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Latest trends in the ever-surprising field of mass measurements

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Abstract. The binding energy of the nucleus, from its mass, continues to be of importance —not only for various aspects of nuclear physics itself, but for other branches of physics such as weak-interaction studies and stellar nucleosynthesis. The number of dedicated programs is increasing worldwide with recent results reflecting experimental achievements worthy of admiration. A brief description is offered of the modern experimental techniques dedicated to the particularly challenging task of measuring the mass of exotic nuclides and detailed comparisons are made in order to present future projects in a critical perspective.

PACS. 21.10.Dr Binding energies and masses

1 Introduction

Mass measurements have a noble (and Nobel) tradition thanks to the pioneering work of Francis Aston. The link between the nuclear binding energy and stellar nuclear synthesis (forged by Eddington, among others) also dates from this *époque*. It is difficult for new results from such a well-established field to be regarded as "hot topics" in the (popular) scientific literature. A recent headline heralded "Attogram mass measurements," performed by solid-state physicists at Cornell [1], who fabricated a nanometer-scale cantilever and measured its vibration frequency when loaded with a cluster of gold atoms. The achievement was qualified with an interesting statement: "To get any better measurement of mass you would have to vaporize the particle and shoot its constituent molecules through a mass spectrometer." [2] —exactly how our community makes it living[1]! By "our community" is meant nuclear physics where the bulk of atomic mass measurements is done principally because of our interest in exotic nuclei for which the binding energy gives us so much information about nuclear structure and decay modes. Masses of radionuclides are also very important in the interdisciplinary fields of weak interactions [4] and astrophysics [5]. Since there are so many more radioactive nuclides than stable ones and we still cannot really predict their masses, our community continues to prosper.

Measurements of the mass are among the most precise performed, as described by a recent article in the journal *Science* by a group at MIT [6]. In addition to the weighing of chemical bonds, a principal motivation of this work is an alternate determination of the fine-structure constant, α[2]. Such metrology often begs the question: what's the point? Apart from efforts at redefining *the* kilogram —the only fundamental standard still represented by an artifact [7]— recent astronomical observations [8] indicate a possible variation of α over time, something that no metric theory of gravity (including general relativity) allows. Theories aiming at the unification of quantum mechanics and gravity (such as string theory) in some cases predict such variation so that experimental limits should provide important constraints (see [9,10] for the latest on precision measurements and α's purported variation). Even the best theory of all, QED, needs to be tested and binding energies from precision mass measurements are starting to allow us to do that [11].

One idea that has considerably stirred our community is that chaos might prevent us from ever providing accurate mass predictions [12]. While this may seem like more of a question for theorists, it seems that models are still insufficiently accurate to establish a real limit (see contribution of Hirsch *et al.* [13]) so that measurements are still required to improve them to the point where this assertion can be verified.

The organizers asked for a review of the achievements regarding experimental mass measurements since the last ENAM conference in 2001 [14]. Having published a review article [15] on the subject in 2003, you would think that it was easy. As it turns out, the prolific activity in the field has conspired to make it a challenge with many new

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[1] Note that the goal of their work is to weigh viruses [3] —things we would never dare ionize and post-accelerate!

[2] D. Pritchard recently donated the MIT trap to Florida State University where this work will continue under the responsibility of E. Meyers.

results in only the last 1-2 years. Only a brief description of the different techniques will be offered here, in the same spirit as [15] to which the reader is referred (as well as the recent proceedings of APAC2000 [16]) for an exhaustive bibliography. Some detailed comparisons are made using the different methods and for examining performance and complementarity. (Near) future projects are then cast in their (exciting) perspective.

2 Measurement programs and context

Traditionally, we speak of two categories of mass measurements: so-called indirect techniques —reactions and decays— that produce Q-values, or energy differences; and direct (or inertial) methods of mass spectrometry where time-of-flight or cyclotron-frequency measurements of the exotic species are combined with those of well-known reference masses, ultimately linking them to ^{12}C (from which the mass unit is defined). The two canonical radioactive-beam production methods, fragmentation (or fusion-evaporation) of thin targets with in-flight separation (FIFS) and thick-target, isotope separation on-line (ISOL), previously offered a clear separation between the mass measurement techniques, namely time-of-flight for the former and cyclotron frequency for the latter. Also, while the in-flight approach is generally more sensitive, the ISOL-based method is generally more accurate. Thanks to the advent of gas cells and RFQ coolers, the best of both worlds is now possible. The high precision brought by holding an ion at rest in a Penning trap can now equally be brought to bear on ions born in fragmentation at relativistic speeds (as explained in [17]).

Mass measurement programs have been underway for many years at GANIL, GSI and ISOLDE. Very recently, ANL, MSU and JYFL have made their first measurements and realization is well underway at MAFF and TRIUMF. With TOFI (LANL) gone, SPEG at GANIL is now the senior program —and still very active. With fragmented projectiles, measurements of time-of-flight and rigidity are combined to determine the mass. Although the resolving power is modest, the tremendous sensitivity of their method allows them to reach the drip line for many light species. So SPEG is in an excellent position to study the migration of magic numbers [18].

Attempts have been made to improve time-of-flight measurements by lengthening the flight path using the many turns that result from injection of fusion-evaporation products into the CSS2 cyclotron [19]. Some difficulties were experienced initially but recently the technique (and corresponding analysis) has been improved and the newly-measured results and revised errors now provide good agreement in all cases (see discussion below).

The same idea of lengthening the time of flight can be realized in a storage ring, as with the ESR at GSI. Relativistic fragments are filtered through a mass separator and injected into the ring operated with a given rigidity where their masses can be measured two ways [20]. One is by detecting the so-called Shottky signal of a charged particle each time it passes an electrode and obtaining the revolution frequency from the Fourier transform. Since the fragmented beam has a relatively large velocity spread, it must be cooled. This is done with an electron cooler but the process requires several seconds [21]. The second method, used to measure short-lived species, requires operating the ring in isochronous mode where the revolution frequency is (to first order) independent of the velocity spread. In this case the ions are monitored in-beam with a thin-foil detector the the revolution frequency is derived from matching successive time signals [22].

An enormous volume of mass data has been produced by the ESR, spanning a sizeable portion of the nuclear chart. In 2002, they used the fragmentation of U to produce neutron-rich species that were measured with the two techniques [23,24]. Recently, their 1997 data was reanalyzed using all the time-correlation information available over the duration of the stored beam. The large mass harvest of heavy proton-rich nuclei out to the drip line was consequently extended and improved (thanks additionally to important α-decay links) [21].

The very drip line itself is a question of binding energy (or rather, its disappearance). We also know that for light, neutron-rich nuclides, halos manifest themselves at the dripline. The mass is a critical input parameter for halo models and due to the extremely small binding energies and very short half-lives, special techniques must be used. MISTRAL is a good example of such a technique. As a transmission, time-of-flight spectrometer using a radiofrequency "clock", its measurement technique is very fast and as it determines the ion cyclotron frequency, it is very accurate [25]. In 2003 MISTRAL measured the mass of ^{11}Li (a "superlarge" nuclide —see [26]) with an accuracy of 5 keV. What is interesting is that the halo binding energy has changed by more than 20% [27]. MISTRAL is located at the end of the mother of all ISOL facilities: ISOLDE, where a few meters of beamline separate it from ISOLTRAP, the mother of all on-line Penning trap installations [28].

ISOLTRAP has pioneered most of the methods now being used (mostly) for radioactive species elsewhere[3]. Starting with a gas-filled, linear RFQ trap, low-energy ion bunches are injected into a large-volume, cylindrical Penning trap for isobaric purification. The isobar of interest is retained and sent to the precision Penning trap where its cyclotron frequency is determined by measurement of its time of flight after excitation and ejection. During 2003, ISOLTRAP measured several masses of neutron-rich Ni, Cu, and Ga isotopes in an attempt to answer the question: is $N = 40$ magic? (for the answer, see [29]). In the course of those measurements, the triple-decker isomer ^{70}Cu was encountered, whose β-decaying branches had complicated spectroscopy efforts. By bringing the enormous reserve of resolving power to bear, ISOLTRAP was able to weigh each laser-selected isomeric state separately, resulting in unambiguous identification [30].

[3] Note that the original use of the Penning trap for precision measurements earned H. Dehmelt a share of the 1989 Nobel prize and that Penning traps had already been developed for mass measurements of stable species (see, *e.g.*, [6]).

Due to its superior precision, ISOLTRAP is able to contribute to the fascinating field of weak interaction physics. The comparative half-life (or Ft value) of a super-allowed beta transition gives us almost unhindered access to the weak vector coupling constant (one leg of V_{ud}, the up-down quark element of the CKM matrix). To determine Ft, the decay Q-value is needed (along with the half-life and branching ratio as well as nuclear corrections and the rate function, f). Nine such decays are sufficiently known to contribute to CVC and unitarity tests and ISOLTRAP has recently provided the masses so that *two* new points [31,32] can be added to this figure (see also [28]).

The superior performance of the versatile Penning trap has naturally triggered new experimental programs. The first "clone" of ISOLTRAP was SMILETRAP located at the Manne Siegbahn Laboratory in Stockholm, generally dedicated to stable species but in high charge states. The next project was the Canadian Penning trap and after a difficult early life with its excommunication from Canada, is now in full-fledged operation [33,34]. It is the first instrument of its kind making use of "the best of both worlds"; the advantages of high energy reactions and low energy precision apparatus —linked by a gas cell (see [17]). Their first success was ^{68}Se, established as a waiting point of the putative rapid proton-capture process, thought to power X-ray bursts [35].

The newest arrival is JYFLTRAP in Jyväskylä, a two-in-one design meaning that the isobaric separator trap and precision hyperbolic trap are located in the same magnet (inspired by SHIPTRAP, see below). The great advantage of JYFLTRAP is the host of neutron-rich refractory elements made available by the IGISOL technique, insuring a *chasse gardée*. In the course of commissioning the isobaric cooler part of JYFLTRAP, new masses of Rh [36], Ru [37] and Zr [38] nuclides were measured (some for the first time). Now the precision trap has been brought into the battle with impressive results (see [39]).

Also reporting exciting prelminary results at ENAM04 were the LEBIT facility at MSU [40] and SHIPTRAP at GSI [41]. Both are new-generation instruments reaching for the best of both worlds by trapping species that issue forth from a gas cell with LEBIT trapping the products of fragmentation reactions and SHIPTRAP, those of particularly heavy-ion fusion-evaporation (eventually seeking trans-uranium elements).

Finally, MAFFTRAP [42] will enjoy the copious production rates of neutron-rich species offered by thermal neutron-induced fission using the FRM-2 reactor, now operating near Garching and TITAN [43], a Penning trap system being built at TRIUMF in Vancouver, will be the first installation to use high-charge states of radioactive species (bred in an EBIT) to achieve higher accuracy (*i.e.*, higher cyclotron frequencies) with shorter trapping times.

3 Comparisons

To compare the performance of the various techniques a composite plot of experimental uncertainty *vs.* (weighted) isobaric distance from stability was offered in [15]. The same figure is presented here (fig. 1) (the same axes and definitions are used for the sake of direct comparison with fig. 6 in [15]) for results published only since [15] went to press. The number of measurements (217) that have appeared in the last 1-2 years is remarkable. Overall, the results show that most of the various techniques have been improved in that lower uncertainty (and in some cases, better sensitivity) has been achieved.

While offering only modest uncertainty, SPEG does offer the mass values that go farthest from stability. The newest SPEG results [18], though they did not reach further than [44] (those data included in fig. 6 of [15]), have seen their precision —and very likely, their accuracy— improved. The new results conform to the systematic extrapolations of the recent Atomic Mass Evaluation [45] whereas the earlier results did not, a *faux pas* that resulted in their exclusion from that work. The same is true for the CSS2 values that now, although having lost a little on the precision axis, now provide reliable results (see below).

The ESR97 data were analyzed using time dependence —an important consequence of the storage ring technique that offers a dynamic profile of the beam. Not only could more mass values be derived but the precision was considerably improved —up to a factor of five in many cases (Note that although hundreds of masses were measured, the values shown here correspond to only the 75 new masses not produced from α-links.) A smattering of proton-rich masses measured using the IMS technique were recently published [22]. Though more modest in uncertainty, the masses in question were farther from stability. A larger IMS data set, of fission products obtained from fragmentation of a U beam, has appeared since [23]. Though not shown here, these preliminary results show a relative uncertainty of roughly 10^{-6} and are of particular interest in light of mass-model predictions for neutron-rich nuclides.

New on this figure for MISTRAL is the result for the very short-lived, drip-line nuclide ^{11}Li [27]. This light nuclide is one of the extreme points on the isobaric axis and the excellent precision offered by MISTRAL makes it a valuable tool in the overall quest for mass data.

There are two newcomers to this figure —both from the North: the Canadian Penning Trap (now at Argonne) and the Finnish Penning Trap in Jyväskylä[4]. Measured back in 2001, the CPT ^{68}Se mass was only recently published [35] due to the obligations of long consistency and error checks. The achieved uncertainty has already improved with more measurements [46]. The JYFL trap results come from on-line tests mentioned above, using the cooler trap [36,37,38] and not the precision trap, however mass measurements using both traps have now been made [39]. The power of traps is nicely illustrated here with the latest cooler trap results showing a similar precision with those of cooled ions in the storage ring.

Last (and least —in terms of experimental uncertainty), ISOLTRAP. Not only has there been an impressive harvest in the past 18 months (77 values here, in-

[4] Both also have a common ancestor in ISOLTRAP.

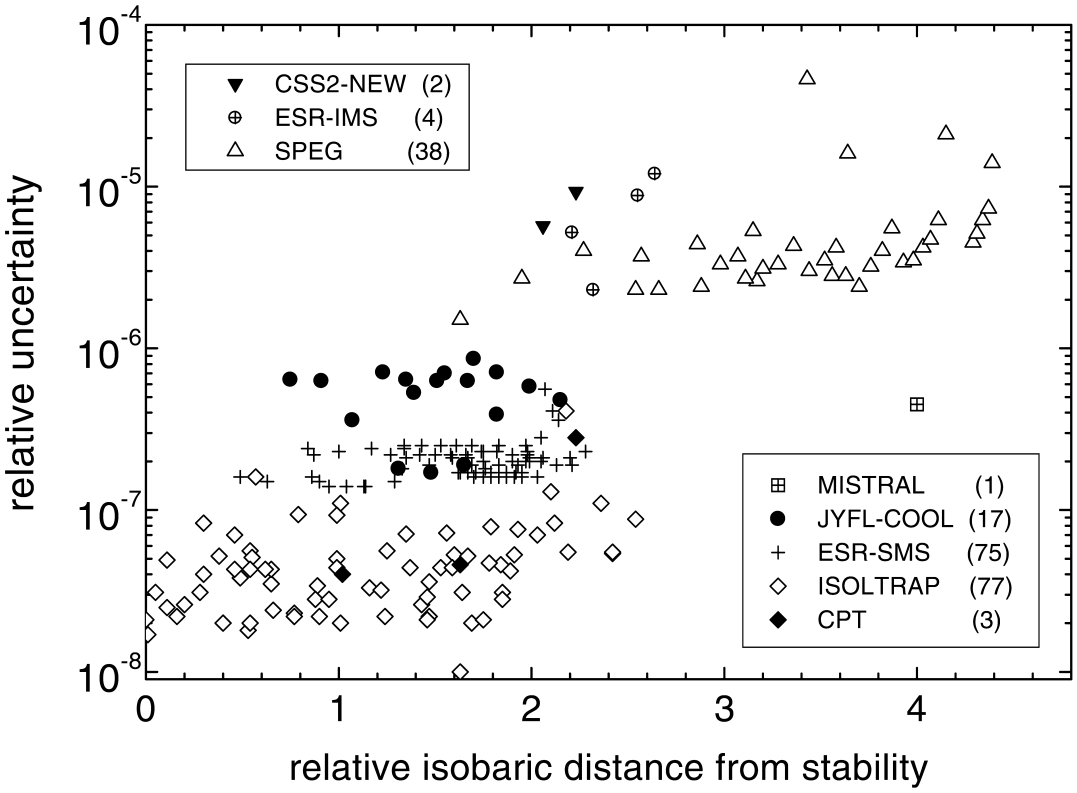

Fig. 1. Experimental uncertainty *vs.* (weighted) isobaric distance from stability (as in fig. 6 of [15]) of new mass results published in only the last two years. Data from SPEG: [18]; CSS2: [47]; ESR-IMS: [22]; ESR-SMS: [21]; MISTRAL: [27]; CPT: [35,46]; ISOLTRAP: [28,29,31,32,48].

cluding some improved masses even for stable nuclides), but thanks to the pioneering error survey using carbon cluster ions [49] the uncertainty is routinely at the 10^{-8} level. Here, even stable nuclide masses can often be improved. Comparison with fig. 6 in [15] is striking since the majority of those earlier ISOLTRAP measurements sat at the 10^{-7} level, corresponding to the overly conservative systematic error addition. The enviable sensitivity of ISOLTRAP —only a few hundred ions per second are necessary to achieve such an uncertainty— combined with the enormous reserve of resolving power, enable the measurement of masses very far from stability. For details and references of these measurements, see [28].

Like any type of measurement, masses can be determined inaccurately, meaning they are wrong —even if repeated determinations with the same apparatus give the same results (*i.e.*, high precision). Due to the high accuracy inherently required for masses, they are particularly prone to systematic errors. Aside from making detailed error surveys and consistency checks, an excellent test comes when comparing results for like masses from different techniques. In [15], several such comparisons were offered and on the whole, the various methods were quite consistent. One exception was CSS2 for which three deviating results had been published [19]. The reason for citing this example is by no means to castigate the CSS2 collaboration but simply to show that their story has a happy ending: measurements with other techniques enabled a re-evaluation and improvement of their technique, deriving a more realistic experimental uncertainty [47]. Shown in fig. 2, the recent measurements of ^{68}Se and ^{80}Y are now in complete agreement with those in the literature. Also shown in fig. 2 are the very recent cases of $^{94-95}$Kr from ISOLTRAP [50] and the ESR [23] as well as ^{22}Mg and ^{22}Na from CPT [46] and ISOLTRAP [32]. Note that the latter are perhaps the most accurate on-line measurements ever made with radioactive nuclides (note the mass difference scale over a thousand times smaller than the first case).

It is worth mentioning again that all methods rely on the availability of reference masses. This gives an inherent complementarity to all of the techniques described here in that the more accurate measurements calibrate the ones that are farther from stability. In many cases for example, MISTRAL and ISOLTRAP results have been used to calibrate ESR and SPEG measurements.

4 On (and beyond) the horizon

We have looked at the established mass measurement programs and what they have spawned. The Penning trap has had an enormous influence in the field. Dominating most of the performance categories, it has also proved versatile

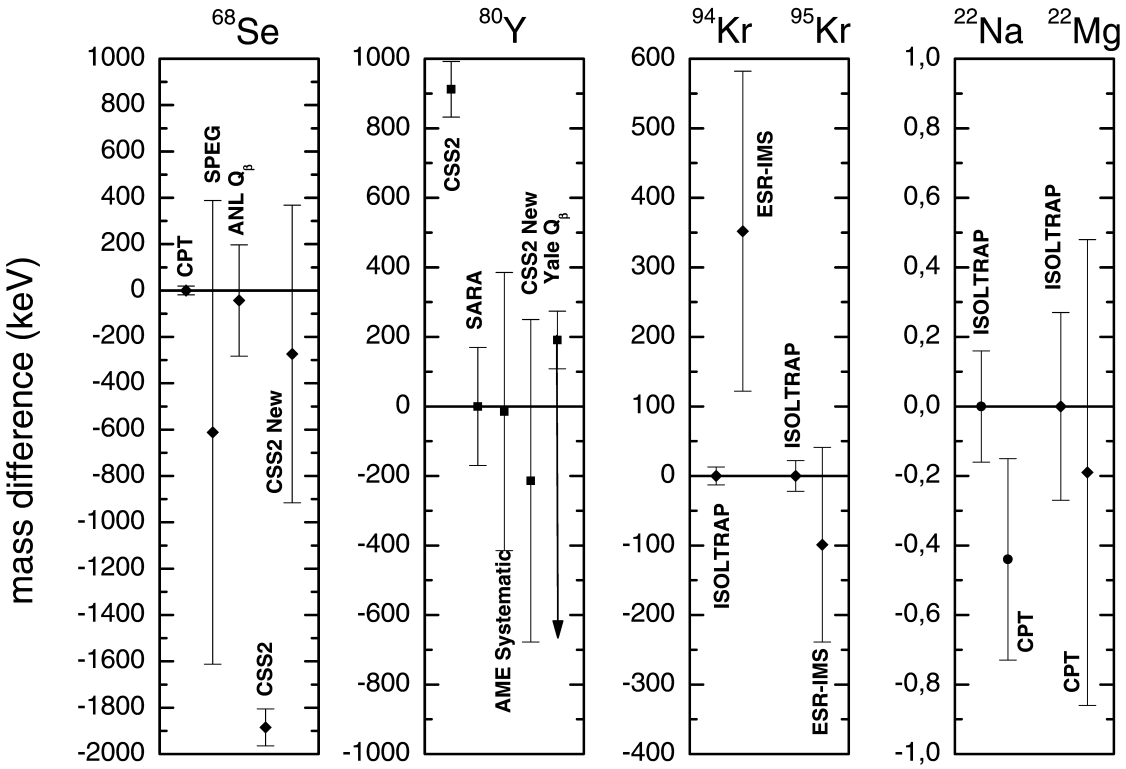

Fig. 2. Comparisons of the masses of different nuclides determined by different techniques. Note the change of overall scale from 3 MeV to 2 keV. Respective data points for ^{68}Se from [35,51,52,19,47]; ^{80}Y from [19,53,54,47,55]; $^{94-95}$Kr from [50,23]; and ^{22}Mg and ^{22}Na from [32,46].

enough to be found at the heart of practically every new measurement program.

After the Penning trap, is there room for another type of mass spectrometer? The answer is yes. The trivial reason is that there are so many nuclides for which masses will need to be measured that a veritable battery of techniques is necessary. For the moment, the only weakness of the Penning trap is in the serial time-of-flight scheme currently utilized which is somewhat inefficient (even impossible for such cases as superheavy nuclides) so that large areas (*i.e.*, spanning several Z values) require a lot of effort. Already this is being remedied with the development of the Fourier Transform (FT-ICR) technique that is non-destructive so that a complete measurement is possible with only one rarely produced ion (see [56]).

Two other techniques are worthy of mention here: the use of electrostatic mirrors [57] and so-called "household appliances" [58]. These schemes offer an attractive alternative: masses of nuclides far from stability (*i.e.*, short half-lives) with decent accuracy and moderate cost and effort. Of course, nothing associated with exotic nuclides comes cheap and easy but the impressive feats of the Penning trap came only with vigorous effort, sustained over many years.

Amongst the myriad applications of mass measurements, perhaps the most demanding is that of nuclear astrophysics. There, the need is for masses as far as possible from stability, almost regardless of the attained precision. Even a rudimentary mass value —provided it is accurate within the associated uncertainty— can give significant insight into the associated nuclear structure. But the key point is that these values far from stability not only provide the greatest test for nuclear mass models but are also used as diagnostics for their improvement and evolution (see [5]).

The enormous harvest of masses from the ESR has proved particularly valuable in this regard. For this reason (and for nuclear structure itself) the future plans of the FAIR facility at GSI are important to mention [59]. In addition to a low energy branch (which will, naturally, include a Penning trap) new storage rings are planned, one for Schottky-type measurements (with another for faster, stochastic pre-cooling) and one for isochronous measurements. The production rates at this facility indicate that new masses will number in the thousands!

Though tremendous improvements in experimental sensitivity and production techniques appear promising, the masses of many exotic nuclei of interest will certainly remain unmeasured for many years to come, leaving no choice but to resort to theory. Thus, the interplay between theory and experiment is crucial and new measurements far from stability are now used as diagnostics in the development of new microscopic mass models. If extrapolation to the drip lines remains an existential issue, at least a veritable articulation now exists between theory and experiment.

I thank my fellow mass measurers for having produced so much new data in the short time since [15] and making me sweat profusely trying to present it all.

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