was dominantly influencing the Mass Table value. A previous measurement published by the same group ⁹ was also strongly deviating, but in the opposite direction. The incompatibility between these two results was explained assuming an isobaric contamination of ^{30}Na by ^{30}Mg in the earliest work. All other data ^{10,11,12,13}

agree nicely.

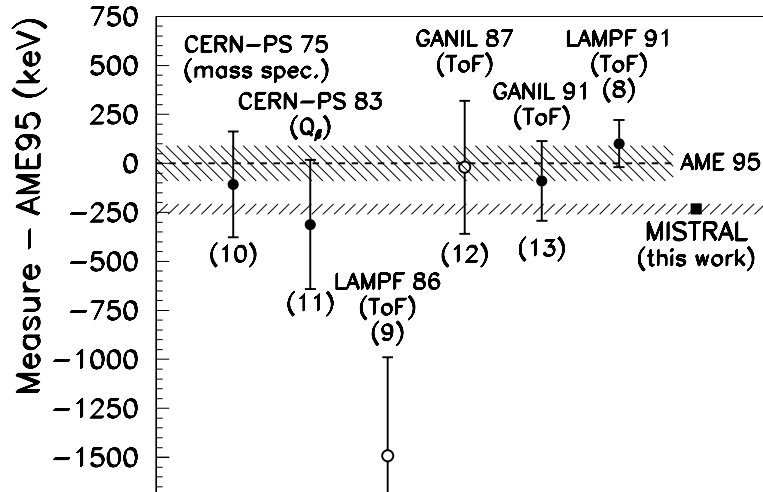


Figure 6. Comparison of all existing measurements for ^{30}Na . Full circles correspond to the results used to adjust the AME'95 mass value.

$^{25,26}\text{Ne}$ and ^{32}Mg masses were measured using a non-selective plasma ion source. Isobaric contamination turned out to be very difficult to eliminate. As MISTRAL produces a transmission peak for each harmonic n , even distant isobars may produce mixed peaks corresponding to different harmonics. The position of possible isobars, and the observed time dependence must be carefully checked. If a contamination is detected during the measurement, it is possible to circumvent it by selecting another radio-frequency in order to modify the relative positions of the two overlapping peaks. This could be done successfully for ^{27}Al - ^{27}Na , but not in the case of ^{31}Al - ^{31}Mg where the problem was not detected in time. ^{32}Mg looks to be free from contamination, but is nevertheless considered as preliminary ^{14,15}.

It is expected to remeasure ^{32}Mg in a cleaner environment by using a

selective RILIS ion source^a. As for ³⁰Na, the MISTRAL determination leads to a more bound nucleus than AME'95, and the disagreement only comes from the last TOFI measurement⁸ which was the most influential one.

Fig. 7 shows the two-neutron separation energy S_{2n} versus the neutron number for Na, Mg, P and S. A smooth function ($f(Z, N) = -3300N + 100NZ + 700Z + 39800$) has been subtracted in order to emphasize the nuclear structure effects. ¹⁶S and ¹⁵P isotopes exhibit a strong shell closure at $N = 20$. Conversely, for ¹²Mg and ¹¹Na, S_{2n} increases for $N \geq 19$, indicating the onset

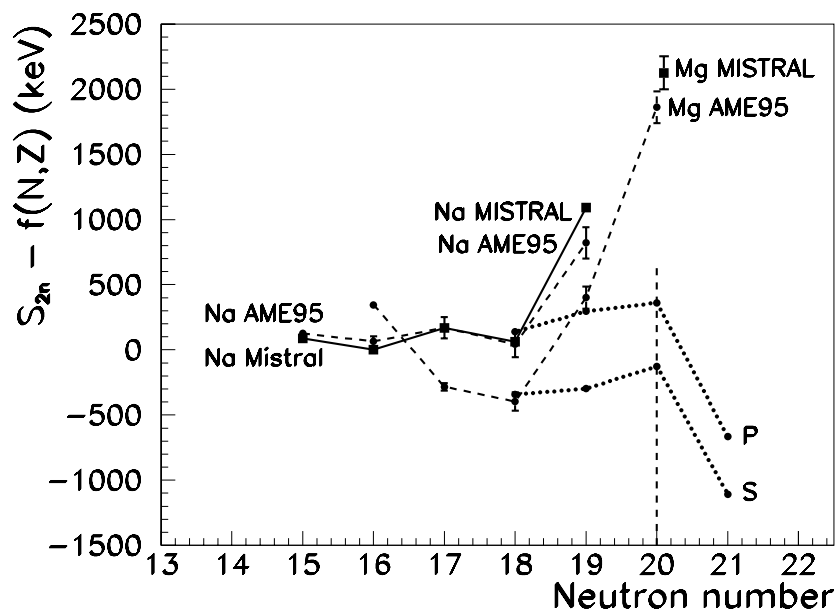


Figure 7. Two-neutron separation energy versus the neutron number N for the elements Na, Mg, P, S after subtraction of a smooth function.

of a deformation as it was first noticed from first direct mass measurements in 1975¹⁰. The MISTRAL new measurement enhances the effect in this region, now known as an island of inversion.

^ain September 2001, using the RILIS ion source, it was possible to measure the whole series ²⁹⁻³³Mg with good statistics. The preliminary value for ³²Mg, here cited, appears to be confirmed.

Fig. 8 compares the Na experimental masses to a few microscopic models. Even if not accurate, the old Hartree-Fock calculation from Campi *et al.* ¹⁶ as well as a very recent one from Goriely *et al.* ¹⁷ reproduce the onset of the deformation at $N = 19$. A recent shell-model calculation by Caurier *et al.* ¹⁸ produced a better fit for $N \leq 18$, but the deformation occurs at $N = 20$, one neutron too late. This island of inversion is predicted to be very localized and to disappear for $N \geq 22$. Accurate measurements further from stability would be welcome...

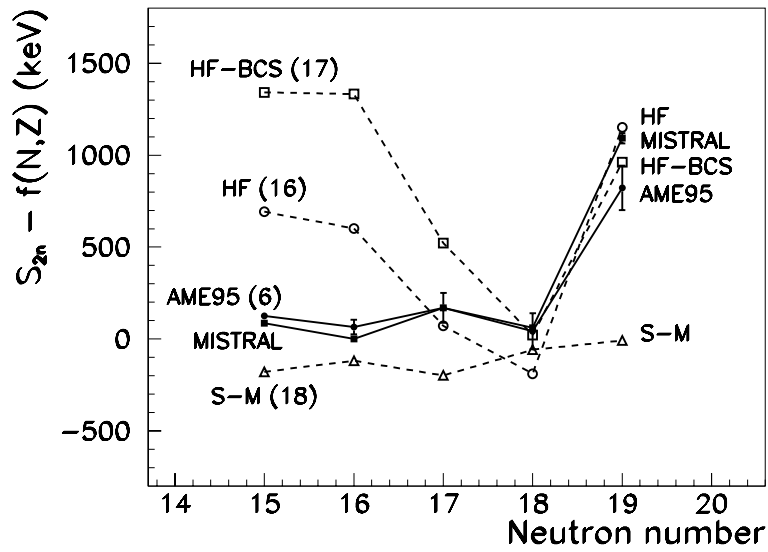


Figure 8. Comparison of the measured two-neutron separation energy (full symbols) with the predictions of different microscopic models (open symbols). Measurements : MISTRAL (squares), AME'95 ⁶ (circles). Models : Hartree-Fock ¹⁶ (circles), Hartree-Fock BCS-1 ¹⁷ (squares) and Shell Model ¹⁸ (triangles).

3.3 ⁷⁴Rb mass measurement at the proton drip line

There are several motivations to measure the masses of $N = Z$ nuclei. A fundamental one is to test the CVC hypothesis by measuring the $\mathcal{F}t$ values

of $0^+ \rightarrow 0^+$, $T = 1$ Fermi super-allowed β -decays. This requires a very high precision together for the masses ($\sim 10^{-8}$), for the half-lives ($\sim 10^{-4}$) and for the branching ratios. The second motivation, requiring less precision, is to study the $n - p$ interaction and the so-called Wigner term.

The difficulty is that ^{74}Rb ($T_{1/2}=65$ ms), a proton drip-line nuclide, is poorly produced at ISOLDE : 1000 ions/pulse, *i.e.* very near the present sensitivity limit of MISTRAL. However three measurements could be achieved using the isobar ^{74}Ge as reference. A small correction (a few 10^{-7}) determined from the comparison of the well known ^{76}Rb with the isobar ^{76}Ge was applied. The three measurements are in good agreement. Their weighted mean value is $73.944230(107)\text{u}$ ¹⁹. The accuracy of $1.3 \cdot 10^{-6}$ is limited by the statistics, but already 7 times better than the previous one measured by Audi *et al.* ²⁰. Another measurement using the ISOLTRAP Penning trap ²¹ was also successful at ISOLDE. This measurement still improves by a factor 4-5 the MISTRAL measurement. Both agree very well.

Aiming at the $\mathcal{F}t$ value, the precision of the ^{74}Rb half-life was also recently very much improved ^{22,23}. However a one order of magnitude improvement on the mass value is still needed.

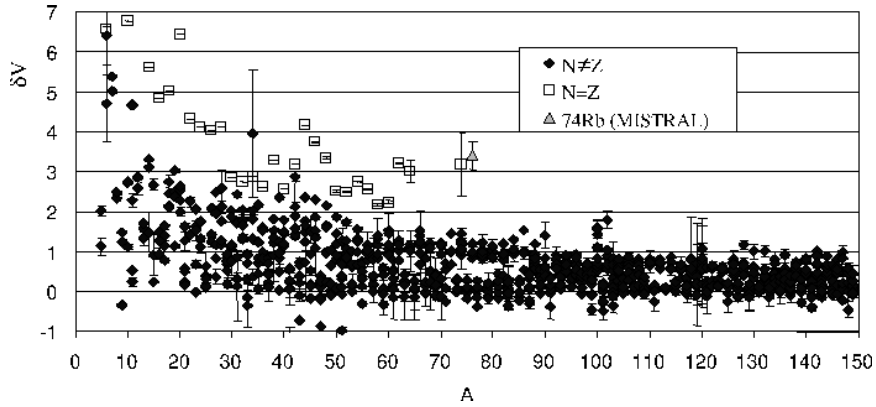


Figure 9. $\delta V = B(N, Z) - B(N - 1, Z) - B(N, Z - 1) + B(N - 1, Z - 1)$ is plotted versus the atomic number A . Diamonds correspond to $N \neq Z$ nuclei. Squares correspond to $N = Z$ nuclei. The MISTRAL result for δV (triangle slightly shifted) is clearly larger than the values obtained for $N \neq Z$ nuclei.

Concerning the $n - p$ interaction, it is usual to calculate double differences δV between the binding energies of four nuclides in a square :

$$\delta V = B(N, Z) - B(N - 1, Z) - B(N, Z - 1) + B(N - 1, Z - 1) \quad (7)$$

If unknown, the masses of $N < Z$ nuclei can be deduced from their mirror nuclei. Fig. 9 shows that δV is enhanced for $N = Z$ nuclei as compared to the $N \neq Z$ ones. However, the effect decreases when A increases and is thought to progressively vanish. The result obtained for ^{74}Rb contradicts this tendency. It was already suggested by the previous mass measurement, but it is very clearly confirmed here by the more accurate MISTRAL measurement. Such a feature is expected in the model developed in the frame of Wigner's SU(4) symmetry by Van Isacker *et al.* ^{24,25} : the SU(4) symmetry, which produces an enhancement of δV for light $N = Z$ nuclei, is progressively broken when A increases because of the increasing importance of the spin-orbit term. However, for $N, Z > 28$, a pseudo-SU(4) symmetry could exist, producing an enhancement as observed for ^{74}Rb .

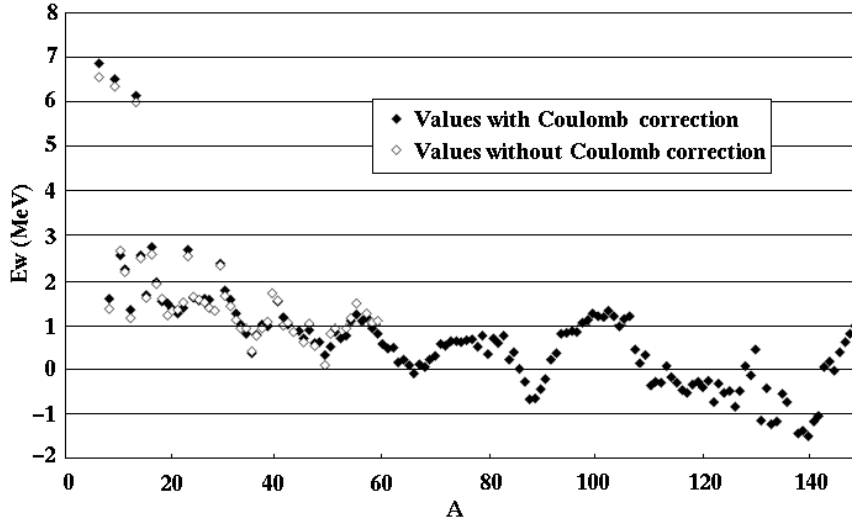


Figure 10. Wigner energy obtained with (full diamonds) and without (open diamonds) Coulomb corrections as a function of the atomic number A .

Measurements along the $N = Z$ line also open the possibility to better study the Wigner term E_W appearing in the Bethe-Weizsäcker semi-empirical mass formula :

$$B = a_v A - a_s A^{2/3} - a_c \frac{Z(Z-1)}{A^{1/3}} - a_a \frac{(N-Z)^2}{A} - E_W |N-Z| + \delta \quad (8)$$

For $N \neq Z$, E_W is found to be inversely proportional to A ^{26,27,28}. In these studies, the Wigner energy is determined by differences corresponding to second derivatives of the binding energy. Here, in Fig. 10, a more direct method, applicable in regions where $N - Z < 0$ nuclei do not exist, was used. For each series of isobars, the Coulomb and pairing contributions, which depend on T_Z , are subtracted. Afterwards $B(T_Z)$ may be fitted to a parabola :

$$B = C_1(A) - 2E_W|T_Z| - C_3(A)T_Z^2 \quad (9)$$

In the case of light nuclei where the masses of $T_Z < 0$ are known, a similar fitting procedure may be applied without any correction. The good agreement between both results validates the first method. The general trend is generally found to follow a law in $40/A$, but a pronounced shell effect is superimposed with maxima corresponding to $Z, N = 20, 28, 40, \text{ and } 50$ ²⁸, which would appear to be correlated to (HO) or (HO + SO) shell closures.

4 Prospects

In the near future, some other interesting cases are in the MISTRAL scope. These are nuclei with $T_{1/2} < 50$ ms sufficiently produced by ISOLDE (≥ 1000 ions/pulse) : ^{11}Li ($T_{1/2} = 8.5$ ms), ^{12}Be , ^{27}Ne , and $^{99-101}\text{Rb}$. ^{11}Li is a halo nucleus for which an accurate mass value is valuable ^{29,30,31}. Furthermore, several detailed microscopic models calculations specially devoted to ^{11}Li and some other halo nuclei have been developed ^{32,33,34,35} very recently, which are adjusted to the experimental binding energies. Presently, only one measurement reaches an accuracy better than 10^{-5} (35 keV): MISTRAL could confirm its validity and improve the accuracy by a factor 5-7.

Another interesting case is that of $^{99-101}\text{Rb}$ since a deformation is known to occur at $N = 60$ (^{97}Rb) ^{36,37}. A better knowledge of this region would also be welcome.

In order to go further, the sensitivity limit of 1000 ions/pulse needs to be overcome. In order to reduce the beam emittance, and hence to increase the MISTRAL transmission, a R&D has been started. The chosen cooling method needs to be fast and non selective. These criteria are fulfilled by using gas interaction inside a radio-frequency quadrupole serving as an ion guide ³⁸(Fig. 11). The first indication of an emittance reduction has been obtained ^{39,40}.

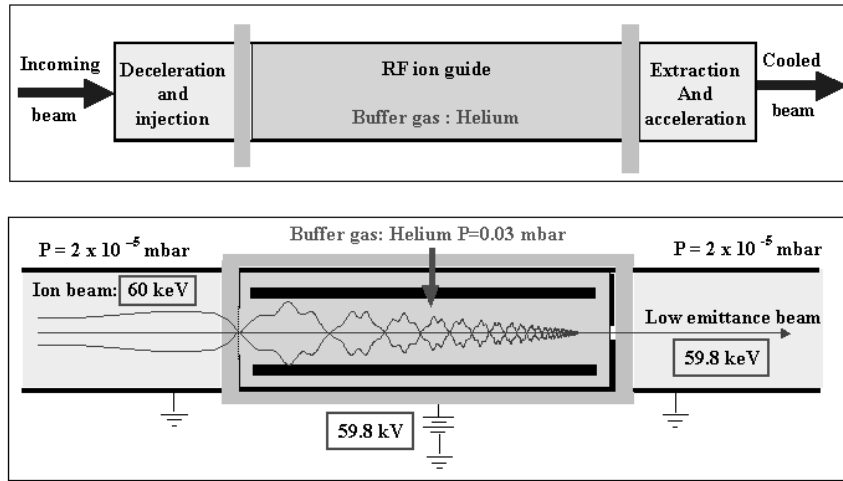


Figure 11. Principle of the cooling device. The 60 keV ion beam from ISOLDE or MISTRAL is decelerated to ~ 200 eV, cooled by interaction with the He gas, and focused on the axis by the RFQ. It is then extracted and reaccelerated.

5 Conclusions

MISTRAL succeeded to measure masses of very short-lived nuclides with an accuracy of a few 10^{-7} as expected. The shortest one, ^{28}Na ($T_{1/2}=30.8$ ms) was measured with an accuracy of $5 \cdot 10^{-7}$. Some more physics cases are still in view, but improving the transmission as expected by the present cooling development would enlarge the possible scope.

Acknowledgments

The authors are very grateful to G. Conreur, M. Jacotin, J.F. Képinski, G. Le Scornet from Orsay, M. Duma from IAP, Bucharest and G. Lebée from CERN for their able technical assistance. This research was funded by the IN2P3, France, and some of the work at ISOLDE benefited from the European RTD Program “Access to Large-Scale Facilities”.

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