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Present knowledge of atomic masses

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Abstract. Progress is reviewed of available data for evaluation of atomic masses since the 1995 one. Many more direct mass measurements have become available, especially in the region of proton-rich nuclides. The number of alpha- and, especially, proton-decay data increased considerably. In the region of superhigh mass numbers, many very interesting new observations were made.

PACS. 21.10.Dr Binding energies and masses – 32.10.Bi Atomic masses, mass spectra, abundances, and isotopes

1 Units. Fundamental masses

Most recent progress in the knowledge of atomic masses has been in the region of "exotic" nuclides, that is in regions far removed from the line of β-stability in a diagram against atomic number of, *e.g.*, neutron excess, or with atomic numbers above about $A = 250$. The precision reached in the former is not better than a few tens of keV, compared with a few keV or even far better near that line.

Yet, let us start with data in the latter region. Then, in the first place, a significant progress in precision has been made in the knowledge of values for fundamental physical constants [1]. Most important for our purpose is the relation between the usual units for mass and energy. No remark is required for the first unit, 1/12 of the atomic mass of ^{12}C in its ground state. But the energy unit, the electronvolt, requires a comment. In addidition to the "international" volt, use is made of a unit V_{90}, the "practical" volt, defined by adopting a values for the constant in the Josephson relation between voltage and frequency. At the time of our last mass evaluation [2] the ratio between mass unit and the latter energy unit was known with a precision of 90 ppb, with the former with as much as 300 ppb. Though this precision was important for the treatment of few items, we then yet decided to use the "practical" eV as unit. But in the mentioned 1998 evaluation of values for fundamental constants, the precisions were improved to 8 and 40 ppb. The difference is not important for our purposes, more so since the ratio between the two differs only 4 ppb from unity. The only exception occurs for the light hydrogen isotope: the reported error in its mass excess in mass units is 15 ppb! We therefore plan still to use the "practical" eV in our next mass evaluation.

A remarkable consequence of the present small error is, that now the relative errors in the mass excess values in mass units and in energy units are no longer significantly different, even for the stable hydrogen isotopes. In our 1995 evaluation, their errors for energy units were twice as large as those for mass units.

The new values for the fundamental constants are of some importance too in deriving values for reaction energies from measurements of voltages, magnetic rigidities, flight times or γ-ray wavelengths. When making corrections for some results of very precise older measurements, one of us (AHW) became unsatisfied about the use of many values recalibrated by earlier authors, of course using then current values for fundamental constants. He therefore undertook the somewhat laborious task of checking these cases and to revise them where judged desirable.

In this work, precise values for masses of some fundamental atoms play a role. And progress was made here too, due to new measurements with precision Penning traps in Uppsala [3], Seattle [4] and Cambridge-Massachusetts [5]. It is worth mentioning, that it then appeared that the new mass values for the stable helium isotopes differ somewhat more from the previous ones than their error estimates.

The mass of the neutron follows from those of the two stable hydrogen isotopes in combination with the energy of the γ-rays emitted in the capture of thermal neutrons in hydrogen. Kessler and Deslattes [6] made a new, very accurate measurement of its wavelength by crystal refraction. The result again deviated somewhat

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more from the earlier value than its adopted error. The error is now so small that, other than earlier, the errors in nuclear binding energies do not differ significantly from those in mass excesses.

2 Backbone

Precision Penning-trap measurements have also been made of some other nuclides along the line of β-stability. Thus, for example, the mass of the ^{133}Cs atom was measured [7,8] and is now known with a precision of 22 eV. The new value is 5 keV higher than the one we gave in our earlier evaluation, to which an error of 3 keV was assigned. Very precise values have now also been reported for ^{23}Na, ^{85}Rb, ^{87}Rb [8], ^{36}Ar [9] and ^{76}Ge and ^{76}Se [10].

For early mass-spectroscopic results, which mostly formed overdetermined sets, we found in their least-squares evaluations that, as a rule, the assigned errors were underestimated by, mostly, 50%. We took this into account in our evaluations of their combinations with one another and reaction and decay energy results. The just mentioned Penning-trap results also form an overdetermined set. We were pleased to find, that for them the consistency factor did not differ significantly from one.

Unfortunately, the most urgent problem for the backbone has still not yet been solved. We showed 20 years ago [11], that the reported mass spectroscopic values for masses of stable Hg isotopes deviated some 20 keV, about 10 times the reported accuracy, from the values derived with the same method for lighter W and Os isotopes, and for heavier Th and U ones, combined with measured nuclear-reaction energies. New measurements under way since a few years have not yielded clues for solving this problem yet.

3 Mass-spectroscopic results for exotic nuclides

New measurements in Los Alamos, using flight time measurements on reaction products, were mentioned at ENAM98 [12]. The authors were so kind to give us a list of the resulting useful mass values of nuclides from ^{44}Sc to ^{77}Zn, with precisions of the order of a few times 100 keV. It is a pity that no discussion of them yet appeared in open literature.

Also announced at ENAM98 [13] was, that GSI started making measurements with, essentially, the same technique —though using a far larger instrument. Two sets of measurements were published [14,15] giving data from ^{114}Sb to ^{201}Po, with precisions of, in some cases, as small as a few tens of keV.

The Mainz-ISOLDE group continued their measurements with a Penning trap. New data, with a precision only slightly worse than 10 keV, became available for nuclides from ^{114}Xe to ^{154}Gd [16,17]; and from ^{182}Hg to ^{203}At [18].

All these measurements were made with resolutions insufficient to separate isomers, with a few exceptions. And in checking this feature, the authors found some surprises. In measurements with a time resolution of some 8 seconds, one does not expect to see isomers with ten times smaller half-lives. Yet, the GSI group [15] observed the isomers in ^{149}Dy and ^{151}Er, with excitation energies of $2\frac{1}{2}$ MeV, and with reported half-lives of about $\frac{1}{2}$ s! But these half-lives refer to neutral atoms. The measurements, however, were made on fully stripped nuclei. And because of the large conversion coefficients of the relevant isomeric transitions, these isomeric nuclei in their stripped states live long enough!

Least-squares evaluation of combination of these new mass-spectroscopic results with decay energies, discussed below, did not indicate a necessity for correction to their errors as mentioned above.

The total result of these measurements is that mass values for proton-rich nuclides are much better known than before.

4 The NUBASE evaluation; corrections to measured isomer mixtures

Already since long, we maintain an "M-file" of approximate mass values for atoms in ground states and in selected isomeric states as input in our computer programs (which, essentially, calculate the differences with the input values). One reason is that, where isomers occur, one has to be careful to check which one is involved in reported experimental data, such as β- and α-decay energies. Cases have occurred where authors were not (yet) aware of isomeric complications. Examples are mentioned below. For that reason, our M-file contains known data on such isomeric pairs (half-lives; spin-parities; excitation energies).

The matter of isomerism became even more important, as just mentioned, for mass-spectroscopic measurements with insufficient resolution for separating isomers. One then obtains an average mass for the isomeric pair. A mass value for the ground state, our primary purpose, can then only be derived using additional information for the isomeric excitation energy and the production rates of the two isomers.

We therefore judged it necessary to extend our M-file to include all known isomers. This turned out to be a major job. And since it was judged possible that the result might be useful for others, the resulting "NUBASE" file was published [19].

In cases where the excitation energy is not known, it must be estimated. Different pieces of information may exist. It may be that information on measured isomeric γ-rays, though not yielding a complete decay scheme, yet gives a lower limit for the excitation energy. If not, it may be known that one of the isomers decays to the other, giving a lower limit $E_{\rm exc} = 0$. Then, it may be that similar isomers are known in isotopes or isotones of the considered nuclide. Extrapolation of their known excitation energies may give an acceptable estimate. In some cases, theoretical considerations may be used; *e.g.* that the Gallagher-Moszkowski rule predicts one of the

combination of shell model levels to be lowest. Quite often, the uncertainty of such an estimate is not large compared with the error in the mass-spectroscopical result for the mixture, allowing the derivation of an acceptable mass value for the ground state.

An extra complication, though, is that the intensity ratio of the isomers in the measured mixture is often not known. In that case, we assume that the probability distribution for the upper isomer is flat between 0 and 1. Again, the resulting precision for the thus derived ground-state mass is often not much worse than that of the measured average mass.

5 The α-decay chains

Measured α-decay energies in such chains yield often quite precise information about differences in the masses of their members. Fortunately, new information on α-decay is still regularly reported, by laboratories in Finland, Germany, Japan and the USA.

For nuclei with both an even number of protons and of neutrons, the transition between ground states is always the most intense one. Except for the very small number of cases, where this transition was later found to occur below a strong transition of another nuclide, these cases give certain information about mass differences. For odd-mass number nuclides, one can also often trust this when the final state is known to have the same Nilsson model assignment as the mother nuclide. This mother, though, may be an upper isomer!

If excitation energies of final states (or that of the mother isomer), are known experimentally, one can again derive a good value for the mass difference of the ground states. Combination with results on γ-rays, preferably in coincidence with α-transitions, may show this.

But for other cases we have made long ago a systematic analysis of a number of them, comparing them with good cases in their vicinity. The idea was that this might lead to the suggestion of an average energy for the final states in the decays with the highest observed α-energy, which could then be used to get a reasonable estimate of the ground-state decay energies. The result was, though, that the best choice for this average energy was zero. Instead, we took care that in such cases 50 keV is then root-sum-squares added to their errors. Adding labels "O" to our input file makes the computer take care of this.

In many cases, such decays occur between nuclides for which, earlier, only estimated atomic mass values were available. We then did not feel a necessity for a careful analysis and therefore just labelled them O rather freely. But now, mass-spectroscopic results, sometimes known for several members of separate α-decay chains, can help to suggest whether observed α-decays feed ground states. We have now analyzed several of such cases more carefully. As a result, we have withdrawn adding the extra 50 keV for several cases. Yet, it does not fully eliminate this problem, as long as the errors in those mass measurements are rather larger than those in reported α-ray energies.

It is interesting to note, that several of the new measurements have shown that α-decays earlier assigned to ground states belong in reality to upper isomers.

6 Mass values derived from systematical trend. Thomas-Ehrman effect

Already since long, we supplement our tables of mass values from experiment with values derived from experimental trends. The original reason was, that many β- and α-decay energies, even chains of them, were known between sets of nuclides, none of which had an experimentally known mass; and we thought the information on masses on such cases to be too valuable to be omitted. But for the purpose of our NUBASE evaluation, we now think it useful to have mass values for all nuclides occurring there.

Earlier, we used only extrapolations from experimental values for binding energies of two neutrons, of two protons, and decay energies in α-decay and double-β-decay. These were chosen since they are little influenced by pairing energies between the last added two nucleons. Lately, we also used extrapolation of differences with simple mass formulas. Mass values derived these ways were duly indicated in the table.

The same methods were also used to locate experimental data badly different from expectation, and therefore probably incorrect. Our experience has been that newer data mostly confirmed our suspicions. We [2] dutifully published both the suspected value and our estimate.

Special extrapolation methods were used for very heavy nuclides ($A > 250$), and for light ones in the region where mass values were known for at least one nuclide with $N < Z$.

One way of there estimating masses of proton-rich nuclides is based on charge symmetry relations. Charge symmetry compares masses at the same mass number. For odd-mass numbers, one then has to take into account the difference in pairing energy between two protons and two neutrons. Values for this difference are given by Jensen, Hansen and Jonson [20]; but for light nuclei, a better estimate can be derived from a study of the β-decay energies between mirror nuclei with $\frac{T=1}{2}$. But the used charge symmetry relations automatically take care of this feature.

Another set of symmetry relations, making use of combinations of such β-decay energies for neighbouring mass numbers, is used by Comay, Kelson and Zidon [21]. The differences with charge symmetry are of the order of 50 keV; rarely above 100 keV; those of both with experimental values, where known, are of the same order. Both are true with exception of experimental values affected by Thomas-Ehrman shifts.

These shifts, occurring for proton-unstable nuclides, cause the real mass value to indicate more stability than that from extrapolation. This effect has been studied by Comay *et al.* [21]. One of us (AHW) updated this study and agrees that it indicates that for the light nuclides studied ($A < 20$) the shift is 1/3 of the proton decay energy

value following from the extrapolation. But he also compared results of such extrapolations with more experimental masses, not only proton unstable ones, and with values obtained from isobaric analogue analyses, see below. This evaluation rather strongly suggests, that, anyhow for heavier nuclides, the shift is rather smaller than the mentioned estimate.

7 The proton drip line for A = 0–100; isobaric analogues

A source of information on masses of light proton-rich nuclides is knowledge of masses of isobaric analogues of their ground states. Such masses were already early [22] found to agree rather nicely with the isobaric analogue mass equation IMME, a quadratic equation between mass values for states with the same isobaric spin T. In several cases, a mass value for the most proton-rich member derived from IMME can be given with a smaller error than that in the direct experimental value, or even where no experimental mass value is known. A decided advantage of this way of extrapolation above the two methods mentioned above is that here experimental data are used not yet used in the determination of other ground-state mass values.

Recently, Herfurth *et al.* [23] found that their new mass-spectrometric value for ^{33}Ar did not agree with IMME. Yet, the difference with its mass as calculated from IMME is only about 18 keV. In an evaluation, several years ago, one of us (AHW) had already decided that it is wise to increase errors as following from IMME. For the most proton-rich nuclide with isobaric spin $T = 3/2$ (or 2), the estimate was to increase them by root-sum-squares adding 20 keV for the most cases. This new result, thus, is no reason for a change.

In our 1995 mass table [2] we give, as said, a list of experimental mass values which in our opinion can better be replaced by given values derived from systematic trends. We might next time consider to add to this table cases for which IMME gives mass values to which lower errors can be assigned.

Recently, there has been progress in deriving, from studies of delayed proton emission, masses of $T = 5/2$ IAS levels in nuclides with $T_3 = -3/2$. From all their IAS sextuplets, the mass of the most neutron-rich item, $T_3 = 5/2$, is known. But only for $A = 39$, 43 and 47 one more IAS mass, that of the $T_3 = 3/2$, is known, so that common IMME can be used to derive mass values for ^{39}Ti, ^{43}Cr and ^{47}Fe. For lighter cases, some authors earlier helped themselves by using the estimates of Anthony *et al.* [24] for the relevant Coulomb-energy difference. This is somewhat questionable, since these estimates do not agree with the difference with the experimentally known masses for $T_3 = +5/2$ items.

In their recent work on ^{31}Ar, Axelson *et al.* [25] instead derive a value for the mass difference with its isobaric ^{31}Cl analogue from extrapolation of known Coulomb-energy differences between the isotopes of this pair with mass numbers 32, 33 and 34, that is with $T = 2$, 3/2 and 1. They use for this a formula earlier discussed by Jänecke [26]. Comparing values, obtained by appling the same method to several more cases, with the results obtained from the two extrapolation methods mentioned leads to a preference for the Jänecke-type results.

8 The proton drip line for A = 100–220

The new mass-spectroscopic results, especially when combined with the data on α-decay chains, yield quite interesting information about the course of binding energies for proton-rich nuclides. Another valuable contribution is the study of proton-decay energies of very proton-rich nuclides. The most recent case is ^{117}La [27], from the groups that published much of this kind of information, from here to ^{185}Bi. For ^{117}La, and several more cases, two proton-emitting isomers were found for the same nuclide. The properties of proton decay then often did not only permit to determine the isomeric excitation energy, but even to get information about their spins and parities. And combined with α-decay measurements on these nuclides and their daughters, information has been obtained about several more isomers and their excitation energies.

The resulting position of the proton drip line is of course valuable. But the new information on the energetical consequences for the pairing of two protons, two neutrons and a neutron with a proton is most interesting. Of the first two, we mention only that the formulas of Jensen, Hansen and Jonson [20], who predict a decided dependence on the neutron excess N-Z, represent the data rather satisfactorily. The data on neutron-proton pairing also agree not badly. But a closer scrutinity indicates a curious dependence on magic numbers. Thus, the semi-magic number $N = 64$ seems to cause a jump in the proton- neutron pairing energy, anyhow if Z is also nearly magic, near $Z = 50$.

Finally, about the Thomas-Ehrman shift. If as large as for light nuclei, it would give a quite observable deviation in the course of proton separation energies, up to more than 0.5 MeV for the observed most proton-unstable isotopes. No such shift is seen; it seems to be at most 1/3 of the value found for cases with mass numbers below $A = 20$, discussed above.

9 The superheavy nuclides, A > 250

Not important for the mass work, but interesting to notice, is the matter of the names of the very heavy elements. In our 1995 evaluation we mentioned, that the International Union of Pure and Applied Chemistry proposed a set of names for the elements 103–109. We used them also in NUBASE. Somewhat unfortunately, the names finally adopted [28] were different for several elements; we will of course use the new ones in the future. No names were proposed already for the elements 110 and 111. Since then, a further new element, $Z = 112$, was discovered at GSI [29],

with mass number 277. A very remarkable fact was that in the two decay chains reported, atoms of the daughter, ^{273}110, occured after delays that were a factor 1000 different. Also, the most delayed one had a very significantly (1.3 MeV) lower α-energy. This points to the influence of a new semi-magic number of neutrons, $N = 162$.

The Berkeley announcement [30] of the discovery of the nuclide ^{293}118, decaying to nuclides of the also new elements $Z = 116$ and 114, was very exciting. Rumours are circulating, though, that new experiments both in Darmstadt and in Berkeley, yet fail to confirm these results.

Newer data on isotopes of the two lighter of these three elements have been reported in Dubna [31,32], from bombardments of heavy actinides with ^{48}Ca. At least some of them are somewhat less certain. Thus, heavy criticism was raised by Armbruster [33] against some Dubna data.

We nevertheless plan to present these data in our tables. This causes no trouble because no conflicting data necessitate a choice. But we will warn our readers that the mass values presented in this region may be rather uncertain. The same is of course true for the mass values of intermediate nuclides in this region that we obtain by interpolation.

The long chains of α-decays in this mass region present a difficulty. Again, the excitation energies of levels fed by the observed α-rays are often not known. As said, the prominent decays for odd-A nuclides are regularly those to states with the same Nilsson model quantum numbers as the parent (favored α-decays). For the lighter nuclides in this region, differences between the positions of such particle levels are often known for isotones or isotopes; and they often do not change drastically as a function of N or Z. We made a study of them, which allows us to make decent estimates for relevant excitation energies of states fed in favored α-decays. But such information is not available for many heavier nuclides. Neglecting this point would lead to progressively too low mass estimates, so we have to do something.

For this purpose, we study the systematics of α-decay energies. It is then very pleasant that recently some data became available [32] for nuclides with both Z and N even: as said the most intense transitions occur there between ground states. By lack of experimental Nilsson level data, more of such α-decay energies would be very welcome.

10 Neutron-rich nuclides

Progress in the study of very neutron-rich nuclides is rather slow. One must hope that more information will soon become available. Yet, interesting new data have become available in some places, *e.g.* near the crossing of a magic number of protons and neutrons at ^{132}Sn [34]. Unfortunately a preliminary report on recent measurements with the Mainz-Isolde Penning trap seems to indicate somewhat different values. Thus here too, some problems remain to be solved!

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