EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

CERN/ISC 99-3
ISC/P107
18 January 1999

Status Report and Proposal to the ISOLDE Experiments Committee

(IS-346)

MISTRAL: MASS MEASUREMENTS AT ISOLDE WITH A TRANSMISSION RADIOFREQUENCY SPECTROMETER ON-LINE

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Abstract. The *MISTRAL* experiment has now measured its first masses at *ISOLDE*. The direct technique utilized by this radiofrequency transmission spectrometer allows concentration on nuclides with particularly short half-lives. In its first experiment in July 1998, neutron-rich Na isotopes having half-lives as short as 31 ms were measured. A second experiment in November 1998 enabled us to pinpoint the cause of a systematic error and improve the measurement precision of the isotopes $^{26\text{-}30}$Na to about 25 keV. Further measurements were made on K, Ca and Ti isotopes out to A = 48. We propose a continuation of the measurement program on neighboring isotopes of Ne, Mg and Al in the N = 20 region and K and Ar isotopes across the N = 28 region. These normally-magic configurations suffer from a competing deformation effect that rather increases the binding energies. This problem has long attracted interest and direct, high-precision mass measurements in these regions would complement on-going work at *ISOLDE* and elsewhere. We request 36 shifts of beam time.

1. Introduction and Motivation

The interest in measuring masses of radioactive isotopes comes, in part, from an ever-present need to constrain nuclear mass models for which extrapolations are in very poor agreement (fig.1) due to differing quantities of microscopic nuclear physics in the model formulation. The topography of the mass surface gives us clues as to the nature of nuclear effects manifested in the binding energy that include pairing, deformation, shell and sub-shell closures. Masses also give very important information of interest to exotic nuclear phenomena such as isospin symmetry breaking and halos. Knowledge of the nuclear binding energy is also extremely important for nucleosynthesis, particularly the rapid neutron capture process, thought to occur in supernovae. Masses of isotopes very far from stability are required to calculate a variety of physical quantities involved in this process and to reproduce the abundances of the heavy elements observed in the solar system [1,2].

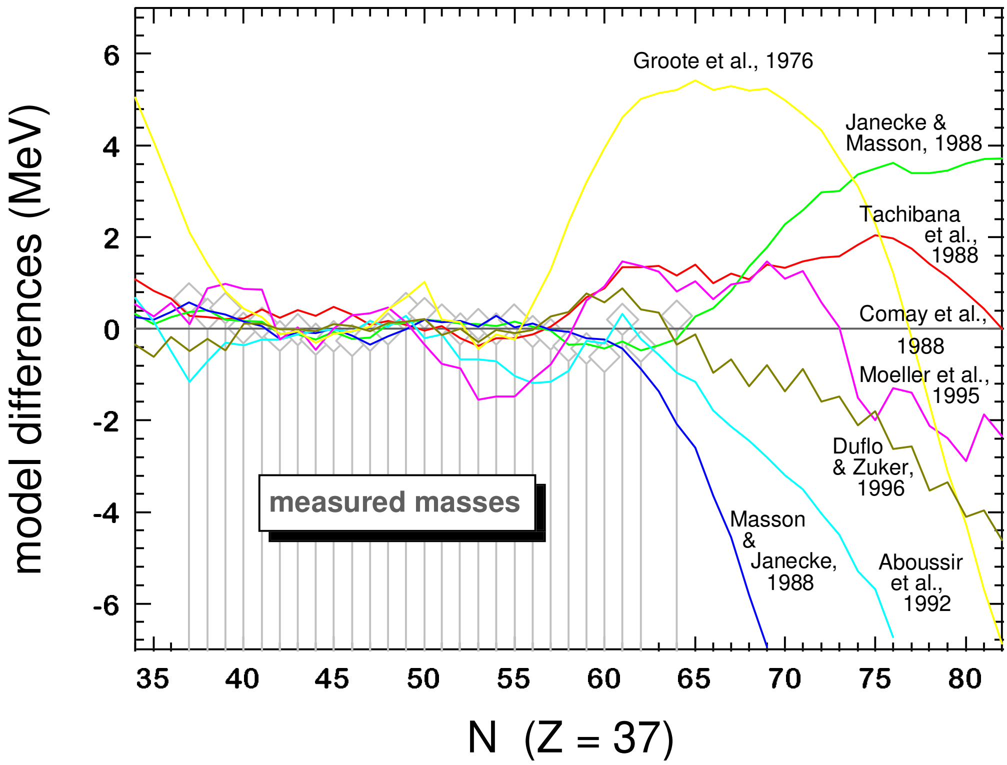

FIGURE 1. Difference in mass predictions of various models as a function of N for $Z = 37$ (Rb). Since the model parameters are adjusted to measured masses, the agreement is very good where masses are known. The rapid neutron capture process path could cross anywhere, depending on the astrophysical conditions, between $N = 65$ and 80.

MISTRAL is one of several programs dedicated to the accurate mass measurement of radioactive isotopes (for reviews see [3,4]). These programs are all complementary in technique and/or applicability. They include time-of-flight techniques that make use of high energy fragmentation reactions in thin-targets such as *TOFI* [5] at Los Alamos and *SPEG* [6] at GANIL. To increase precision by increasing the flight path, cyclotrons have now been used to accelerate the reaction products [7]. Storage rings offer nuclear lifetime measurements as an added bonus to huge mass harvests from high energy fragmentation reactions [8]. Penning traps offer an excellent environment for mass measurements and consequently hold the records for precision [9,10]. The key is that a single ion may be held for as long as necessary to make a measurement. *ISOLTRAP* (IS-302), the first on-line Penning trap spectrometer, has long provided excellent, precise measurements of radioactive isotopes at *ISOLDE* [11]. It will soon be joined by the CPT (Chicago/Canadian Penning Trap) which will measure masses of nuclides produced at the fragment mass analyzer at Argonne [12].

The *MISTRAL* spectrometer, also at *ISOLDE*, is a sort of hybrid between a cyclotron and a Penning trap. Its rather special technique of radiofrequency modulation of the cyclotron motion at the full beam transport energy allows very rapid measurements of excellent precision thus rendering it particularly suitable for short-lived isotopes [13,14]. Thus, *MISTRAL* complements *ISOLTRAP* which must store ions for longer periods in order to make a very precise measurement.

MISTRAL has now achieved its first results - only eighteen months after its arrival at *CERN*. Twenty-two masses have now been measured (see sec.3), fifteen of which are radioactive and three of which have lifetimes of less than 50 ms. Of existing high-precision, direct techniques, *MISTRAL* is the only spectrometer capable of reaching the sub-100 ms regime and is in no way limited in half-life for *ISOLDE*-produced isotopes.

This capacity for extremely short-lived isotopes is of great interest for a key problem in nuclear physics which we have now started to study and expect to continue in the upcoming period namely, the so-called "island of inversion" around the $N = 20$ shell closure. Fig. 2 illustrates the problem as the two-neutron separation energies for Na increase when crossing the shell closure where normally they should decrease. Experimental evidence for this shell erosion was put forth originally with mass measurements by Thibault *et al.* [15] while pure *sd*-shell calculations [16] failed to produce this increase in binding energy. Further mass measurements followed from *TOFI* [5] and *SPEG* [6] but the results showed a discrepancy with *TOFI* finding the effect to be attenuated.

Further anomalous behavior in Na came from laser spectroscopy measurements that showed an increase in charge radius starting at $N = 18$ [17]. This and the observation of an exceptionally low-lying 2^+ level in the isotonic ^{32}Mg [18] gave rise to the hypothesis of a competing deformation effect, later confirmed by a large *B(E2)* value determined via Coulomb excitation [19].

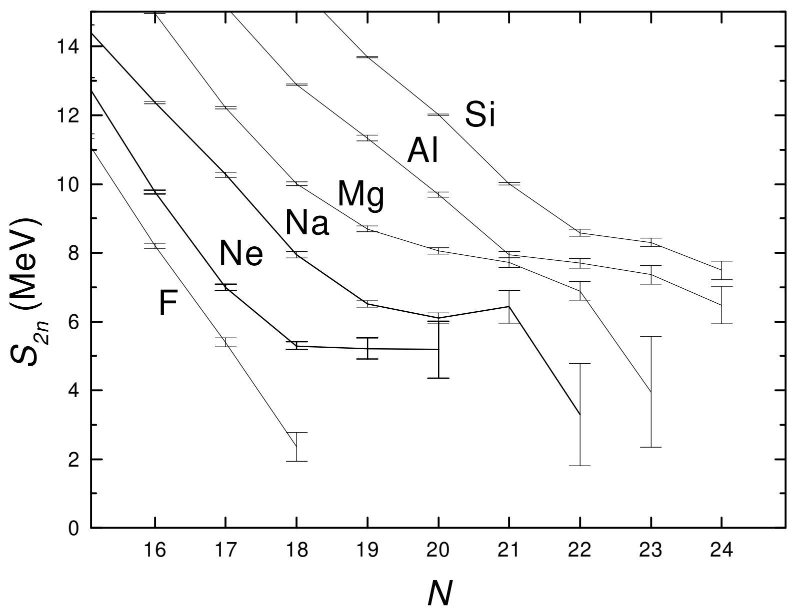

FIGURE 2. S_{2n} values for F, Ne, Mg, Na, Al and Si in the region of the $N = 20$ pseudo shell-closure, from [35].

Hartree-Fock calculations performed by Campi *et al.* [20] offered the explanation of neutron excitations to the *fp*-shell for increased binding energy - a hypothesis strengthened further by shell model calculations with an extended space [21,22] as well as HFB calculations [23]. However, microscopic models that advocate shell erosion far from stability do an unsatisfactory job in predicting deformation. For example, in a recent HFB calculation, Bonche *et al.* [24] show shell erosion at $N = 28$ but in their calculation ^{32}Mg is spherical. Thus, controversy, if not shell-structure, persists!

2. *MISTRAL*

A schematic diagram of the *MISTRAL* spectrometer with its nominal trajectory is shown in fig.3. Ions injected at the full *ISOLDE* beam energy (60 kV) follow a two-turn helicoidal trajectory inside the annular, homogeneous magnetic field (fig.3, inset center) and are counted using a secondary electron multiplier. With an injection slit size of 0.4 mm and orbit radius of 0.5 m, a mass resolution of 2500 is obtained using no radiofrequency. In order to make a measurement, a longitudinal kinetic energy modulation is effected using two symmetric electrode structures (fig.3, inset right) located at the one-half and three-half turn positions inside the magnetic field. This way the ions make one cyclotron orbit between the two modulators. A radiofrequency voltage is applied to the central modulator electrodes. Depending on the phase of this voltage when the ions traverse the structure, the resulting longitudinal acceleration produces a larger or smaller cyclotron radius than that of the nominal trajectory since all the trajectories are isochronous. The ions are transmitted through the 0.4 mm exit slit when the net effect of the two modulations is zero. This happens when the radiofrequency voltage is an integer-plus-one-half multiple of the cyclotron frequency which means that during the second modulation the ions feel exactly the opposite of what they felt during the first. For high harmonic numbers (e.g. larger than 1000) and a radiofrequency voltage of about 200 V, the ion signal over a radiofrequency scan shows narrow transmission peaks having resolutions of about 50,000 (over 100,000 is possible) evenly spaced at the cyclotron frequency (fig.3, inset left).

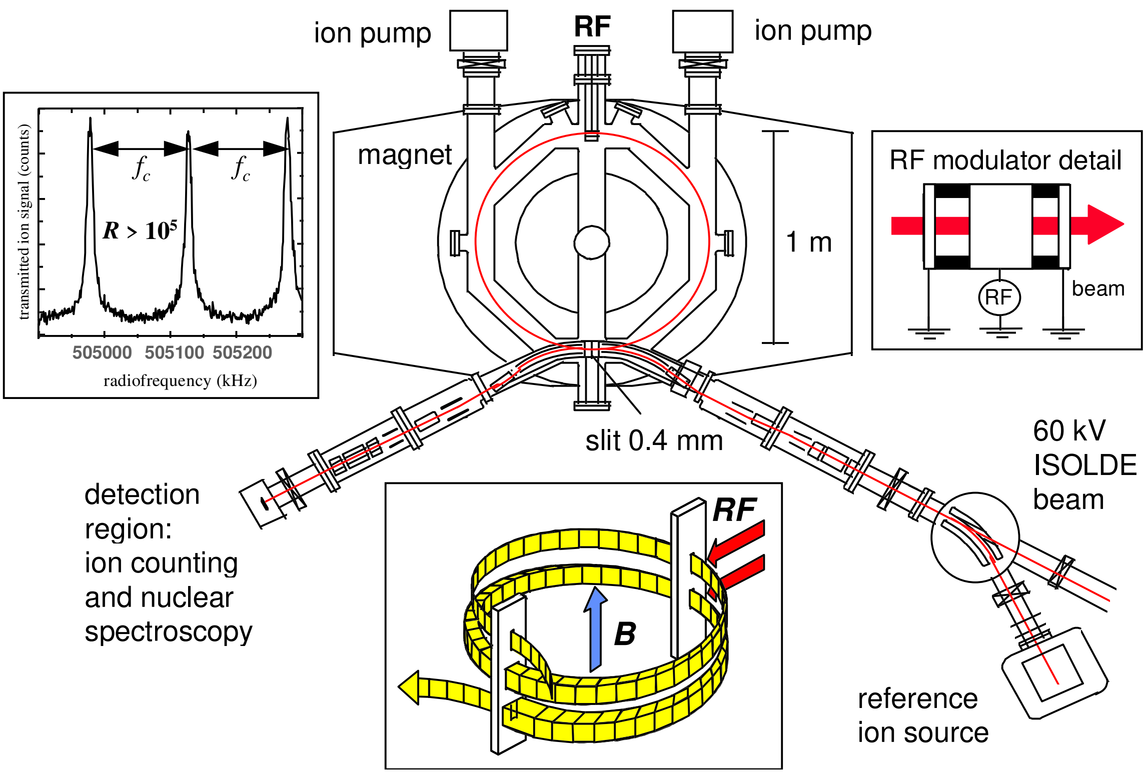

FIGURE 3. Layout of the *MISTRAL* spectrometer showing the nominal ion trajectory. Ions are injected from the *ISOLDE* beam line at the full transport voltage of 60 kV while the reference mass is alternately injected (without changing the magnetic field) at its required energy. Inset (right) shows the modulator electrode structure, the geometry of which is selected for a certain mass range. Inset (center) shows an isometric view of the trajectory envelope with the 0.4 mm injection slit followed by the first modulator at one-half turn, the phase-definition slit (of variable width to control resolution), the second modulator at three-half turns and finally the exit slit. Inset (left) shows the transmitted ^{39}K ion signal as a function of radiofrequency spanning three harmonic numbers (around 3400). The mass resolution here is about 50,000 but can be pushed to over 100,000 by closing the phase-definition slit.

A mass measurement is made when an unknown mass is alternately injected with a reference mass *without* changing the magnetic field. Comparing masses in this way requires changing not only the transport energy of the reference beam but the voltages of all electrostatic elements in the spectrometer (two triplets, eight pairs of steering plates, and two benders plus the injection switchyard bender). These comparisons are done in rapid succession (seconds) in order to eliminate short-term drift in the magnetic field.

In the case of short-lived isotopes (as well as elements with very rapid release times from the target matrix, such as Na) it is impossible to scan the entire required frequency range in time after the impact of the proton pulse. For each radioactive beam pulse, the ion transmission signal is recorded for only one radiofrequency point (determined randomly) and the resonance peak is reconstructed at the end. This point-by-point mode not only allows us to increase statistics in the peak but for each point, the ion signal may also be recorded with the radiofrequency switched off so that not just the intensity but the true transmission is measured in order to correctly normalize the peak. Moreover, since we count the number of ions in the beam that have been separated with very high resolution, we can produce very clean release curves and consequently, measure half-lives.

3. Summary of the first results from MISTRAL

The *MISTRAL* spectrometer arrived at *CERN* in May 1997 and was installed in the new *ISOLDE* beam hall extension. It was recording its first resonances only four months later. A first test run (5 shifts) using radioactive isotopes around $A = 27$ took place in November 1997 using a UC_2 target coupled with a plasma ion source. The spectrometer was able to cleanly separate the isobaric components with relatively good sensitivity and encouraging indications for measurement precision.

In July 1998, *MISTRAL* again took radioactive beam (13 shifts) from a UC_2 target but this time with a surface ionization source to get a clean beam of Na isotopes. This element is challenging since the release time from the target matrix is very fast. During this run the masses of $^{23\text{-}30}$Na were measured. The half-lives of the last three isotopes are all less than 50 ms.

Shown in fig.4 is a recorded peak for ^{30}Na from the July 1998 run. This measurement corresponds to a sum of 25 series of 64 (random) frequency steps each of which is triggered by the PS booster proton pulse with a period of (at least) 1.2 s. The center frequency is derived from a triangular fit which is the theoretically expected lineshape [25]. In this case, the frequency corresponds to harmonic number 2421 of the cyclotron frequency of ^{30}Na in the 0.3868 T field at a beam energy of 60 keV. The mass resolution in this case is 25,000 (reduced on purpose in order to favor transmission) but has been measured at over 1.2×10^5.

When we compare our preliminary values to those in the mass table we perceive an offset (proportional to ΔM, the mass-doublet difference) of about $7 \times 10^{-7} \times \Delta M$ as shown in fig.5(left). If we correct this offset, the residual differences scatter randomly about zero within a value of about ±25 keV, an uncertainty already better than the present one for ^{28,29,30}Na (80, 90, 90 keV).

This systematic error is due to a lack of congruency between the reference ion trajectory and that of the mass being measured. Ions of differing trajectories do not experience the same magnetic field because of residual gradients (~10^{-5}/cm).

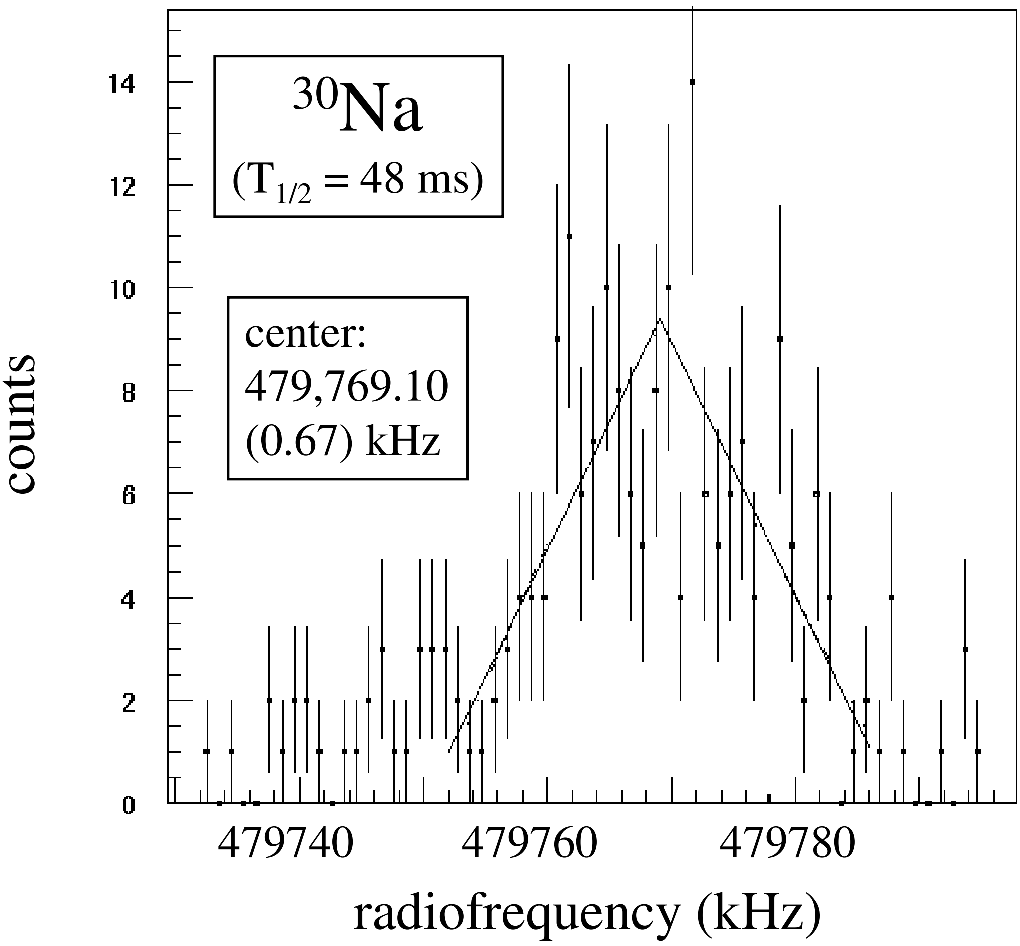

FIGURE 4. A recorded peak for ^{30}Na ($T_{1/2}$ = 48 ms). This measurement is a sum of 25 series of 64 (random) frequency steps each recorded after one proton pulse. The center frequency is derived from a triangular fit [25] and corresponds to harmonic number 2421 of the cyclotron frequency of ^{30}Na in the 0.3868 T field at a beam energy of 60 keV. The mass resolution is about 25,000 (reduced to favor transmission).

In November 1998, again using a UC_2(W) target (21 shifts), we remeasured $^{23\text{-}30}$Na but this time using two reference masses thanks to an upgrade of our high voltage network (sec.4). This way we are able to interpolate the systematic error to zero between the two reference masses as shown in fig.5(right). In cases where we used the same mass from *ISOLDE* and our reference source, the agreement is better than 3×10^{-7}. Using this correction method the masses obtained with *MISTRAL* can be compared to the mass table as in fig.6. The mass uncertainties for the isotopes further from stability are naturally dominated by statistical error.

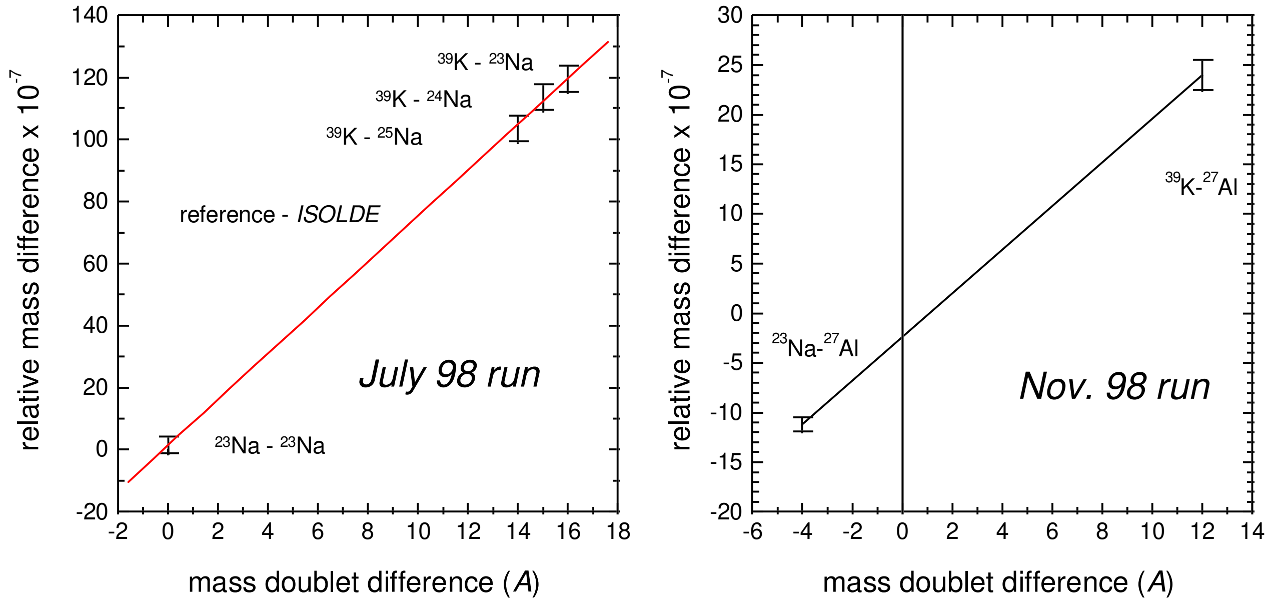

FIGURE 5. Evaluation of the systematic error of the *MISTRAL* measurements. Plotted is the mass difference of the measured value with respect to the mass table [35] versus the mass doublet difference in mass units (reference mass - *ISOLDE* mass). (left) Stable and well known isotopes with respect to the heavier reference ^{39}K. (right) ^{27}Al measurement with respect to both ^{23}Na and ^{39}K allowing an interpolation to zero mass difference giving a precision of about 3×10^{-7}.

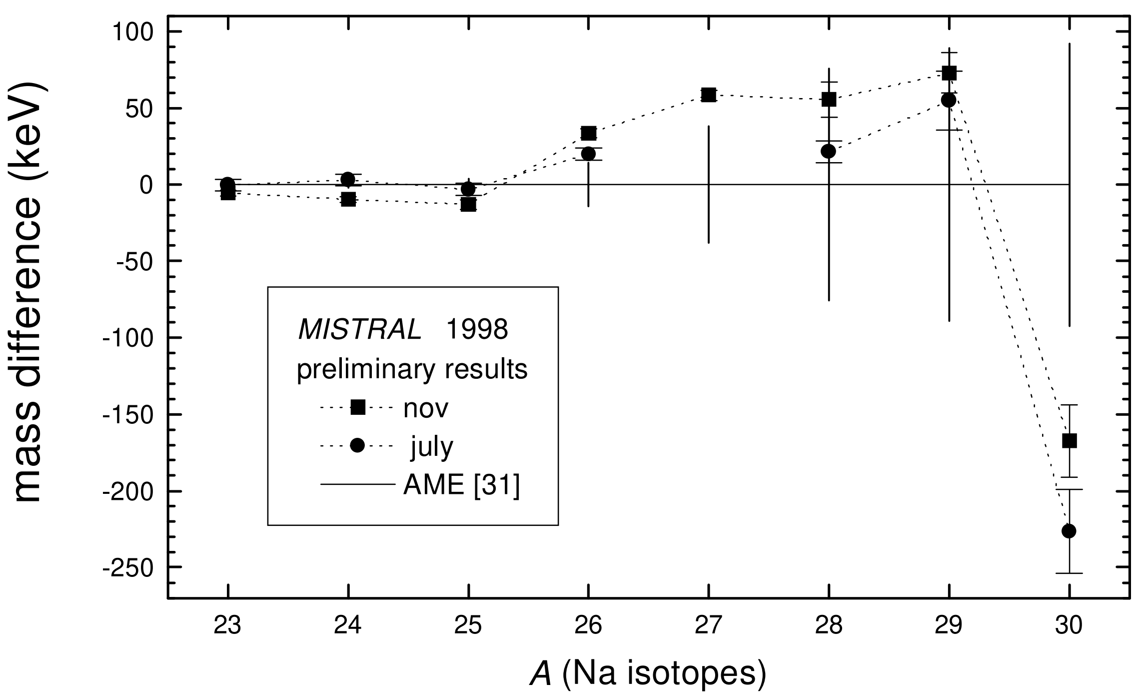

FIGURE 6. Preliminary mass values measured by *MISTRAL* with repect to the adjusted mass value from the Atomic Mass Evaluation [35]. In the July run there was a problem with the isobaric contamination of ^{27}Al such that ^{27}Na was incorrectly measured. This problem was eliminated in the November run with the stable Al mass being measured to within 0.3 ppm (fig.5). The deviation from the mass table - especially at A = 30 (about 7×10^{-6}) is present in both sets.

In the last run we also measured the masses of isobaric contaminants. At $A = 47$, we determined three masses: K, Ca and Ti, by optimizing the magnetic field setting for each. In the case of the ^{27}Na-^{27}Al doublet, the mass difference corresponded just by chance to one harmonic number but by changing the radiofrequency (and hence the relative cyclotron frequency difference for the harmonic numbers) we were able to perfectly resolve the two peaks. The mass value for ^{27}Al agrees with that of the mass table to better than 3×10^{-7}. We also measured the isotopes $^{39,41\text{-}47}$K and some isobars of Ca and Ti (see table 1 for summary).

isotopes	stable	unstable	total
$^{23\text{-}30}$Na	1	7	8
^{27}Al	1	0	1
$^{39,41\text{-}47}$K	2	6	8
$^{46\text{-}48}$Ca	2	1	3
$^{47\text{-}48}$Ti	2	0	2
total	8	15	22

TABLE 1. List of isotopes whose masses have been measured by *MISTRAL*.

4. Technical Improvements

In parallel with the installation of *MISTRAL*, the *ISOLDE* beam line was extended in a configuration determined by calculations made by our group (we also performed the calculations for the *NICOLE* experiment) but with a compromise in the *RC0/RC2* section of the beam line. Repeated tests using stable beam resulted in a very difficult beam transport (due to vertical mis-alignment) and a maximum transmission of only 50%. Not only is transmission an issue but the beam must be injected into the spectrometer through a 0.4 mm slit and must also be perfectly aligned with the axis of the spectrometer. Improvements are planned to cure the mis-alignment by providing successive vertical steering and a discussion is underway to remove the constraint in the RC0/RC2 section that causes the envelope of the beam to be too large.

Between the two runs in 1998 we upgraded the high voltage system of the spectrometer. This allows us to now operate the reference source at 80 kV enabling the use of a reference mass lighter than the unknown mass. This was crucial for using ^{23}Na as a reference for all the sodium isotopes and enabling the determination of the systematic error contribution caused by the residual field gradients (~10^{-5}/cm). We plan to reduce these gradients through the use of current shim coils [26] which will be installed in early 1999.

The important question of sensitivity is being addressed during the current shutdown when we will eliminate some beam loss in the exit line of the spectrometer with the hope of gaining between a factor of five and ten as a result. So far we have only managed a transmission of 5×10^{-5} from the *GPS* focal plane to the *MISTRAL* detector despite measurements of the *ISOLDE* beam emittance that projected much better. This situation is aggravated by the fact the spectrometer is very far downstream from the target (and from *GHM*, where the measurement was made). The emittance, though conserved, is nonetheless degraded by repeated foci and, especially, the two strong deflections *en route*.

For the longer term, this important question is being addressed by the development of a beam cooling device at the entrance of the spectrometer. By reducing the emittance of the incoming beams not only is the transmission through the many slits inside the spectrometer improved but both the reference and *ISOLDE* beam are “brainwashed” before they go through, forgetting their differing characteristic divergences, positions and energy spreads and increasing the congruence of the two trajectories through the spectrometer. Cooling a beam can be done several ways but the one technique that seems the fastest and most universal makes use of a light, neutral buffer gas. We plan to install a gas-filled ion guide that will use the alternate focusing of a radiofrequency quadrupole field to continually refocus the ion beam onto the axis while it loses kinetic energy from collisions with the gas [27]. The prototype is now under test in Orsay with a targeted installation in 2000. We are also collaborating with *ISOLTRAP* on the development of a device based on the same principle. Both experiments are members of the *EXOTRAPS* European Research (TMR/LSF) Network with beam cooling being one of its principle themes.

5. Proposed measurement program

With uncertainties of less than 10 keV for abundant isotopes and 25 keV for exotic ones, *MISTRAL* offers considerable improvement in measurement precision. Though preliminary, the sodium masses seem to reinforce the idea that the N = 20 shell effect is washed out as originally shown by Thibault *et al.* [15] even though the later measurements made with the *TOFI* spectrometer [5] found the effect to be attenuated. Two problems with this type of measurement which influence the results are: short-lived (μs) isomeric states and a very complicated calibration function. At *ISOLDE,* such short lived isomers - if even produced - would decay before diffusing out of the target. The very moderate resolution of *TOFI* is also conducive to large incertitude. Therefore, the new *MISTRAL* measurements would appear to revive this interesting question. Detailed analysis is now underway.

Looking at fig.2 we can see that one of the critical questions is the error bar on the $^{31-32}$Na measurements. Since the mass for ^{30}Na would already seem incorrect, it is imperative to verify the following two isotopes in addition to reducing the error bar (currently 160 and 480 keV). Clearly the error bars of the last Ne isotopes are too large (110, 300, 820 keV) and the values of these masses must not only be refined but also checked. Ne is particularly interesting as it would seem that the drip line intervenes at N = 21. Finally, there is some questionable behavior in the region of ^{33}Mg (150 keV uncertainty) and ^{34}Al (90 keV) as the two values almost overlap. Moreover, there is a large discrepancy between the *TOFI* and *SPEG* values for these isotopes - in both cases a 400-500 keV difference. A clarification of the mass values here is therefore necessary.

Since shell structure is considered a universal feature of nuclear structure, the same question can be raised at other magic numbers *e.g., N* = 28. Recent results from *SPEG* in this region (on isotopes of Si, P, S and Cl) [28] will give us a glimpse. But while this method is complementary to *ISOLDE* in its production mechanism, the uncertainty of these measurements is greater than 5×10^{-6} (about 300 keV) which does not put a very strong constraint on theory. Further measurements with *MISTRAL* will allow us to refine some of the results from *SPEG* and *TOFI* and perhaps to calibrate a systematic error that may be present.

Meanwhile, extensive activity is underway on other fronts including work at *ISOLDE*: *e.g.,* recent laser spectroscopy results on the quadrupole moments of $^{26-31}$Na [29] and also the envisaged Coulomb excitation and neutron-transfer reactions in these mass regions with *REX-ISOLDE* [30]. Recent Coulomb excitation results from MSU on ^{46}Ar [31] and ^{44}Si [32] do not provide evidence for an N = 28 shell closure and microscopic calculations by Werner *et al.*, [33] predict the breaking of the shell gap in this region whereas Cottle and Kemper [34] argue for its persistence.

Mass measurements, *e.g.* ^{49}K (uncertainty 70 keV), ^{50}K (280 keV, measured by *TOFI*), ^{47}Ar (100 keV) and ^{48}Ar (unknown), joined with these other complementary techniques will continue to provide important experimental input to accompany the vigorous theoretical activity in addressing these interesting questions.

6. Beam time request

MISTRAL has now obtained its first results which are of excellent quality and offer great encouragement for future experiments. However, the experiment must still be considered to be in the start up phase. The upcoming experimental program will be largely based on the plasma ion source which is a new step forward as we are extremely sensitive to the beam characteristics and this may make the initial beam tuning more lengthy. Furthermore, though we have now demonstrated the correct handling of isobaric contaminants, they will slow down the progression along an isotopic chain.

We would prefer somewhat longer, uninterrupted beam times due to the meticulous stable beam and trajectory set-up procedures that precede the measurements (section 4). However, this offers excellent possibilities for parallel users (*e.g.* in *GLM* and *GHM*).

For the program presented above we ask for 36 shifts - to be distributed over two or three longer beam times:

$N = 20$: Ne, Mg, Al isotopes using the ThC_2 or UC_2 target with a hot plasma ion source

$N = 28$: Ar isotopes using the ThC_2 or UC_2 target with a hot plasma ion source (naturally measuring any K and/or Ca isotopes present as well)

$N = 20/28$: Al, K, Ca isotopes and eventually $^{30\text{-}31}$Na using ThC_2 or UC_2 target with a (W) surface ionization source

For continuing systematic error evaluation and transmission improvement tests we ask in addition for 12 shifts of stable beam.

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