Methods of atomic-mass measurement in nuclear physics

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The values of the atomic masses are fundamental in the construction and testing of various nuclear models. The masses of nuclei, which determine their binding energy, reflect the delicate balance between the nuclear and Coulomb forces and give information about the structure of nucleon levels in the nucleus. Comparison of the results of theoretical nuclear models with experimentally measured mass values is particularly interesting for short-lived nuclei far from the beta-stability line, for which large discrepancies have been observed between the predictions in various mass models. The values of the atomic masses are needed to calculate the energies of various nuclear decays and reactions. This review focuses on the basic concepts in atomic-mass measurement: the unit of atomic mass and its energy equivalent, the mass number, the mass excess and mass deficit, the mass defect, and the nuclear binding energy. The three types of basic mass formula—from the liquid-drop and the droplet models, the shell model, and phenomenological models—are analyzed. A wide range of problems in nuclear physics requiring the use of mass analyzers is discussed. The requirements on the mass analyzers used to solve these problems are studied. The second part of this review, now being prepared for publication, deals with the detailed discussion of the physical methods of mass analysis and specific mass analyzers.

1. INTRODUCTION

The problem of atomic-mass measurements is so multifaceted that a thorough introduction to it needs to be divided into several parts.

1.1. Goals of the measurements

Atomic-mass measurements represent an area of science whose goal is to create a metrological system of atomic-mass values, which is important both for fundamental knowledge and for broad applications in various scientific disciplines. In nuclear physics, for example, the experimental values of the atomic masses are the initial data determining the energies of all nuclear processes involving nuclides and individual nucleons. Examples of such processes are beta and alpha decay, proton and cluster radioactivity, and spontaneous and induced fission of heavy nuclei. The energy released in a decay process determines whether or not a particular nuclear transformation is possible, and also to a large degree determines the transformation rate, which depends on the penetrability of the potential barrier or on the accessible volume in the momentum phase space of the final transformation products.

The values of the atomic masses are the foundation for constructing various models of the nucleus and the touchstone for testing these models. Of particular importance for the theory of nuclear models are experimental data on the masses of nuclides far from the beta-stability line, for which the results of various theoretical predictions disagree most strongly. The exact, predicted, and measured values of the masses of nuclides lying at the neutron stability limit of nuclei are crucial for constructing cosmological theories of nucleosynthesis in the Big Bang scenario.

Moreover, atomic-mass measurements make it possible to identify the nuclides formed in nuclear reactions and nuclear transformations. The simplest initial step in the identification of a nuclide is the determination of its mass number A (class-C identification). More accurate measurement of the atomic masses with resolution of the nuclide–isobar mass multiplet makes it possible to identify nuclides also according to their atomic number Z (class-A identification). The more subtle difference of the nuclide atomic masses depending on the atomic number is used in this case. This is the second applied aspect of atomic-mass measurements. Such applied mass analysis is a very useful analytical tool also in the identification of complex molecular compounds with molecular weight greater than hundreds of thousands of daltons in chemical studies performed in biology, geology, ecology, medicine, and criminal law.

Atomic-mass measurements involve the use of an extensive arsenal of scientific devices, which can be termed generally as mass analyzers. Here by "mass analyzer" we mean a device used to separate in space or in time a mixture of nuclides with different masses, where the nuclide masses are measured with some accuracy by using either a mass spectrograph or a mass spectrometer. The term "mass spectrograph" was originally used to refer to a mass analyzer which "drew" a static picture of the ion mass spectrum on a photographic plate. If we adhere to this historical definition, in a modern classification of mass analyzers a mass spectrograph would be a device operating in a time-independent mode and simultaneously recording the nuclide masses in some range of values. Naturally, this does not include retuning of the mass spectrograph from one mode to another in order to cover a wider range of mass values. In contrast to the mass spectrograph, the mass spectrometer is a device operating in a time-dependent mode and successively recording and measuring the nuclide masses. It is impossible to use a photographic method for "drawing" the mass spectrum in this mode, and therefore ions in mass spectrometers are detected by electrical methods, which are widely used to record

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ions also in modern mass spectrographs. This is apparently the only fundamental difference between these two types of mass analyzer, although in some cases mass analyzers are classified according to the difference in the device acceptance of the atomic mass, which is a quantitative rather than a qualitative characteristic. Along with such terms in nuclear physics one often finds the term "mass separator." This refers to a mass analyzer which separates and analyzes by weight quantities of matter and large ion currents in the mass-spectrograph mode. Such devices require special technical equipment and are further classified as low-, intermediate-, or high-current.

The modern mass analyzers used in nuclear physics allow measurement of the atomic masses of all the chemical elements and a large number of their compounds with a relative error of down to 10^{-10}. They can be used to separate nuclides of interest from a mixture of other nuclides for relative content of the nuclei of interest as small as 10^{-12} of the sample mass, while, if necessary, recording individual atoms. They can also be used for mass analysis of short-lived radioactive nuclides on time scales of microseconds.

In addition to the direct measurement of atomic masses by means of mass analyzers, for indirect determination of atomic masses in nuclear physics it is very common to measure the energy released in a particular nuclear transformation or nuclear reaction, thus relating a nucleus of unknown mass to a known nuclide. Here the energy or momenta of the transformation products are often measured by using analyzers of the same type as for direct atomic-mass measurements.

1.2. Content of this review

In this review, we discuss the methods of measuring atomic masses and the devices realizing these methods which are used primarily in nuclear physics. We present the basic definitions of atomic mass used in measurements. We discuss the systematization of the experimental values of the atomic masses and the so-called mass formulas, which represent the natural theoretical complement of the information based on experimental data. We consider several problems from nuclear physics which require the use of mass analyzers. We give the requirements on the accuracy of atomic-mass measurements and on the mass analyzers themselves in these problems. We present the fundamentals of the theory of charged-particle motion in magnetic and electric fields and the principles of atomic-mass measurement. We study the different variants of static magnetic mass analyzers. We describe the mass analyzers which use an electric field in addition to a magnetic field. We study resonance mass analyzers, including the very accurate ones based on an electrostatic hyperbolic trap in a uniform magnetic field operating according to the ion cyclotron resonance principle. We also describe the use of ion cyclotrons and storage rings for measuring nuclear masses.

In this review the author has not only had to present information from the studies cited, but also sometimes to resort to critical analysis of certain questions in these studies which in the opinion of the author are controversial, in order to free the reader from this task.

1.3. The main literature sources

There are no modern reviews of the methods and techniques of atomic-mass measurement in nuclear physics which are sufficiently complete and detailed. There are only some reviews which appeared many years ago and so do not reflect the recent advances in the field of mass analysis. An example of such a review is the detailed and thorough article by Bainbridge entitled "Charged particle dynamics and optics, relative isotopic abundances of the elements, atomic masses," published in the first volume of the three-volume encyclopaedia *Experimental Nuclear Physics* (Ref. 1). This review was completed at the end of 1952, and it describes practically all the results in the theory of mass analysis and mass-measurement technique obtained up to that time. Now this review is interesting not only as a source of information on the early stage of development of mass-analysis techniques, but also as a compendium of basic analytic formulas from the ion optics of mass analysis, the foundations of which were laid at the beginning of this century and which have not undergone fundamental changes since then.

As an illustration of the historical aspect of the Bainbridge review, let us just mention the detailed explanation from an outsider's viewpoint of the situation prevailing at the beginning of the century regarding the possible existence of species of atoms of different chemical elements, which are the subject of the present review, and also the role of Thomson and his mass-spectrograph experiments. The Thomson experiments and the chemical studies carried out by Soddy revealed the existence of isotopes, species of atoms of the same chemical element differing from each other in mass (Ref. 1, pp. 560–561).

The proceedings of recent international conferences on atomic masses and fundamental constants are among the useful publications on atomic-mass measurement. The first such conference was held in West Germany in 1956 and is numbered zero in the sequence. Then followed eight conferences in this series, the sixth[2] and seventh[3] of which were, respectively, held in 1979 and 1984. The eighth conference was not held, and the last one[4] was held in Germany in 1992 in conjunction with a conference on nuclei far from the beta-stability line.

A great deal of useful information on the techniques of atomic-mass measurement can also be found in the proceedings of the regular international conferences on "Electromagnetic Isotope Separators and Techniques Related to Their Application," the most recent of which (the 12th) was held in 1991 (Ref. 5). The proceedings of the international conferences on mass spectrometry, which mainly reflect the use of mass analysis in chemistry research, are also useful.

2. BASIC INFORMATION ABOUT ATOMIC MASSES

The technique of measuring atomic masses naturally requires definition of the objects which are measured.

2.1. The atomic mass, the mass number, and the mass excess and deficit

The mass of an atom is concentrated practically entirely in the nucleus, which contains Z protons and N neutrons; a

small fraction of the mass is in the Z electrons orbiting in planetary shells around the nucleus. At the beginning of the century the unit of atomic weight was defined to be one sixteenth of the atomic weight of the chemical element oxygen, and this determined the scale of atomic weights used in chemistry research (the unit of atomic weight, awu). However, in studies of the isotopic content of various chemical elements, the standard became, following the suggestion of Aston,[6] the mass of the single isotope of oxygen known at that time, a mass exactly equal to 16 mass units. It was assumed that oxygen is an element with only one isotope, and therefore the former chemical scale of atomic weights should be kept. However, there soon followed the discovery of two other oxygen isotopes with masses 17.004 and 18.004, which occur in nature only in very small amounts. Owing to these discoveries, the new physical scale of atomic masses did not correspond to the old chemical scale of atomic weights, and the atomic weight unit (awu) in the chemical scale came to be related to the atomic mass unit (amu) on the new physical scale as the ratio 1.000275 to 1.

The masses of all nuclides except, of course, those of the lightest isotope of oxygen, expressed in terms of the new mass unit, had values close to integers, i.e., they were greater or less than an integer by only a small amount much less than 0.1. Because of this, Aston proposed the so-called whole-number rule and the unique assignment to each nuclide of an integer A which he called the mass number.[6] We note that it would have been impossible to do this if the mass of, for example, an atom of a hydrogen isotope were used as the mass unit, as was done at the beginning of the last century by the English scientist Dalton. In that case the masses of heavy atoms would differ strongly (by more than half a mass unit, and in extreme cases by up to two units) from integer values, so that there would not be any one-to-one correspondence between the atomic masses of nuclides and a series of integers.

We note that the first scale of atomic weights in which the unit of atomic weight was taken to be the weight of the lightest chemical element, hydrogen, is still used. For example, in advertisements of companies producing mass spectrometers, the range of measured masses is often given in daltons in honor of the scientist who advanced the hypothesis of the atomic structure of matter.

In 1922 Aston was awarded the Nobel Prize in chemistry "for his discovery, by means of his mass spectrograph, of isotopes in a large number of nonradioactive elements, and for his enunciation of the whole-number rule." The second reason for which he was awarded the Nobel Prize demonstrates the now-forgotten pioneering role that Aston played in establishing the discrete structure of the atomic nucleus.

We note that only after more than ten years, namely, after Chadwick's discovery of the neutron, for which he was also awarded the Nobel Prize, was the proton–neutron structure of the nucleus hypothesized, and it became clear that the mass number A introduced by Aston through the value of the nuclide mass was also the number of nucleons in the nucleus. Unfortunately, in the literature the mass number A is not always presented in the correct historical order. For example, in the single monograph in Russian on atomic-mass systematics by Kravtsov,[7] which is essentially a detailed university textbook, the mass number A is first defined as "the total number of nucleons in the nucleus," and only later does he give "another definition of the mass number as the integer closest to the nuclide mass." We note that in modern nuclear physics there is no way to determine directly the number of nucleons in the nucleus, except by measuring the nuclear mass and expressing it in terms of the mass number A.

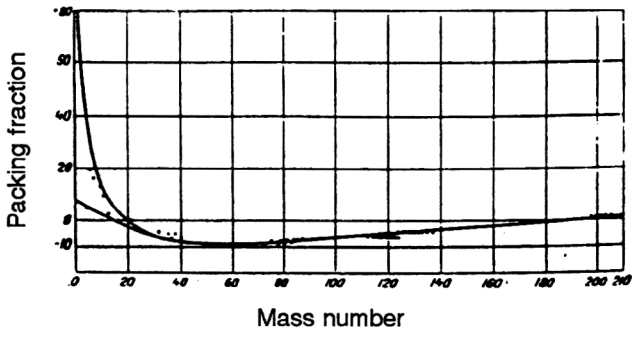

FIG. 1. First dependence of the packing fraction $P(Z,A)$ on the mass number A (Ref. 6).

Thus, in the numerous measurements of the masses of isotopes of various elements made at the beginning of the century by Aston and others it was shown that the nuclide masses $M(A,Z)$ on the new mass scale can be expressed as

$$M(A,Z)=A+\Delta(A,Z), \tag{1}$$

where the mass number A, as is now well known, is equal to the sum of the number of protons Z and the number of neutrons N in the nucleus, $A=Z+N$, and $\Delta(A,Z)$ is a number small (less than 0.1) compared to unity and can be either positive or negative. In those cases where the nuclide mass was larger than the mass number, Aston referred to Δ as the mass excess, and when the nuclide mass was smaller than the mass number he referred to the absolute value of Δ as the defect of the isotopic weight. In the latter case it would have been more logical to refer to Δ as the mass "deficit," the antonym of mass "excess."

2.2. The packing fraction and the mass defect

The first and most important result of defining the concept of mass excess was the analysis of the dependence of this quantity on the mass number. To simplify the analysis, Aston introduced[6] the ratio

$$P(A,Z)=\Delta(A,Z)/A \tag{2}$$

as a measure of the relative density of packing of particles in the nucleus. He referred to this quantity as the packing fraction, and it has for some reason entered the Russian scientific literature as "packing factor" or "packing coefficient."

In Fig. 1 we show Aston's first curve systematizing the packing fractions $P(A,Z)$ as a function of the mass number A for the first measured masses of isotopes of 20 different elements. The packing fraction was expressed in relative tenthousandths. The author believed at that time that the curve

demonstrated to a very high degree an interesting general regularity reflecting a relation between the atomic mass and the intra-atomic energy.

The curve begins with very large positive values for hydrogen. Then it drops, passing through zero in the region of neon. It reaches a minimum near iron and nickel, and then rises slowly, again passing through zero near mercury and taking positive values for the heaviest elements.

The decrease or "vanishing" of the mass δM is accompanied, as follows from the Einstein theory of relativity, by a release of energy

$$E=\delta M c^2, \tag{3}$$

where c is the speed of light in vacuum. The difference between the total mass of the nucleons making up the nucleus and the actual mass of the nucleus,

$$\delta M(A,Z)=NM(1,0)+ZM(1,1)-M(A,Z), \tag{4}$$

where $M(1,1)$ and $M(1,0)$ are respectively the masses of the hydrogen atom and the neutron, determines the total binding energy of the nucleus:

$$E_b(A,Z)=[NM(1,0)+ZM(1,1)-M(A,Z)]c^2. \tag{5}$$

The behavior of the packing curve reflects the main sources of possible release of nuclear energy. In particular, for light nuclides it is possible to have reactions like the fusion of hydrogen nuclei to form helium nuclei and the further fusion of helium nuclei to form nuclei of larger mass with a large energy release per unit mass (3 MeV per nucleon). Such reactions are responsible for the energy radiated by the Sun and other heavenly bodies, including supernovae. At the present time, under terrestrial conditions thermonuclear reactions of the fusion of light nuclei have so far been realized only in uncontrolled thermonuclear explosions, and there are attempts to realize them in controlled thermonuclear processes in special installations.

For heavy nuclides it is the reverse process which is exoenergetic, namely, the transformation of heavy nuclei into lighter ones. When the first edition of Aston's book[6] appeared, the only such process known was that of the successive alpha decay of uranium and thorium isotopes into final products—lead isotopes. Now we also know the process of induced fission of heavy nuclei into two nuclei of intermediate mass, discovered by Hahn and Strassman in 1939, and the process of spontaneous fission, discovered by Flerov and Petrzhak in 1940. These two processes are accompanied by the release of $\sim$200 MeV of energy per fission event or $\sim$0.7 MeV per nucleon of the nucleus undergoing the fission. Induced fission of uranium or plutonium is used, as is well known, in uncontrolled nuclear explosions and in controlled (but sometimes going out of control) nuclear reactors as a powerful source of thermal energy.

A number of authors later constructed packing-fraction curves by using a large number of nuclide mass values. These curves, which improved on, for example, the effects of nucleon filling of nuclear shells, repeated the basic regularity of nuclear packing with increasing mass number discovered by Aston.

The decrease of the mass in the formation of bound nuclei or as a result of nuclear transformations was referred to as the "mass defect" by Aston. This aggravated the ambiguity of using the terms "defect of the isotopic weight" and "mass defect," and also the term "mass excess." For example, in the monograph on atomic physics[7] authorized as a textbook at the college level we read: "The mass deficit of hydrogen is +0.008123 (on the mass scale $M(^{16}\mathrm{O})=16$), the neutron mass deficit is +0.00893," and so on, instead of Aston's definitions of the mass "excess" of hydrogen and the mass "excess" of the neutron. Similar variants of the definitions can be found in the modern chart of nuclides,[8] where the mass excess is referred to as the mass defect. In the current Russian scientific literature the mass number is sometimes referred to as the mass, the atomic weight, or the "number of the mass," a literal translation of the English term into Russian.

The correct or, at least, uniform terminology in questions concerning nuclide masses is also important because for very light nuclei, which at present are the subject of intensive theoretical and experimental study, the values of the mass excess and the mass defect for some nuclides are quite close. Therefore, because of the confusion in the definitions of these concepts it is sometimes impossible to understand what is being referred to, even if only approximate, incompletely established values of these quantities are known. For example, for an exotic and insufficiently studied nuclide like the ^{7}B two-proton emitter, the mass excess and mass defect are $\Delta M(^7\mathrm{B})=23-29$ mu and $\delta M(^7\mathrm{B})=23-29$ mu according to the data of various theoretical models. (Here the mass excess and mass defect of the nuclide are expressed in the new unit of mass u which will be discussed below.)

2.3. The modern unit of atomic mass

A drawback of the physical mass scale $M(^{16}\mathrm{O})=16$ was the difficulty of comparing the masses of various nuclides with the reference nuclide ^{16}O owing to the fact that oxygen does not form molecular compounds with many elements. Carbon is more convenient in this regard, as it forms a great many different compounds, making it possible to compare the isotopes of nearly all the elements with each other by mass-analytic methods. Accordingly, in 1959 the Commission on Atomic Weights of the International Union of Pure and Applied Chemistry, IUPAC, recommended that carbon be used as the standard mass and that the unit of mass be one twelfth of the mass of the ^{12}C atom in the ground atomic and nuclear states. This proposal was supported and adopted in the General Assembly of the International Union of Pure and Applied Physics, IUPAP. On 13 December 1961, as noted in Ref. 7, the Bureau of the Division of Physical and Mathematical Sciences of the USSR Academy of Sciences, according to the report of Academic Secretary L. A. Artsimovich, resolved to "accept the new unit for nuclide masses (atomic weights) $^{12}\mathrm{C}=12$, recommended by the Tenth General Assembly of the IUPAP and adopted by the IUPAC Council in the summer of 1961." This measure of atomic mass became known as the unified mass unit and was abbreviated as u in the international literature. The ratio of the new mass unit to the atomic mass unit of the scale $M(^{16}\mathrm{O})=16$ is

$$1\ \text{u} = (1.000317917 \pm 0.000000017)\ \text{amu}.$$

We note that the establishment of a new atomic mass unit in terms of the nuclide ^{12}C, which in turn is defined in terms of the atomic mass unit in the system of units $^{16}O = 16$ (the mass of ^{12}C is equal to 11.996 amu, and the mass number is 12), is not of sufficient metrological rigor. It would seem to be more logical to define the new unit of mass as one twelfth of the mass of the lightest of the two known stable isotopes of carbon, which does not require any preliminary "weighting" of the reference mass in some mass scale.

The numerical value of the unified mass unit is defined as

$$1\ \text{u} = m(^{12}C)/12N_A = (1660540.2 \pm 1.0) \cdot 10^{-33}\ \text{kg}, \quad (6)$$

where $m(^{12}C)$ is the molar weight of the carbon-12 isotope, equal to 12×10^{-3} kg/mole and N_A is Avogadro's number, equal to $(6.0221363 \pm 0.0000068) \times 10^{23}$/mole. The accuracy of the numerical value of the mass unit is determined by the accuracy of Avogadro's number, which was last measured in 1992 by determining the ratio of the molar volume to the atomic volume for superpure crystalline silicon.

2.4. The energy equivalent of the mass unit

The internal energy of the nucleus, the energy of nuclear transformations and nuclear reactions, the energy of electromagnetic radiation, and the kinetic energy of neutral products of transformations and reactions are usually expressed in a special energy unit used in nuclear physics: the electron volt (eV), even though the electron volt by definition is the kinetic energy of a charged particle with a single elementary charge q_0 accelerated by an electric potential difference of one volt. Therefore, in order to use the large amount of numerical data on the energies of nuclear transformations in calculations of the atomic masses of unknown nuclides and, conversely, to use the data on nuclear masses in calculations of nuclear transformation and reaction energies, we need the factor for converting the mass unit into its energy equivalent, expressed in eV/c^2.

The expression of the mass unit in eV/c^2 determined by relativity theory (3) is given by

$$1\ \text{u} = [(10^{-3}\ \text{kg/mole}) \cdot c^2/(N_A q_0 V)]\ \text{eV}/c^2,$$

in which the numerical values of the speed of light c, the elementary charge q_0, and the unit of electric potential V are expressed in fundamental units (meter, kilogram, second, ampere) of the international system of units (SI) adopted at the Eleventh General Conference on Weights and Measures in 1960. Even though the electric potential producing in an electric circuit a constant current of strength 1 A with a power of 1 W is taken as the unit of potential in the SI system, throughout many years of practical experience the unit of potential (volt) has been defined by electrolytic voltage elements maintained in national metrological laboratories and periodically compared with the principal standard kept in the Bureau International de Poids et Mesures, BIPM. After the realization of the effect, predicted by Josephson,[10] of the tunneling of electron pairs through a dielectric separating two superconductors, the BIPM commission proposed that this effect be used for a more accurate maintenance of the electric voltage standard. The Josephson effect will be explained below in connection with our discussion of the analogous effect in nuclei. Here we only note for now that when a potential difference V is applied to superconductors, a nonstationary (variable) electric current with frequency

$$f = 2q_0 \cdot V/\hbar \quad (7)$$

passes through the dielectric; here $\hbar$ is the Planck constant. Equation (7), which determines the frequency of the variable current and the electromagnetic radiation accompanying it, is analogous to the expression for the frequency of electromagnetic radiation by an atom induced by a transition of an electron from one level to another separated from the first by an energy $q_0 V$. The difference between Eq. (7) and the expression for radiation by an atom amounts only to replacement of the coefficient 2 by 1, which arises becauses in the Josephson effect a pair of electrons rather than a single electron is involved in the transition from one superconductor to the other.

As reported in Ref. 11, in 1969–1972 it was suggested that the value 483594.0 GHz/V be used as the coefficient of proportionality between the frequency of electromagnetic radiation and the potential difference in Eq. (7). This corresponds to the value of the international voltage standard supported at that time. This defined the energy equivalent of the mass unit to be

$$1\ \text{u} = (931501{,}2 \pm 0{,}3)\ \text{keV}/c^2.$$

However, after the proton gyromagnetic ratio was improved in 1986 the new value $2q_0/\hbar = (483597.67 \pm 0.14)$ GHz/V was recommended. It is larger than the former value by $7.6 \cdot 10^{-6}$. Accordingly, a new equivalent of the mass unit was defined:

$$1\ \text{u} = (931494.32 \pm 0.28)\ \text{keV}/c^2.$$

In 1990 the constant in the expression for the Josephson effect (7) was again changed, owing to improvement of the resistance standard, and became

$$2q_0/\hbar = 483597.9\ \text{GHz/W*}$$

without allowance for possible error.

Accordingly, at the present time

$$1\ \text{u} = (931493.86 \pm 0.07)\ \text{keV*}/c^2, \quad (8)$$

where W* is the voltage standard, established in 1990 according to Eq. (7). The small difference in the coefficients of the conversion of the unit of atomic mass to its energy equivalent in different historical periods should be borne in mind when converting mass values and their energy equivalents from different years if the relative error in the values in question must be less than 10^{-5}.

3. PREDICTIONS AND SYSTEMATIZATION OF THE MASS VALUES

As noted above, the set of experimental values of the atomic masses is one of the fundamental bases for the con-

struction of various nuclear models. A special role in such constructions is played by equations predicting the atomic masses or, as they are still called, mass formulas, which determine hypothetical values of the atomic masses as a function of the atomic number Z and the mass number A. The mass formulas are phenomenological; they are empirical analytical expressions based on certain theoretical ideas. These analytic expressions contain free parameters which are selected according to the experimental (empirical) values of the atomic masses, and also according to the data on the nuclear radii and the nuclear magnetic dipole and electric quadrupole moments. The total nuclear mass in a model is made up of a macroscopic part determining the fundamental component of the nuclear mass on the basis of a general approximate description and a microscopic part representing a not-necessarily-small addition due to special features of the nuclei in question. The mass formulas are required not only to represent the values of the masses of all known nuclei as accurately as possible, but also to give a sufficiently reliable prediction of the masses of nuclides not yet discovered. The latter requirement is checked not only by practical comparison of the predicted mass values with the masses of new nuclei, but also by mathematical methods based on the maximum-likelihood principle.

All mass formulas can with some degree of arbitrariness be classified into three groups. Formulas of the first type are based on the classical representation of the nucleus as a liquid drop. Those of the second express the mass–energy of the nucleus on the basis of the quantum-mechanical model of nucleon shells in the nucleus. Finally, those of the third class use certain phenomenological (observable) relations between the mass values of known nuclei.

3.1. Mass formulas based on the liquid-drop model

The first classical mass model of this type was proposed about 60 years ago in the form of an expression referred to as the Weizsäcker or Bethe–Weizsäcker formula.[12] This formula determines the mass $M(N,Z)$ or the energy $E(N,Z)$ of a nucleus by representing the nucleus as a spherical drop of a uniformly charged, incompressible liquid. The Weizsäcker formula is written analytically as

$$M(N,Z)=a_v A+a_s A^{2/3}+3q_0^2Z^2/5r_0A^{1/3}+a_{\rm sym}I^2/A, \tag{9}$$

where $I=N-Z$ is a number referred to as the neutron excess, q_0 is the proton electric charge, and a_v, a_s, r_0, and $a_{\rm sym}$ are fitted parameters determined from the experimental values of the atomic masses. The first term in the formula is the volume energy of the classical nuclear drop, the second is the surface energy, and the third is the Coulomb energy of a charged liquid. We have also introduced into the formula an additional quantum-mechanical symmetry energy, more precisely, an asymmetry energy, associated with the nucleon content of the nucleus.

The numerical values of the parameters in the mass formulas are determined from the experimental values of the atomic masses by fitting. For this a system of linear conditional equations is written down which equate the experimental mass values to the so-called theoretical ones. In this system the parameters of the formula are treated as unknown quantities. Since the number of unknown parameters of the mass formula m is much smaller than the number of equations n, equal to the number of experimental atomic masses used, the system is overdetermined and likely to be inconsistent. For example, Eq. (9) contains only four fitted parameters, in modern liquid-drop mass formulas $m\simeq 50$, while the number of experimental values of the atomic masses used to adjust the coefficients is $n\simeq 2000$. The system of inconsistent equations does not have an exact solution and can be satisfied only approximately to some degree. This solution is sought by the method of least squares, i.e., by minimizing the rms deviation of the nuclide masses found from the mass formula from the reference experimental values of the masses:

$$\sigma_{\rm rms}=\left[(1/n)\sum_{i=1}^{n}(M^i_{\rm exp}-M^i_{\rm th})^2\right]^{1/2}. \tag{10}$$

The rms deviation of the calculated masses from the experimental values for the liquid-drop mass formula is about $\sigma_{\rm rms}=2.5$ MeV.

Equation (9) does not include the deformation of the nuclear drop or the compressibility of nuclear matter; moreover, it is applicable only for nuclei with small neutron excess $|I|<A$.

In Ref. 13 Myers and Swiatecki represented the nuclear mass as an expansion in a small dimensionless parameter, the ratio of the nuclear force range and the linear dimension of the nucleus, which is roughly proportional to $A^{-1/3}$ and $[(N-Z)/A]$.[2] Keeping the lowest terms in the expansion, the authors obtained the following series for the nuclear mass:

$$\begin{aligned}M(N,Z)=&a_1A+a_2A^{2/3}+a_3A^{1/3}+a_4I^2/A\\&+a_5I^4/A^{4/3}+a_6I^4/A^3+c_1Z^2\\&\times(1+\alpha/A^{1/3})/A^{1/3}+c_2Z^2A^{1/3}\\&+c_3Z^2/A+c_4Z^{4/3}/A^{1/3}.\end{aligned} \tag{11}$$

The first and second terms in Eq. (11) are the volume and surface energies of the nucleus already contained in (9). The third term is the additional correction to the energy from the compressibility of nuclear matter and from the change of curvature of the nuclear surface. The fourth and fifth terms reflect the corrections to the volume and surface energies arising from the asymmetry of the nucleon content of the nucleus. The following terms include the change of the Coulomb energy due to the difference between the neutron and proton distributions in the nucleus, the diffuseness of the nuclear surface, and several other factors.

Because the appearance of the terms in (11) not present in (9) led to relatively large values of the expansion parameter, the above-quoted ratio, for nuclei with small values of A, i.e., for nuclei of small size, the new model became known as the droplet model.

The deviations of the calculated values of the atomic masses from the experimental values in the droplet model turned out to be roughly the same as in the liquid-drop

model, and they repeated the sequence of filling of nuclear shells by nucleons. For these reasons it was found that additional shell corrections of the microscopic type had to be included in the nuclear energy.

The simplest correction of the microscopic type to the macroscopic nuclear mass which qualitatively reproduced the shell deviations was again proposed by Myers and Swiatecki.[14] They started from the model of motion of independent nucleons in the finite nuclear volume and assumed that the microscopic correction arose from the splitting of the levels of these nucleons. As the starting point for the microscopic part they chose a system of uniformly spaced, degenerate levels of a Fermi gas in a spherical volume. They then introduced the level-splitting function depending on the nucleon filling of the next shell, after relating the shell filling to the deformation of the nuclear shape. The nuclear deformation led to a change of the energy of the split levels and made them mix strongly. Then they found the minimum of the total energy of the nucleus, equal to the sum of the liquid-drop mass and the microscopic correction, as a function of the deformation and calculated the energy of the nuclear ground state, i.e., the lowest state, obtained in this way. The model coefficients were found by a fit.

The numerical values obtained for the microscopic correction qualitatively reproduced the deviations of the masses calculated by using the liquid-drop model from the experimental masses, but individual deviations reached several MeV. The deviations in the region of light nuclei with $N,Z \leqslant 29$ and particularly for $N=Z$ were especially large. The discrepancy in the latter case was eliminated by introducing the so-called Wigner correction $-7\exp(-|6I|/A)$ MeV, introduced earlier by Wigner in studying nuclear forces.

The method of including the shell correction in the mass formula was developed further by Strutinsky.[15] As the initial microscopic state he chose single-particle proton and neutron energy levels in the Nilsson potential, which was more realistic than the degenerate Fermi-gas levels. Then he considered the effect of deformation of the nuclear shape, described by even Legendre polynomials of second and fourth order, on the location of the single-particle levels near the limiting Fermi energy. To determine the nonregular component of the additional energy arising in deformation he used special averaging of the energies of the single-particle levels on a finite energy interval approximately equal to the energy spacing between shells (7–10 MeV). The equilibrium shape of the nucleus and its energy were determined according to the minimum of the additional energy. In addition to the shell correction, the nucleon pairing energy was also included in this model.

The calculated nuclear masses agreed with the experimental values considerably better than in the earlier models: the average deviation was 0.733 MeV. The additional nuclear deformations that were found agreed with the experimental values within several percent.

Without dwelling further on subsequent models of this type, let us just quote the most recent liquid-drop mass formulas. One of the formulas[16] is distinguished by the fact that it contains a small (according to current ideas) number of free parameters ($m=12$) and is therefore, in the opinion of the authors, better justified theoretically and more likely to be correct in predicting the masses of unknown nuclides than formulas with many parameters. Another feature of this formula is the simple linear relation between the energy of the shell correction and the deformation. The formula of Ref. 16 gives the masses of 652 known nuclides with rms deviation $\sigma_{\text{rms}}=0.55$ mu and has been used to predict the masses of 4162 nuclei, but with N and Z greater than 50.

Another version of the macroscopic–microscopic mass formula based on the droplet model is described in Ref. 17. The essence of this mass formula can to some degree be understood from its full name: "nuclear mass formula in the droplet model with finite nuclear-force range and based on the one-particle bounded Yukawa potential" and from its macroscopic–microscopic terms expressing the volume and surface energies, the energy of the surface curvature, the Coulomb energy, the volume and surface energies due to inadequacy of the neutron and proton distributions in the nucleus, the Coulomb exchange energy, the correction for the proton form factor, the charge asymmetry energy, the Wigner energy, the nucleon pairing energy, and, finally, the binding energy of the electrons in the atomic shells. To calculate the 29 parameters of the model the authors used the values of the atomic masses of 1593 nuclides from ^{16}O to ^{263}Rf. The masses of 4678 nuclides from ^{16}O to ^{318}122 were calculated.

The model of Ref. 17 is referred to by the shortened name FRDM, for finite-range droplet model.

The accuracy of the parameters of the mass formula and the errors intrinsic to the calculated formula were estimated in Ref. 17. Let us discuss these. As noted above, the errors of the mass model are usually estimated as rms deviations given by Eq. (10). The use of Eq. (10) to estimate the accuracy of the mass formula is justified if all the experimental errors σ^i_{exp} are small compared with σ_{rms}. However, this requirement is often not met for masses of nuclides far from the beta-stability line. The masses of such nuclei, which, as a rule, are short-lived and formed in nuclear reactions with small cross sections, are measured with considerably larger errors than the masses of stable and long-lived nuclides. In these cases the rms deviation (10) always overestimates the error introduced by the mass formula itself.

To estimate the quality of the mass model the authors of Ref. 17 used the results of Ref. 18 and separated the model error from the total error. On the basis of the statistical theory of distributions of random deviations they assumed that the theoretical error depending on the many parameters of the mass model is distributed according to a normal law of random deviations and does not have any systematic deviation. Then the deviations of the error μ^i_{th} from zero are determined by the deviation of the normal distribution σ_{th}. In turn, the value of σ_{th} is determined, as shown in Ref. 18, by minimizing the function

$$S=\sum_{i=1}^{n}\left[M^i_{\text{exp}}-(M^i_{\text{th}}+\mu^*_{\text{th}})\right]^2/(\sigma^{i2}_{\text{exp}}+\sigma^{2*}_{\text{th}}),$$

with the condition of solving the equations

$$\sum_{i=1}^{n} \{[(M^i_{\rm exp}-(M^i_t+\mu^*_{\rm th})]^2-(\sigma^{i2}_{\rm exp}+\sigma^{2*}_{\rm th})]\}$$

$$/(\sigma^{i2}_{\rm exp}+\sigma^{2*}_{\rm th})^2=0,$$

$$\sum_{i=1}^{n} [(M^i_{\rm exp}-(M^i_{\rm th}+\mu^*_{\rm th})]/(\sigma^{i2}_{\rm exp}+\sigma^{2*}_{\rm th})=0.$$

In these equations $\sigma^{2*}_{\rm th}$ is the approximate value of the true error of the mass formula $\sigma^2_{\rm th}$ found by iteration. A perfect mass model would have $\sigma_{\rm th}=0$.

In Ref. 17 the error of the theoretical model without error due to inaccuracies of the reference values of the masses was $\sigma_{\rm th}=0.769$ MeV. On the other hand, the theoretical error found in the same manner using the masses of 1443 nuclei known with an error of less than 0.1 MeV was 0.782 MeV. The fact that the error $\sigma_{\rm th}$ did not decrease when the model was based on more accurate reference masses is viewed by the authors as confirmation of the correctness of the method of estimating the theoretical error of the mass formula.

3.2. Mass formulas based on the nuclear shell model

The simplest mass formula of this class is based on the representation of the nucleus as a system of spherical nucleon pairs with total angular momentum equal to zero filling the nuclear shells in a spherically symmetric potential.[19] The nuclear energy in this model is written as the energy of the N_0 neutrons and Z_0 protons in a filled shell plus the energy of the n neutrons and p protons above a filled shell. The pairing energies of two neutrons, two protons, and also a neutron and a proton are introduced into the model, along with the nuclear effective interaction of the nucleon pairs. As a result of this representation, the nuclear masses are represented as four parabolic surfaces separated by intervals according to the evenness or oddness of the numbers of neutrons and protons in the nuclei. The shell boundaries are clearly manifested as sharp changes in the neutron and proton binding energies.

For nuclei with mass numbers in the ranges between magic numbers, inside which there are many intermediate subshells, an additional function is introduced for the nuclear deformation energy due to the configurational interaction. This function, which is symmetric with respect to particles above the shell and holes in the shell adds to the smooth parabolic energy surfaces oscillatory corrections reflecting the real behavior of the nucleon separation energies. This effect is especially clearly manifested in the change of the binding energy of neutron pairs in the rare-earth region for $N=90$. Although this model does not have a clear physical interpretation, the mass formula based on nuclear shells explains the experimental values of the atomic masses with an error of 0.1–0.2 MeV.

Another version of a mass formula based on the shell model is presented in Ref. 18. The macroscopic part of the model is based on the exponentially bounded Yukawa potential. The starting point for the microscopic part is the bounded single-particle Yukawa potential. The new model has been used to take into account neutron pairing, proton pairing, and also neutron and proton pairing. The pairing energy in even–even nuclei is taken as the zero reference point from which the pairing energy is measured, in contrast to the average pairing energy in even–even and odd–odd nuclei usually used as the zero. This model came to be referred to as the FRLDM, like the abbreviated name of the model of Ref. 17, but with the word "droplet" in the full name replaced by "liquid-drop."

The FRLDM has been used to calculate the masses of 4678 nuclides in the range from ^{16}O to ^{263}122.

There is also an estimate of the theoretical error in the mass formula for this model, as for the model of Ref. 17. The accuracy of the mass formula constructed from the experimental values of the same masses as in Ref. 17, 1593 nuclei from ^{16}O to ^{263}Rf, is somewhat worse; the error in the theoretical formula is estimated to have the rms value $\sigma_{\rm th}=0.832$ MeV.

The mass formula of Ref. 20 is based on an empirical concept of shells with a general equation of the form

$$M(N,Z)=M_g(N,Z)+M_{\rm eo}(N,Z)+M_s(N,Z),$$

where M_g is the basic (approximate and smooth) function of N and Z, $M_{\rm eo}$ is a function taking into account the averaged even–odd component of the mass, and M_s is a function taking into account the shell corrections. For the 1657 masses of the reference nuclides, $\sigma_{\rm rms}=0.538$ MeV. The formula has been used to predict the masses of 7204 nuclides.

What appears to be the most recent mass model based on the idea of nuclear shells is given in Ref. 22. This model, which gives "nuclear masses in the model of fermion dynamical symmetry," is based on the theory of the effective nucleon–fermion interaction (in contrast to the interaction of paired nucleon–bosons) with filled spherical shells and restricted dynamical symmetry. The single-particle energies based on the Woods–Saxon potential and the Strutinsky modified shell correction are used as the microscopic part of the model. The general structure of the mass formula is

$$M(N,Z)=M(^{208}{\rm Pb})+n_nM(1.0)+n_pM(0.1)+\langle M\rangle,$$

where $n_n=N-126$ and $n_p=Z-82$ are the numbers of valence neutrons and protons in the nucleus, and $\langle M\rangle$ is the expectation value (after iteration) of the Hamiltonian. The model gives calculated masses which agree with 332 experimental masses in the region of isotopes of actinide elements with $\sigma_{\rm rms}=0.34$ MeV. The model predicts the possible existence of superheavy nuclides with $Z=114$ and $N=164$. The energy of the shell correction for the nuclide ^{278}114 amounts to -12.6 MeV, which is comparable to the large shell correction of -13.75 MeV for the doubly magic ^{208}Pb nucleus.

The authors of Ref. 22 note that their model gives values of the masses of heavy nuclides, the shell corrections, and the separation energy of the last neutron and proton in better agreement with experiment than does the model of Ref. 18.

We note that there are also other predictions of the location of the "stability island" in the "sea" of superheavy nuclides. In the mid-1960s the values $N=184$ and $Z=114$ were taken as the magic numbers of neutrons and protons in the region of heavy elements. In 1986 in Ref. 23 it was first

shown that the most long-lived nuclides are apparently the deformed nuclei $^{288}110$ and $^{290}110$. These predictions are based on increasing the contribution to the stability of heavy nuclei from the shell correction near $N=162$ and $Z=108$ due to the large gap in the scheme of corresponding Nilsson levels.

The experimental results of Ref. 24 were recently added to these predictions. In the bombardment of ^{248}Cm by ^{22}Ne ions two new nuclides ^{265}Rf and ^{266}Rf were discovered, which have higher stability as $N=162$ is approached.

3.3. Mass formulas based on phenomenological relations

The mass formulas of this class are based on the known phenomenological relations between values of atomic masses known from experiment. The first formula of this type was that of Garvey and Kelson.[25] It is based on the following relation between, for example, any six values of atomic masses found by analysis of the experimental data:

$$M(N+2,Z-2)-M(N,Z)+M(N,Z-1)-M(N+1,Z-2)+M(N+1,Z)-M(N+2,Z-1)=0. \quad (12)$$

Equation (12) was checked by substituting 621 atomic masses for $A\geqslant 16$ and $N\geqslant Z$ with the condition that if $N=Z$, then N is even. The value of the average deviation of the left-hand side of (12) from zero turned out to be only 0.198 MeV. The deviations were more or less random, and no regularity or trend was seen in them. It was only noticed that the deviations decrease with increasing A.

It is easily verified that Eq. (12) is satisfied exactly if it is assumed that the nuclear mass depends additively only on the number of neutrons, the number of protons, and the mass number, which is more or less justified in the independent-particle model, i.e.,

$$M(N,Z)=g_1(N)+g_2(Z)+g_3(A), \quad (13)$$

where the g are arbitrary functions of their arguments. After substituting (13) into (12) and canceling terms of the same absolute value entering pairwise with plus and minus signs we identically obtain zero.

In the literature several methods are given which clearly illustrate the phenomenological coupling of the mass values (12), using nuclear level-filling schemes and other simple ideas. Here we present a similar illustration which has been proposed by the present author.

We write Eq. (12) as

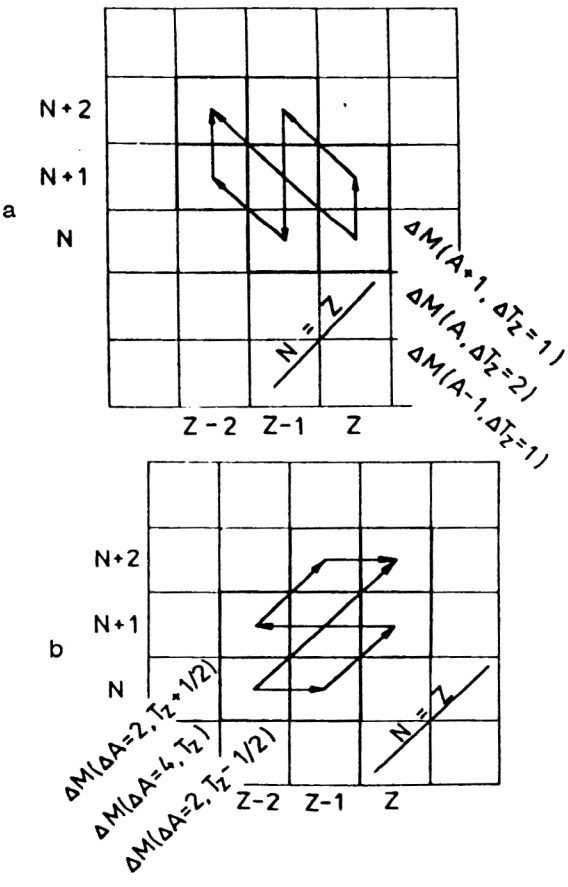

FIG. 2. Illustration of the scheme for extrapolating values of the atomic masses in the phenomenological Garvey–Kelson model. (a) Transverse extrapolation. The heavy solid lines show $\Delta M(A,\Delta T_z)$. (b) Longitudinal extrapolation. The heavy solid lines show $\Delta M(\Delta A,T_z)$.

$$\Delta M(A,\Delta T_z=2)=\Delta M(A+1,\Delta T_z=1)+\Delta M(A-1,\Delta T_z=1), \quad (14)$$

where ΔM is the mass difference of two nuclide-isobars with mass numbers A and difference of the isotopic-spin projections equal to ΔT_z. For example, $\Delta M(A,\Delta T_z=2)=M(N+2,Z-2)-M(N,Z)$, $T_z=(N-Z)/2$ is the projection of the isotopic spin on the selected direction in isotopic-spin space. Equation (14) is represented in Fig. 2a by a scheme which shows that the mass difference of two nuclide-isobars with mass numbers A and isospin-projection difference $\Delta T_z=2$ is the sum of two other analogous nuclide mass differences, but with half the value $\Delta T_z=1$ and mass numbers respectively one unit larger and one unit smaller: $A+1$ and $A-1$. This scheme reveals the path, shown by the heavy solid lines, of the approximate extrapolation from one known atomic mass $M(N,Z)$ to another, unknown mass $M(N+2,Z-2)$ through two other differences of known nuclear masses. The change of nuclear mass in going from $M(N,Z)$ to $M(N+1,Z)$ and from $M(N+1,Z-2)$ to $M(N+2,Z-2)$, shown by the thin lines, is approximately canceled by the change of mass in going from $M(N+2,Z-1)$ to $M(N,Z-1)$. The validity of this approximate extrapolation is certain if along the extrapolation path

there are no sharp jumps in the mass values as the mass number A changes. This condition essentially ensures that the extrapolation lines do not intersect the line $N=Z$.

In addition to the above so-called transverse mass extrapolation leading to new values of the nuclide masses located to the side of the beta-stability line, a longitudinal extrapolation was also proposed in Ref. 25 which leads, in particular, to masses of isotopes of new, distant transuranium elements. For this case

$$M(N+2,Z)-M(N,Z-2)+M(N+1,Z-2) \\ -M(N+2,Z-1)+M(N,Z-1)-M(N+1,Z)=0 \quad (15)$$

with the condition that $M(N,Z)=f_1(N)+f_2(Z)+f_3(N-Z)$, where again the f are arbitrary functions of their arguments. Equation (15) has also, like (12), been checked using the experimental masses of 755 nuclei, and the average deviation from zero was found to be 0.189 MeV.

Equation (15) leads to a formula analogous to (14) for a clear longitudinal extrapolation of the nuclide masses to the region of nuclides of unknown mass:

$$\Delta M(\Delta A=4,T_z)=\Delta M(\Delta A=2,T_z+1/2) \\ +\Delta M(\Delta A=2,T_z-1/2), \quad (16)$$

where, for example, $\Delta M(\Delta A=4,T_z)=M(N+2,Z)-M(N,Z-2)$. The longitudinal scheme of determining an unknown nuclide mass using Eq. (16) is illustrated in Fig. 2b. In this case the extrapolation to the unknown mass runs along lines with constant projection of the isotopic spin T_z instead of lines with constant value of the mass number as in the method (14).

The Garvey–Kelson mass formula was used in Ref. 26 to predict 6537 nuclide masses. There, to raise the accuracy of the predictions, the reference values were taken to be experimental masses known with an error of less than 0.250 MeV. The standard deviation of the calculated values from the reference experimental values is $\sigma_{\text{rms}}=0.219$ MeV. As a result, nuclide masses in the ranges $2\leqslant Z\leqslant 109$, $4\leqslant N\leqslant 157$, and $7\leqslant A\leqslant 265$ were predicted. These predictions also included neutron-depleted nuclei, but without nuclides with odd N equal to Z for $Z<27$.

In another study[27] the Garvey–Kelson method was used to calculate about 5600 mass excesses of nuclides with $2\leqslant Z\leqslant 103$ and $4\leqslant N\leqslant 157$ for $N\geqslant Z$, but without nuclides with odd $N=Z$ for $A<40$. The standard deviation in reproducing the experimental mass excesses is $\sigma_{\text{rms}}=0.103$ MeV. The mass excesses were also calculated for neutron-depleted nuclides using the charge-symmetry relation. In this case values of another 250 mass excesses were obtained with the standard deviation of the calculated values from the reference ones equal to $\sigma_{\text{rms}}=0.231$ MeV.

There are also many model approaches to the calculation of the masses of a limited range of nuclei or calculations of individual, mainly light, nuclides on which it is not possible to dwell here. As an exception, we note only that apparently the most recent such publication is Ref. 28, where the principle of mirror symmetry $M(Z,N)_{\text{micro}}=M(N,Z)_{\text{micro}}$ is used, together with inclusion in the macroscopic part of the nuclear mass of the change in the Coulomb energy due to replacement of neutrons by protons in the mirror nucleus. This model was used to improve 451 values of the masses of nuclides with $Z>N$ in the region $9\leqslant Z\leqslant 64$, $17\leqslant A\leqslant 120$. A detailed comparison of the results with those of 11 other mass models in the preprint version and with the results of 17 models in the journal version was also made in that study. We note that these nuclear masses are expressed in terms of what was called the "mass excess" in the English-language preprint and the "nuclear mass defect" in the Russian-language journal article.

3.4. Compilation of mass formulas

Above, we discussed the basic mass formulas which generalize and predict large sets of mass values. Naturally, these are not the only mass models known at present. The results of calculations using ten of the most sophisticated mass models of different authors, completed in 1986–1987, six of which (Refs. 16–18 21, 26, and 27) have been discussed here, are presented together in one issue of the journal *Atomic Data and Nuclear Data Tables* (Ref. 29). In addition, the compilation includes atomic mass values[29] found experimentally in most cases and, in only a small number of cases, calculated on the basis of systematic trends in the experimental values. All the results, both calculated and experimental, are presented in a single table which is convenient to use.

By now there have been scores of calculations of large sets of nuclear masses based on various macroscopic–microscopic models with different versions of their macroscopic and microscopic parts. Calculations of the microscopic part of the nuclear mass typically use various representations of the single-particle potential like the bounded Yukawa potential, the Woods–Saxon potential, and the oscillator potential. Different local parameters are used in each microscopic approach. This great diversity of models, which have been tested and found to be consistent for known nuclei, lead to different results for masses of unknown nuclides far from the beta-stability line. Estimating the plausibility of the calculated results for as yet unknown nuclides is an important problem.

3.5. Comparison of the mass formulas

The summary[30] written by the editor of the mass-formula compilation compares the models according to the number of fitted parameters, the number of reference experimental masses, the average and rms deviations of the calculated values from the reference experimental ones, and the number of predicted mass values. Using these results and assuming that the mass formula most weakly justified in the sense of the arguments presented in Ref. 16 is that giving the minimum value of the product $m\sigma_{\text{rms}}$, which takes into account not only the number of parameters m, but also the quality of the formula σ_{rms}, we find that this formula is the one in Ref. 16.

A critical study of various mass models has recently been performed in Ref. 31. For estimating the accuracy of

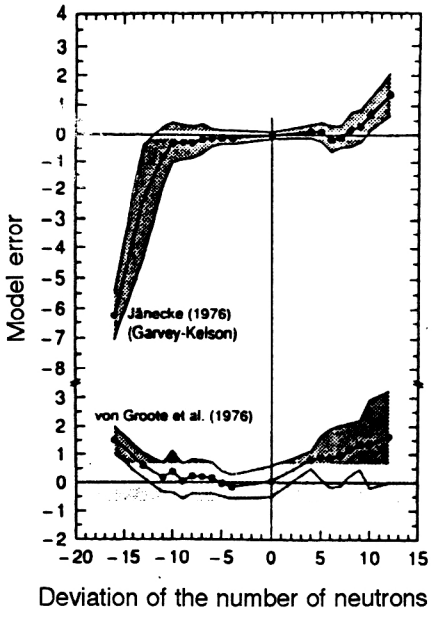

FIG. 3. Theoretical errors in various mass models.[31] The distance of a nuclide from the beta-stability line is represented by the number of neutrons along the horizontal axis. The average error μ_{th} is shown by the points; μ_{th} plus the standard deviation σ_{th} is shown by the dark shaded region; σ_{th} found from the reference values of the atomic masses is shown by the light shaded region.

predicting the atomic masses by a model itself the authors used a method developed by them in an earlier study,[18] which we have described above.

In Figs. 3 and 4 we show the theoretical errors of the various models, which in Ref. 31 are indicated by the names of their authors or the name of the model without literature references. Along the vertical axis we give the error of the model in estimating relatively inaccurately measured masses of nuclei located a certain number of neutrons off to either side of the beta-stability line. The relatively narrow, straight band in these figures lying right on the horizontal axis represents the rms deviation determined from the entire set of nuclide masses used to fit the parameters of the model. The black points show the average errors for several masses of relatively new nuclides not included in the data used to fit the model parameters. The crooked band shows the standard deviations from the average error.

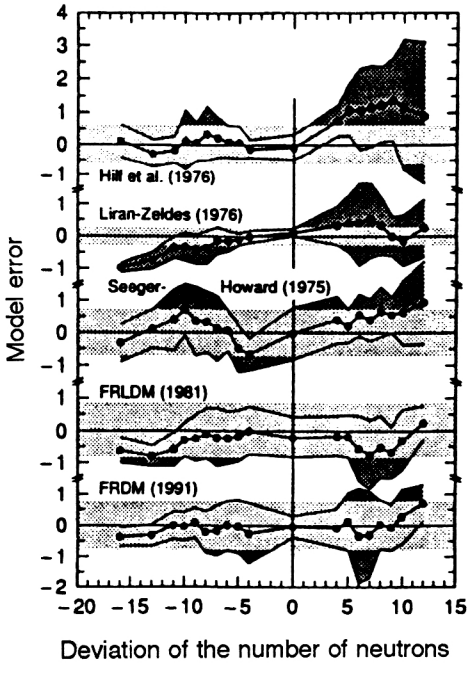

FIG. 4. Theoretical errors in various mass models.[31] The distance of a nuclide from the beta-stability line is represented by the number of neutrons along the horizontal axis. The notation is the same as in Fig. 3.

It is noted in Ref. 31 that in models based on a quantum-mechanical treatment of the nuclear interaction, in particular, in the three models shown in the lower part of Fig. 4, only one point of all the points representing the average deviations of the mass values in question lies outside the errors found originally. In these models also the total errors for these nuclei lie mainly within the bounds of the original total error. Moreover, for the last two models there is no systematic trend for the error to increase dramatically in going away from the beta-stability line. In contrast to these two models, all the other models give results which differ strongly outside the beta-stability region.

3.6. Table of atomic masses

The individual experimental values of the atomic masses of various nuclides are presented in the form of a table. In doing this the experimental data are evaluated and made consistent and uniform in order to obtain the most reliable mass values. This procedure is necessary because every new experimental value of the atomic mass is the result of a single measurement and needs additional comparison with known data confirmed by several results. In connection with this, all the experimental data are categorized as fundamental (or primary) or secondary. The primary mass values are those which are calculated from several experimental results and, as a rule, from direct measurements of the differences of the masses of the measured nuclide and a reference nuclide. The secondary values are those found by using only one type of data. The primary data ensure cross-checking and uniformity of the secondary data.

The most recent, at the time of publication, evaluated atomic mass values are given in the table of the study by Audi and Wapstra.[32] This table lists the nuclide masses, mass excesses, nuclide binding energies, and beta-decay energies for more than 2500 nuclides. The values of the nuclide masses are given in mass units u (6) and in the energy equivalent (8), for some reason expressed not in eV/c^2, but in eV. The decay energies are expressed in eV. The values obtained directly in experiments are given for about 1850 nuclides, and for the other nuclides the quoted values were obtained by calculation on the basis of noticed trends. These nuclides are labeled by a special tag.

Nuclides for which the experimental values of the masses and decay energies deviate from the regular trends following from the experimental mass values of neighboring nuclei stand out strongly in this table. The experimental mass

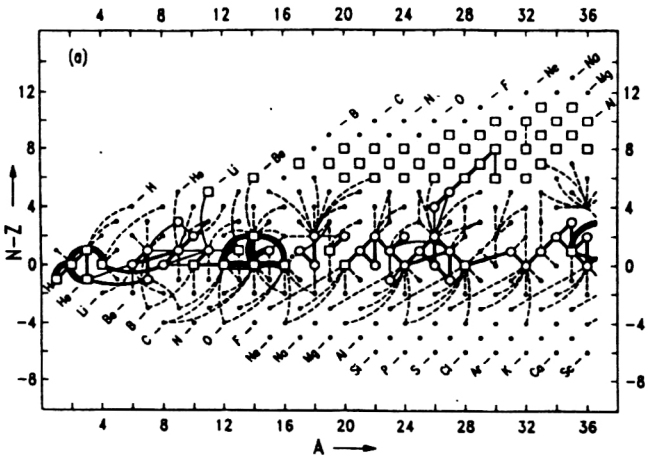

FIG. 5. Connection diagram for the values of the nuclide masses at the beginning of the chart of nuclides. Primary data: For nuclides whose atomic masses have been measured by the absolute-mass doublet method, i.e., using the atomic masses of ^{12}C,^{35}Cl, and ^{37}Cl as the reference masses—□. For other primary nuclides—○. Mass-spectrometric connections— =. Primary connections from nuclear-reaction data— —. Secondary data: For secondary nuclides whose masses have been determined only by one experimental measurement—●. For secondary nuclides whose masses have been determined from systematic trends in the experimental mass values—○. Secondary connections ----.

values are given for these nuclides, but the nuclides are given a special tag and mass values calculated on the basis of systematic trends by extrapolation and interpolation of the experimental data are recommended for them.

A special feature of the table is the use of the flow-of-information method,[33] where special matrices are used to analyze the influence of any input datum on the output adjusted mass values. This made it possible to judge the significance of any input results.

The connections of the input data are given in the table of Ref. 32 in the form of a diagram. As an example, in Fig. 5 we show a diagram of the connections of the input data for the initial part of the nuclide chart, i.e., for nuclides with mass numbers from 1 to 36. The connections of these data are explained in the figure caption. We see from this figure that the mass value of the above-mentioned nuclide ^{7}B is classified as a secondary, insufficiently established, datum.

3.7. The naming of transfermium elements

In the table of Ref. 32, as in the nuclide chart of Ref. 9, provisional, not officially confirmed, names were used for the last chemical elements. As is well known, the naming of the transfermium elements, i.e., the elements coming after fermium in the Periodic Table, has a long and confused history, and has been settled so some degree only very recently. There have been two groups of researchers, one at Berkeley in the USA and one at Dubna in Russia, who over many years have been involved in the synthesis of new transuranium elements. In recent years they have been joined by a group of German physicists working at Darmstadt. Each of these groups has had its own approach to the problem of discovering new elements and has made its own contributions to the synthesis of new elements and won the right to claim discovery and, consequently, to name the synthesized elements. The situation regarding the naming of new elements became so complicated that in the mid-1980s the IUPAP and IUPAC stepped into the problem in order to clear things up. A "Transfermium Working Group (TWG)" was created. This group was made up of prominent scientists with international credentials in nuclear physics and chemistry. The Working Group evaluated the contributions of individual groups to the synthesis of transfermium elements, published three reports on their activity,[34] and developed a recommendation for the naming of these elements for the Commision on Nomenclature of Inorganic Chemistry of IUPAC. In turn, the Commission on Nomenclature, guided by the then-current practice of naming chemical elements after the place where they were discovered, the properties of the element, or in the memory of a scientist, but not in honor of a living scientist, selected[35] the names of the transfermium elements given in Table I from those proposed by the scientists who had synthesized the new elements.

TABLE I. Naming of transfermium elements.

Atomic number Z	Name	Symbol
101	Mendelevium	Md
102	Nobelium	No
103	Lawrencium	Lr
104	Dubnium	Db
105	Joliotium	Jl
106	Rutherfordium	Rf
107	Bohrium	Bh
108	Hahnium	Hn
109	Meitnerium	Mt

The choice of names of the transfermium elements is explained in Ref. 35:

"Regarding elements 101–103, the Commission accepted the 'status quo' even though it recognized the conclusion of the TWG that an error had been made in the initial report on the discovery of element 102 (Nobelium).

"Element 104 was named Dubnium to recognize the distinguished contributions to chemistry and modern nuclear physics of the international scientific centre at Dubna near Moscow.

"Joliotium was chosen as the name for element 105 to recognize the French scientist F. Joliot-Curie who contributed greatly to the development of nuclear physics and chemistry, and who shared the Nobel prize in 1935 with Mme. I. Curie.

"Elements 106 and 107 were named after Ernest Rutherford (New Zealand) and Niels Bohr (Denmark), respectively, to recognize their distinguished contributions to our knowledge of atomic structure. The Commission recommends the name Bohrium (Bh) for element 107, instead of the proposed Nielsbohrium, so that it conforms to the names of the other elements named after individuals.

"Naming the adjoining elements 108 and 109 after Otto Hahn (Germany) and Lise Meitner (Austria) recognizes their decisive role in the discovery of nuclear fission."

This solution proposed by the Commission on Nomen-

clature is subject to ratification by the IUPAC council, which should occur on 10–11 August, 1995. This solution was published before its ratification so that comments could be collected on the naming of the transfermium elements.

In relation to this, we note that the American discoverers of element 101 (Ref. 36), "In recognition of the pioneering role of the great Russian chemist, Dmitri Mendeleev, who was the first to use the periodic system of the elements to predict the chemical properties of undiscovered elements, a principle which has been the key to the discovery of the last seven transuranium (actinide) elements," named the new element after Mendeleev, proposing the Latinized version Mendelevium. This name entered the Russian literature translated literally as "mendelevij," whereas in the Russian language all names connected with D. I. Mendeleev have spelling and pronunciation closer to that of the actual family name, for example: the settlement Mendeleevsk, the mineral mendeleevite, the Mendeleevskaya Line (a street in St. Petersburg), the Mendeleevskaya prize, the Mendeleevskiĭ Congress, and so on. Obviously, it is more correct to call element 101 "mendeleeviĭ" in Russian and "mendeleyevium" in English. This remark is independent of whether or not this name will be officially accepted; the goal is to use the name for element 101 which is the most accurate in the view of the compatriots of the great Russian chemist.

We also note that certain scientists responsible for the synthesis of transfermium elements have expressed disagreement with the solution proposed by the Commission on Nomenclature of Inorganic Chemistry of IUPAC and use or propose other names. In one issue of *Bulletin of the Atomic Scientists*, Seaborg, coauthor of the discoveries of more than ten transuranium elements and winner with MacMillan of the Nobel Prize in 1951 "for their discoveries in the chemistry of the transuranium elements," writes: "For the first time in history it may happen that the recognized, indisputed discoverers of an element are deprived of the privilege of naming it." The occasion for this was the refusal of the Commission to accept the suggestion that element 106 be named "Unnilhexium" (meaning one hundred six), Seaborg's name, already used in, for example, Ref. 9.

In this review we shall use the names proposed by the Commission in order to avoid confusion in the topic under discussion.

4. SOME PROBLEMS IN NUCLEAR PHYSICS REQUIRING THE USE OF MASS ANALYZERS. REQUIREMENTS ON THE ACCURACY OF ATOMIC MASS MEASUREMENTS

There are several problems in nuclear physics which require the use of a mass analyzer of some type for the identification and measurement of the nuclide masses. The accuracy with which the atomic masses of nuclides should be measured is determined by the requirements in solving specific problems. These requirements in turn define other conditions under which the mass analysis is carried out, namely, its sensitivity, efficiency, and speed. Below, we shall consider several of these, including some incompletely studied problems in nuclear physics which require the use of mass analyzers. These problems are either already being solved, or may be solved in the near future.

4.1. Nuclide identification

One of the problems frequently encountered in nuclear physics is that of identifying nuclides in nuclear reactions and nuclear transformations according to their mass number A and atomic number Z.

Identification of nuclear fission fragments

Identification according to A becomes necessary in, for example, studying the fine structure of the mass yield of fission fragments of heavy nuclei, which reflects the influence of filled shells on the fission process. Determination of the fine structure of the mass yield requires mass analysis with an error of at least no more than one mass unit u. Since the average mass of, for example, heavy fragments is 140 u, to solve this problem the mass analyzer must at least be able to separate the fragments and measure their masses with an error of no more than $\Delta M/M = 1:140 = 0.007$ or 0.7%. [Here Δ denotes the spread in mass values, in contrast to $\Delta(A,Z)$, denoting the mass excess.]

Magnetic and electric fields separate ions on the basis of the ratio of their mass and ionic charge q, i.e., M/q. Determination of the fine structure of the mass yield of fission fragments requires rapid mass analysis, excluding the change of the masses of the primary fragments as a result of delayed-neutron emission due to beta decay of the fragments. It is also necessary that the acceptance of the mass analyzer be independent of the atomic number Z, i.e., of the chemical nature of the fission fragments. Mass analyzers which directly use the fragment momenta and ionic charges obtained during the fission process best satisfy these requirements. Mass analyzers whose operation is based on deceleration of the fragments, neutralization of their ionic charge, and then reemission and reionization down to, for example, the singly charged state followed by acceleration do not work well for this problem.

For heavy fission fragments the equilibrium ionic charge in the target matter is $q = (20-25)q_0$, and therefore $\Delta q/q = 0.04$. Therefore, heavy fragments can have values of A/q which either differ by a very small amount ($131/22 = 5.955$ and $138/23 = 5.957$) or which are equal to each other ($132/22 = 6.0000$ and $138/23 = 6.0000$). In the second case the fragment separation can be based only on the difference between their mass excesses. In summary, in this and in other cases the total resolution of the mass analyzer in $\Delta(M/q)/(M/q)$ must be at the level $(\Delta M/M) \times (\Delta q/q) = 3 \cdot 10^{-4}$.

Separation of the fission fragments in this problem requires, to get a bit ahead of ourselves, use of a mass analyzer simultaneously with magnetic and electric deflection of the fission fragments ensuring velocity focusing of the fragments and thereby the needed resolution.

A very interesting problem in fission physics is that of the correlation between the yield of a certain pair of fission fragments and the number of emitted neutrons, which is determined by the mass defect in the fission. It was found in

Ref. 37 that in the spontaneous fission of ^{252}Cf the average neutron multiplicity is 4.0 ± 0.1 in the fission into two complementary fragments of a number of isotopes of Mo and Ba identified according to their gamma radiation. Here deviations of up to 10 are observed in the number of emitted prompt neutrons.

A more complete solution of this problem can be obtained by using two mass analyzers which record and identify each pair of complementary fission fragments according to A, with simultaneous detection of prompt neutrons in coincidence.

Extensive information about problems in the physics of nuclear fission can be found in the detailed monographs of Refs. 38 and 39.

Separation and identification of light nuclear fragments

A similar situation with spatial separation and resolution arises in the isolation and identification of several light products of nuclear reactions produced as a result of the fragmentation of energetic bombarding particles. For example, in the bombardment of a hydrogen or carbon target by lithium ions of energy 50A MeV, ^{6}He and ^{3}H nuclei are formed with large cross section and leave the target with identical velocities, roughly equal to that of the bombarding particles. Energetic ^{6}He nuclei are interesting for use as exotic bombarding particles, for example, for exciting a soft dipole resonance. This requires fairly reliable separation of the beam of secondary ^{6}He nuclei from the primary bombarding particles and from the other reaction product,^{3}H. Monochromatization of the secondary nuclei is also necessary.

The values of M/q for ^{6}He and ^{3}H, which are close to 3, differ only by 0.2% just because of the difference between their mass excesses $\Delta(A,Z)$. The resolution of only a magnetic mass analyzer is insufficient for accurately separating two beams of these products having some velocity spread. In this case it is necessary also to use a degrader in the magnetic mass analyzer, which differentially lowers the kinetic energy of the analyzed nuclides as a function of their atomic number Z and thereby ensures the separation and identification of nuclides with close values of M/q. There are three types of kinetic-energy degrader which differ in the details of their ion–optical operation.

Identification of heavy products of fusion reactions

The requirements on the identification of the products formed in the fusion of heavy ions and heavy nuclei with formation of a compound nucleus can be illustrated for the example of a possible reaction synthesizing nuclei of a new, for example, the 110th, element:

$$^{244}\mathrm{Pu}+^{34}S=^{(278-xn)}110+xn \quad \text{for } x=4-8.$$

For energy of the bombarding ions $\cong$250 MeV, needed to overcome the Coulomb barrier between the colliding nuclei, the reaction products (recoil nuclei) acquire an energy $\cong$30 MeV, and their equilibrium ionic charges in the target material are $q=(10-15)q_0$. The relatively broad spectrum of values of M/q of the reaction products makes it very difficult to separate them from the primary bombarding particles and to spatially focus the reaction products by a magnetic mass analyzer. In this case the problems of separation and focusing are solved by using either a gas-filled magnetic mass spectrograph or a mass spectrograph with magnetic and electric fields. In the first case, along with magnetic separation the recoil nuclei undergo focusing in their ionic charge as a function of velocity by charge exchange of the ions as they move in the low-pressure, light gas filling the chamber of the magnetic mass spectrograph. In both cases it is possible to separate the recoil nuclei from the beam of bombarding particles passing through the thin target and to identify the reaction products, at least according to their mass number A. This requires a relative resolution of $\Delta A/A=0.4\%$.

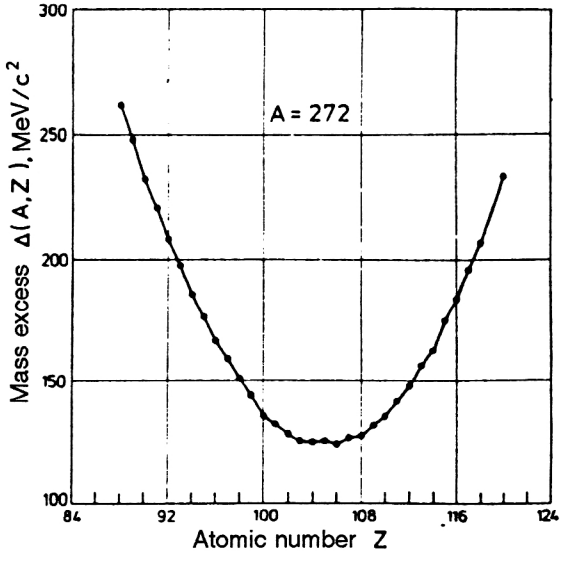

FIG. 6. Mass excesses of nuclide-isobars with mass number $A=272$ as a function of the atomic number Z.

The identification according to the atomic number Z of heavy reaction products undergoing alpha decay is usually done through the "genetic" coupling of the initial parent nucleus with the following known daughter, granddaughter, and great-granddaughter nuclei, established by recording the alpha-particle emission sequence. In addition, it is possible to identify heavy nuclei according to Z by measuring their mass excess $\Delta(A,Z)$. In Fig. 6 we show the dependence of the mass excesses for nuclide-isobars with mass numbers $A=272$ constructed on the basis of the averaged predicted masses according to the mass models of Ref. 29. It follows from this figure that the reliable identification of the element with $Z=110$ in the above reaction in the presence of possible atoms of meitnerium, hahnium, bohrium, and rutherfordium from a mass analyzer requires a mass resolution ΔM of at least 3 MeV/c^2, i.e., at the level $\Delta M/M=0.001\%$.

In this problem a resolution at this level can be ensured by, for example, the use of a Penning trap on-line with a mass spectrograph which in turn functions on-line with a heavy-ion accelerator. Here the trap must ensure fast capture of recoil nuclei, decrease of their velocity by "cooling" by means of a gas or an azimuthal electric field, and precise measurement of the ratios M/q with the accuracy needed to determine not only the mass number A, but also the atomic

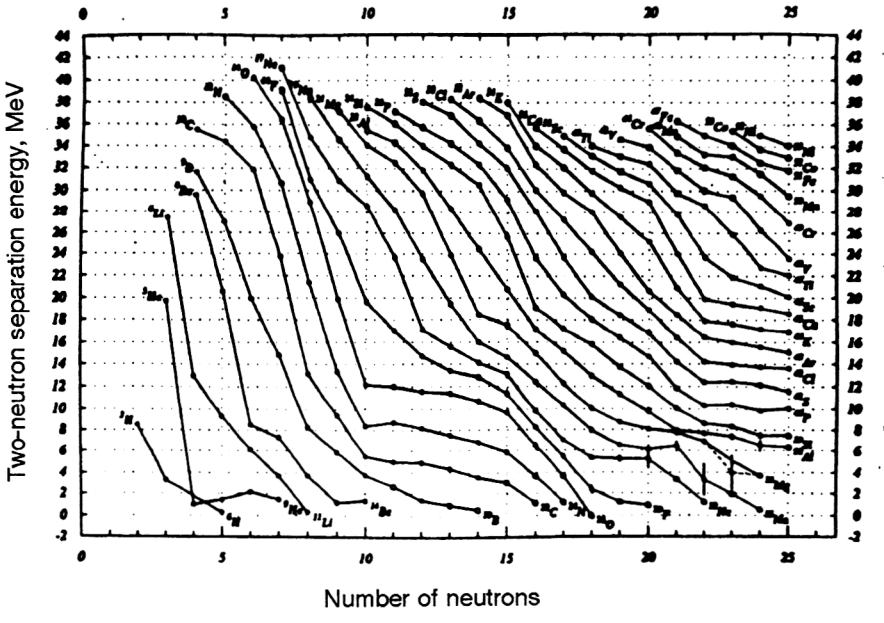

FIG. 7. Two-neutron separation energy for isotopes of elements from H to Ni (Ref. 41).

number Z. Projects for such setups are being developed at the present time.

Isomer separation and identification

An accuracy at least this high is required in mass analysis for the purpose of separating and identifying nuclides in the ground state and in a metastable state close to the ground state. As an illustration, let us describe the requirements on the separation of isomers of ^{178}Hf in the ground state and in the high-spin ($I^{\pi}=16^{+}$) long-lived ($T_{1/2}=31$ yr) metastable state with excitation energy 2.447 MeV. This high-spin state is a unique case of the realization of a trap on the so-called "yrast" line. In nuclear reactions[40] the high-spin isomer is produced in a ratio of about 1 to 20 with respect to the ground-state isomer, and the total amount of it is small (up to 10^{14} atoms).

For studying the nuclear-physics properties of the metastable isomer of ^{178}Hf without interfering admixtures it is necessary to separate it from a mixture of various nuclear reaction products. The extracted high-spin isomer can be used to prepare a target for study of the effect of exposure to neutrons and charged particles. In addition to chemical separation, here it is necessary to perform mass separation with a resolution $\Delta M/M=0.001\%$.

4.2. The neutron separation energy

The packing fraction $P(A,Z)=\Delta(A,Z)/A$ (2) and the binding energy per nucleon of a nuclide, $E_b(N,Z)/A$, referred to as the specific binding energy, depend smoothly on the mass number and do not reflect the periodicity of the nuclear structure. For more detailed understanding of nuclear structure a great deal of information can be obtained from the binding energy or, equivalently, the energy to remove the last nucleon or last two nucleons. This quantity is also expressed in terms of the nuclide mass. For example, the separation energies of the last neutron or of the last two neutrons are, respectively,

$$S_{1n}(N,Z)=[M(N-1,Z)+M(1,0)-M(N,Z)]c^2,$$

$$S_{2n}(N,Z)=[M(N-2,Z)+2M(1,0)-M(N,Z)]c^2,$$

and the separation energies of the last proton or last two protons are

$$S_{1p}(N,Z)=[M(N,Z-1)+M(0,1)-M(N,Z)]c^2, \quad (17)$$

$$S_{2p}(N,Z)=[M(N,Z-2)+2M(0,1)-M(N,Z)]c^2. \quad (18)$$

The two-nucleon binding energy is not sensitive to the effect of evenness or oddness of the number of nucleons in the nucleus and more prominently reflects the nuclear structure, for example, the nucleon filling of the nuclear shells.

In Fig. 7 we show the separation energy of the last two neutrons, $S_{2n}(N,Z)$, taken from Ref. 41 for a group of nuclei at the beginning of the nuclide chart, constructed according to the data of recent measurements of the nuclide masses. The violation of the monotonic behavior of the curves in the region of neutron-rich isotopes of Ne, Na, Mg, Al, and Si with $N=20$–23 and in the region of neutron-rich isotopes of Ca, Sc with $N=22$–23 should be noted. This effect of the relative increase of the two-nucleon separation energy by 1–2 MeV above the expected value is thought to be related to significant deformation of the nuclear shape.

The establishment of this effect required precise measurements of the masses of short-lived nuclei, which were begun in the 1960s in France, using a mass spectrometer with magnetic and electric fields, and were then continued by American and French experimentalists, respectively, using a time-of-flight mass spectrograph in the beam at a meson factory (LAMPF) and an energy-loss mass spectrograph in the beam of a heavy-ion accelerator (GANIL). In the recent experiments a resolution $\Delta M/M=10^{-4}$ was obtained, and the uncertainty in the mass measurement was better than $\Delta M/M=10^{-6}$.

Another feature to be noted in Fig. 7 is the decrease of the two-neutron separation energy for isotopes of N, O, F,

and Ne in the region $N=15$ and particularly for ^{27}F compared with the neighboring nuclides. On the other hand, the curves for S_{2n} show an increase of the stability of helium isotopes with $N>4$, known as the helium anomaly. This and many other problems of light nuclei are discussed in the detailed review of Ref. 42.

Further refinement of the behavior of the nuclide binding energy in this mass range apparently requires improvement of the resolution and accuracy of the mass measurements by at least an order of magnitude better than the values quoted above.

4.3. The nucleon pairing energy

Accurate measurements of the nuclear masses make it possible to analyze another important quantity, the nucleon pairing energy characterizing the subtle interaction between the particles inside the nucleus. The pairing energy, for example, for a pair of neutrons $P_{nn}(N,N-1;Z)$, where the parameters N and $N-1$ imply that the quantity of interest pertains to both the Nth, for example, even neutron and the $(N-1)$th odd neutron, is given by

$$S_n(N,Z)=S_{2n}(N,Z)/2+P_{nn}(N,N-1;Z) \tag{19}$$

$$S_n(N-1,Z)=S_{2n}(N,Z)/2-P_{nn}(N,N-1;Z). \tag{20}$$

Although the pairing energy arises directly from an even paired nucleon and disappears when it is removed, according to established tradition the pairing energy pertains to the two nucleons of the pair and is measured from the average value, as is done in (19) and (20). Then from (19) and (20) we find

$$P_{nn}(N,N-1;Z)=[S_n(N,Z)-S_n(N-1,Z)]/2. \tag{21}$$

The definition (21) differs from the analogous definition in Ref. 7 (p. 9) by a coefficient of $\frac{1}{2}$, owing to the above-noted ambiguity in the choice of the zero for measuring the pairing energy. For neutron pairs lying above and below in the order of filling of the levels we have

$$P_{nn}(N+1,N;Z)=[S_n(N,Z)-S_n(N+1,Z]/2, \tag{22}$$

$$P_{nn}(N-1,N-2;Z)=[S_n(N-2,Z)-S_n(N-1,Z]/2. \tag{23}$$

From Eqs. (21), (22), and (23) we directly find the average pairing energies characterizing the Nth even and $(N-1)$th odd neutrons in the nucleus:

$$P_{nn}(N,Z)=[S_n(N+1,Z)-2S_n(N,Z) + S_n(N-1,Z)]/4, \tag{24}$$

$$P_{nn}(N-1,Z)=[S_n(N,Z)-2S_n(N-1,Z) + S_n(N-2,Z)]/4. \tag{25}$$

In Eqs. (24) and (25) let us change from first differences to second ones, i.e., from the neutron separation energies directly to the atomic masses. We then obtain

$$P_{nn}(N,Z)=[M(N+1,Z)-3M(N,Z)+3M(N-1,Z) - M(N-2,Z)]/4, \tag{26}$$

$$P_{nn}(N-1,Z)=[-M(N,Z)+3M(N-1,Z) -3M(N-2,Z)+M(N-3,Z)]/4. \tag{27}$$

Equation (26) for the pairing energy of an even neutron in the nucleus is given in the monograph of Bohr and Mottelson,[43] but in a somewhat different form and without derivation, which makes it impossible to judge the origin of the scale on which the pairing energy is measured.

It follows from Eqs. (26) and (27) and relations similar to them that the determination of the nucleon pairing energy in the nucleus, which is very important for studying processes of double proton decay, double neutron decay, nuclear superfluidity, and the nuclear Josephson effect, requires measurement of the atomic masses with high accuracy, owing to the small value of the pairing energy and its dependence on second mass differences. For example, in order to calculate the pairing energy with an error of even 10% for its typical value of 2 MeV in the region of superfluid nuclei near tin, measurements of the atomic masses with a relative error of no more than $\Delta M/M=10^{-7}$ are required.

4.4. Radioactive decays of nuclides

Study of the radioactive decays of nuclides also requires precise values of the nuclear masses. As noted above, the nuclear-mass values determine the possibility or impossibility of any particular nuclear transformation and, in extreme cases, the possibility or impossibility for a particular nucleus to exist. A nucleus is considered to exist if it is energetically stable to instantaneous (in a time 10^{-22} sec) spontaneous emission of a proton or a neutron. This condition determines the two limits of the region of existence of nuclides from the neutron-rich extreme to the neutron-depleted extreme. Within these limits there are at present predicted to be about six to seven thousand nuclei. Under natural conditions on the Earth there exist 273 stable nuclides and 11 long-lived radioactive nuclides in secular equilibrium with which there exist another 41 radioactive nuclei. Approximately 1850 radioactive nuclides of the six to seven thousand possible ones are produced artificially at present.

Radioactive proton decay

The determination of the boundaries of stable nuclei is in general a difficult problem, since small errors in the reference values of the masses lead to uncertainty in the type of decay of the boundary nuclei (i.e., beta decay or nucleon emission) and move the boundaries closer or farther away. Figure 8, which is taken from Ref. 44, one of the first studies of the possibilities of nuclear proton decay and its experimental discovery, shows the predicted boundaries separating proton-stable and proton-unstable nuclei. That article mainly repeats the results of the working report of the USSR Academy of Sciences of 1955 by the same authors.

The boundaries between stable and proton-active nuclei are constructed on the basis of a liquid-drop formula like the Weizsäcker formula, taken from Ref. 45 with parameters going back more than 40 years. The following equation obtained from the mass formula was used to calculate the separation energy of the last proton:

$$S_p(N,Z)=931.14\{a_1-2a_2/3A^{1/3}+a_3[(A/2-Z)$$

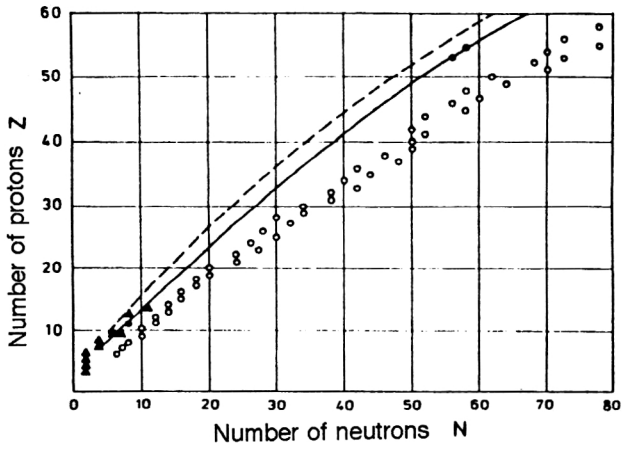

FIG. 8. Boundaries between nuclei which are stable and unstable to proton emission.[44] Lightest stable isotope of a given element—○; boundary of stable nuclei with odd Z— —; boundary of stable nuclei with even Z— ---; proton-unstable nuclei known at the present time—▲; proton-active nuclei known at the present time—●.

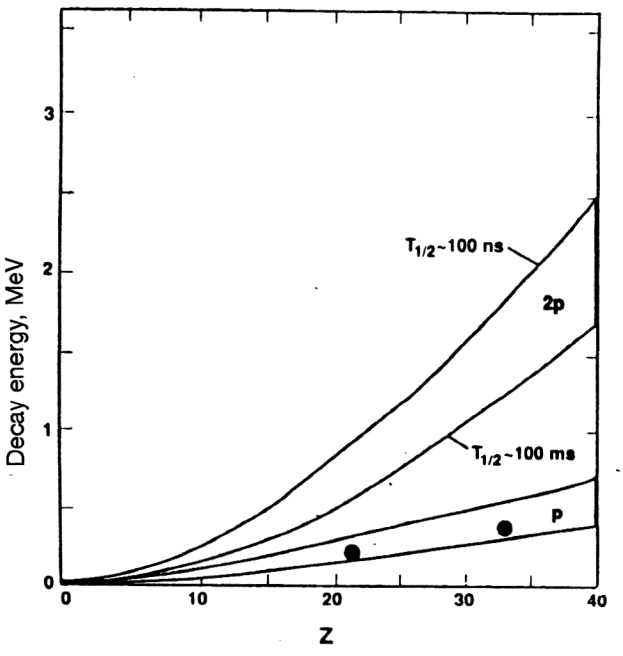

FIG. 9. Calculated values of the nuclear decay energies for proton decay (p) and double proton decay ($2p$) as a function of atomic number Z for two values of the half-life: $T_{1/2}=100$ nsec and $T_{1/2}=100$ msec (solid lines).[46] The points show the analogous calculated energies taken from Ref. 44 for the proton decay of the nuclei ^{39}Sc and ^{63}As with half-life $T_{1/2}=100$ msec.

$$+1/4]/(A-1)-a_4(2Z-1)/A^{1/3}+\delta\}, \quad (28)$$

where $a_1=0.015$, $a_2=0.014$, $a_3=0.083$, $a_4=0.00063$, and $\delta=\pm 0.036A^{-3/4}$, where in turn the plus and minus signs in δ respectively refer to nuclei with even and odd Z.

The boundary separating regions of nuclides which are stable and unstable to proton emission have been estimated by using Eq. (28) with the condition $S_p(N,Z)=0$.

The lifetime of nuclides lying beyond the boundary of stability to proton emission is to a large degree determined by the time for the proton to pass through the Coulomb barrier of the nucleus. In those cases where this time is significantly larger than the nuclear time, for example, equal to $\sim 10^{-12}$ sec, one can speak of the proton radioactivity of nuclei.

In the same figure we give additional nuclides that have been discovered up to now: the proton-active nuclides ^{19}Na,^{109}I, and ^{113}Cs, and nuclides undergoing prompt proton emission. We see from Fig. 8 that altough the predictions are quite old, there is rather good agreement between the results calculated in Ref. 44 and the current experimental results. Figure 8 is one example illustrating how mass formulas will be tested by future results.

Accurate values of the masses in proton decay are important not only for determining whether or not a decay is possible, but also for estimating the lifetime of a prospective proton emitter. Figure 9, taken from the review of Ref. 46, illustrates the importance of accurate mass prediction for the correct planning of experiments to seek proton-radioactive nuclides: an uncertainty of 0.2 MeV in the value of the proton separation energy for proton emitters in the region $Z=25$ leads to possible variations of the half-life from 100 nsec to 100 msec, i.e., by a factor of 10^6. In the same figure we give some of the results for the predicted half-lives of proton-active nuclei as a function of the decay energy $Q_p=-S_p$, taken from Ref. 44.

Double proton decay

More accurate predictions of nuclide masses are especially important in searching for another analogous phenomenon: double proton decay, or two-proton decay, which so far has not been discovered. This phenomenon was predicted by Goldanskiĭ in Ref. 47. It is completely analogous to the double beta decay known since the 1930s.

We recall that Goeppert–Mayer, in her study[48] published about a year after the creation of the Fermi theory of beta decay, noted that the masses of nuclide-isobars can vary with increasing Z in such a way that the emission of a beta particle by a nucleus becomes energetically impossible, whereas the emission of two beta particles at the same time by this nucleus will be energetically allowed. She wrote: "This nucleus would then be metastable, since it cannot go over into a more stable one by consecutive emission of two electrons. ... A metastable isobar can, however, change into a more stable one by simultaneous emission of two electrons."

The condition for double beta-minus decay for an atom of mass $M(A,Z)$ reduces to

$$M(A,Z+1)-E_{e1}>M(A,Z)>M(A,Z+2)-E_{e1}-E_{e2}, \quad (29)$$

where E_{e1} and E_{e2} are the binding energies of the last and next-to-last electrons in the atomic shell. It is necessary to include the binding energies of the electrons in atomic shells in order to exclude the possibility of single beta decay not only into the continuum, but also into a bound state, and to make double beta decay into a bound state possible. The

analogous conditions also occur in double positron decay and for paired simultaneous positron decay and orbital electron capture.

For double proton decay we have the condition

$$M(N,Z-1)+M(0,1)>M(N,Z)>M(N,Z-2)$$
$$+2M(0,1) \text{ or } +M(0,2). \quad (30)$$

The binding energies of electrons in shells of the hydrogen atom ^{1}H or helium atom ^{2}He, respectively, equal to 13 and 80 eV, are omitted here.

In a relatively recent study[49] the generalized Garvey–Kelson model was used to predict the most likely candidates for two-proton decay: the nuclei ^{39}Ti, ^{42}Cr, ^{45}Fe, and ^{49}Ni. However, experiments carried out specially to seek double proton decay of ^{39}Ti (Ref. 50) by using a magnetic mass spectrograph showed that this candidate for a two-proton emitter does not undergo double proton decay.

Figure 9 also illustrates the dependence of the lifetime of a two-proton emitter on the uncertainty in the value of the energy released in the decay. From this illustration and from the example given above of searching for double proton decay it is clearly seen to be necessary to raise the accuracy of direct measurements of the atomic masses in the region of neutron-depleted nuclides for more definitive prediction of possible two-proton emitters.

We note that there are some publications on the observation of double proton decay or two-proton decay, but not in the sense of the definitions (29) and (30). These are Refs. 48 and 47, and here the sense is that of the successive emission from a nucleus of two separate protons.

Double neutron decay

A phenomenon analogous to double proton decay can also occur in neutron emission from neutron-rich nuclei. Mass-formula calculations (see, for example, Ref. 29) predict that for many neutron-rich nuclei it is impossible to emit a single neutron and possible to emit two neutrons simultaneously. Among the possible nuclei of low atomic number which can be candidates for double neutron decay, ^{14}Be and ^{32}Ne turned out to be beta-emitters, and the others have not yet been synthesized. In relation to this it is interesting to study the recent results of measurements of the mass of the exotic nucleus ^{10}He, which lies near the neutron stability boundary.

The reaction of two-nucleon counter-exchange ^{10}B (^{14}C, ^{14}O) ^{8}He$+n+n$, namely, $(-2p+2n)$ relative to the target nucleus, was used to obtain ^{10}He in Ref. 51. The essential feature of this reaction was the use of maximally neutron-rich nuclides: a beam of radioactive ^{14}C nuclei and a radioactive and toxic ^{10}Be target. This made it possible to obtain a fairly low absolute value of Q of the reaction with negative sign. The experiments were carried out at the VIKSI accelerator complex in Germany.

In spite of being doubly magic, the ^{10}He nucleus is neutron-unstable. Therefore, its mass was measured in Ref. 51 by the well known method of measuring the mass of the second stable product of the binary nuclear reaction of the ^{14}O nucleus conjugate to the unstable ^{10}He nucleus.

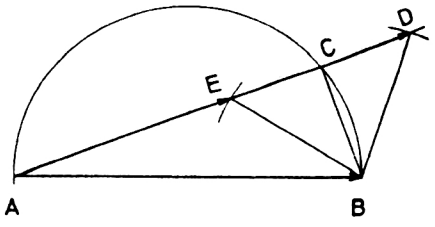

FIG. 10. Geometrical scheme for the connection between P_1, P_3, Q, E^*, and θ in a binary reaction according to Eq. (32). Given: AB$=P_1M_3/(M_1+M_2)$, BE$=$BD$=\{M_3M_4[2E_1M_2/(M_1+M_2)+Q-E]/(M_3+M_4)\}^{1/2}$, the angle BAD$=\theta$. We have $P_3=$AE and $P_3=$AD.

1. Measurement the extra (missing) mass. Here the mass of the unstable product of a binary reaction is determined by measuring the momentum of the second stable reaction product, and the value of the momentum is used to determine the reaction energy and, accordingly, the mass of the associated unstable product.

From the law of energy conservation in a binary reaction

$$M_1+M_2=M_3+M_4+Q/c^2 \quad (31)$$

and the law of momentum conservation

$$P_1=P_3+P_4, \quad P_2=0,$$

where M and P are the rest masses and momenta of the initial nuclei (1, 2) and the final products of the binary reaction (3, 4), Q, the energy released in the reaction appears in the nonrelativistic expression:

$$P_3=P_1M_3\cos\theta/(M_1+M_2)+\{2M_3M_4$$
$$\times[E_1M_2/(M_1+M_2)+Q-E^*]/(M_3+M_4)$$
$$-[P_1M_3\sin\theta/(M_1+M_2)]^2\}^{1/2}, \quad (32)$$

where P_3 is the momentum in the lab frame of the nucleon-stable reaction product M_3, conjugate to the nucleon-unstable product M_4, θ is the emission angle of the product M_3 in the lab frame, E_1 is the kinetic energy of the bombarding particle M_1, and E^* is the excitation energy of the secondary reaction products. The Q of the reaction and the excitation energy E^* are found from (32) by using the momentum P_3 of the nucleus M_3, and the former are then used in (32) to calculate the mass of the decaying nucleus M_4.

In Fig. 10 we show the scheme for writing the momentum P_3 (the vector AD or AE) as a function of the angle θ, together with the Q of the reaction and E^* (the vectors BD and BE) according to Eq. (32). In the construction we have also made use of the right angle ACB inscribed in the circle, using the circle diameter and the Pythagoras theorem.

The authors of Ref. 51, who had ^{14}C ions with well defined energy 334.4 MeV and a magnetic spectrograph of the Q3D type excluding kinematic effects of the reaction, found the energy of the ^{10}He ground resonance state to be $E(^{10}\text{He})=1.07\pm0.07$ MeV relative to the energy of the ^{8}He$+n+n$ state with a statistical reliability 98.7% and resonance width $\Gamma=0.5\pm0.1$ MeV.

In Ref. 52, ^{10}He was obtained from the breakup of ^{11}Li into ^{8}He$+n+n+p$. In turn, the beam of ^{11}Li ions with ki-

netic energy $(43\pm3)A$ MeV was obtained as a secondary radioactive beam by the fragmentation of ^{18}O ions accelerated in the RIKEN cyclotron in Japan. Production of ^{10}He was established by recording ^{8}He, using a dipole magnetic pulse analyzer with the simultaneous detection of two neutrons in coincidence with ^{8}He. The ^{10}He mass was determined by the following method.

2. The invariant-mass method. The essence of this method is described in Ref. 53. If we accept the usual definitions, the theory of the method can be reduced to the following.

From the general relation of relativity theory $M=M_{\text{rest}}/(1-\beta^2)^{1/2}$ it follows that

$$M=(M_{\text{rest}}^2+P^2/c^2)^{1/2}, \tag{33}$$

where M is the dynamical mass, M_{rest} is the nuclear rest mass, v and P are the velocity and momentum in the c.m. frame, and $\beta=v/c$. If we neglect small losses of the kinetic energy of the ^{11}Li nuclei in the thin target, we can write

$$M(^{11}\text{Li})=M(^{10}\text{He})+M(^{1}\text{H}), \tag{34}$$

i.e., we assume that the dynamical mass is invariant in the ^{11}Li breakup reaction. Using Eqs. (33) and (34), the authors determined the rest mass $M_{\text{rest}}(^{10}\text{He})$ by using the time-of-flight method to measure the velocity of the primary particles and using a dipole magnet to measure the momenta of the secondary reaction products. Such measurements did not require strict monochromaticity of the primary particle beam, which was very helpful in using nonmonoenergetic nuclear reaction products as the bombarding particles.

In Ref. 52 the values $E(^{10}\text{He})=1.2\pm0.3$ MeV and $\Gamma<1.2$ MeV were found for the ^{10}He resonance state. We note that from the relations (12) and (14) and from the currently known values of the ^{9}H, ^{9}Li, ^{10}Be, ^{11}Be, and ^{11}Li masses[32] we obtain the mass excess $\Delta(^{10}\text{He})=49.09$ MeV/c^2 or $E(^{10}\text{He})=1.35$ MeV.

The three authors of Refs. 51, 52, and 54 were awarded the G. N. Flerov prize (JINR, 1995) for their investigations of the properties of light nuclei.

The weighted results of Refs. 51 and 52 are shown in the scheme constructed in Fig. 11. There we give the results of these and several other studies on the energy of the lowest $^{9}\text{He}=^{8}\text{He}+n$ resonance state: $E(^{8}\text{He}+n)=1.27$ MeV with resonance width $\Gamma=0.30\pm0.07$ MeV (see Ref. 54).

The scheme in Fig. 11 shows that one of the decay branches of the ^{10}He resonance state, which lives $\hbar/\Gamma=2\times10^{-22}$ sec, can apparently be double neutron decay. This is expressed in the fact that emission of a single neutron from the most probable state of ^{10}He to the most probable state of $^{8}\text{He}+n+n$ does not occur, since $E(^{10}\text{He})=1.07$ MeV$<E(^{9}\text{He}+n)=1.27$ MeV, while emission of two neutrons at the same time by the ^{10}He nucleus, allowed by energy conservation $E(^{10}\text{He})=1.07$ MeV$>E(^{8}\text{He}+2n)=0$, does occur.

Whether or not the successive emission of two neutrons from ^{10}He via the $^{9}\text{He}+n$ resonance state is possible is determined only by the region of overlap of the widths of the ^{10}He and $^{9}\text{He}+n$ resonance states. This overlap, assuming the simplest so-called triangle distribution of level densities,

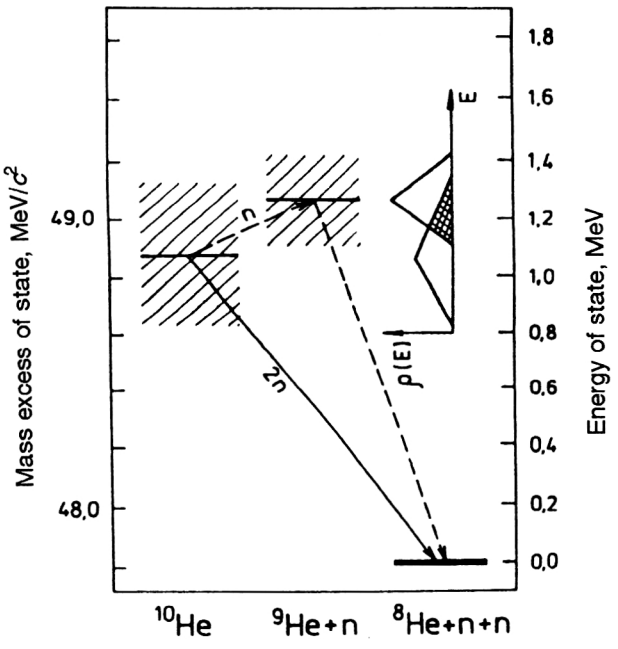

FIG. 11. Energies of the ^{10}He, $^{9}\text{He}+n$, and $^{8}\text{He}+n+n$ states (Refs. 51, 52, and 54).

also shown in Fig. 11, makes up about one fourth of the integrated value of the density of states of the initial level. Moreover, the emission of a single neutron from ^{10}He can be additionally suppressed by the centrifugal barrier (more precisely, the centripetal barrier hindering neutron emission) due to the p state ($l=1$) of the eighth neutron in ^{10}He. There is no centrifugal barrier for the emission of a pair of neutrons ($j=0$).

More accurate determination of the frequency of cases of the successive emission of two neutrons requires more detailed study of the quantum-mechanical transition between the two levels.

For the detailed study of the neutron-emission mechanism in the double neutron decay of ^{10}He, it might be more informative to use measurements of the actual momentum distributions of the ^{8}He nuclei produced in the decay of ^{10}He than measurements of the momenta of the conjugate stable reaction products. In this case the width of the distribution of momentum spreads will directly reflect the mechanism of two-neutron emission from the unstable ^{10}He nucleus. The shape of the distribution can be used to judge how neutrons are emitted: independently or in a bound virtual dineutron state.

For example, in the case of the reaction considered above, $^{10}\text{Be}(^{14}\text{C},^{14}\text{O})^{8}\text{He}+n+n$, for ^{14}C ion energy equal to 334 MeV, the relative spread of momenta of ^{8}He nuclei will be $\Delta P/P=5.4\%$ in the case of double neutron decay and $\Delta P/P=3.8\%$ in the case of the successive emission of two neutrons via the ^{9}He resonance state. The momentum of the background ^{8}He nuclei produced in the reaction $^{10}\text{Be}(^{14}\text{C},^{16}\text{O})^{8}\text{He}$ will differ considerably from the measured momentum.

However, the large ^{8}He momentum in this reaction due to the sum of the primary momentum and the momentum

from the deexcitation of the binary nuclear system (AD =AC+CD in Fig. 9) may be inappropriate for a magnetic analyzer. The reaction $^{14}C(^{26}Mg, ^{30}S)^{10}He$ should be more accessible for testing the studied effect. This reaction has $Q=-48$ MeV, and for the bombarding particles of energy $\sim 20\ A$ MeV the ^{8}He momentum due to the difference of the constituent momenta (AE=AC−CE in Fig. 9) is ~1 GeV/c, which is quite acceptable for an analyzer of average magnetic rigidity. The spread of the ^{8}He momentum distribution in this reaction will be 24% and 9%, respectively.

We note that the pairing energy of the seventh odd neutron in the ^{10}He nucleus determined from Eq. (27) is 1.25 MeV. This energy may be manifested as the binding energy of the two simultaneously emitted neutrons in the initial stage of double neutron decay.

A high-resolution magnetic spectrometer of the QD type with cancellation of the kinematic effects of nuclear reactions is being built at the present time in the Flerov Laboratory of Nuclear Reactions of the JINR. The spectrograph will be based on a tunable magnetic channel of the primary and secondary beams of the U-400M cyclotron and will operate in the energy-loss mode, making it possible to have high quasimonochromaticity of the beam of bombarding particles, while preserving the full beam intensity by means of gapless "monochromatization." This will be ensured by combining the dispersive capabilities of the magnetic channel of the primary ion beam and the QD magnetic spectrograph.

Double alpha decay

If one attempts to further extend the study of double-decay processes—double beta decay, double proton decay, and double neutron decay, the question of whether or not double alpha decay is possible necessarily arises. Comparison of the nuclide mass values shows that there are apparently no nuclei satisfying exactly the condition for double alpha decay analogous to (29) and (30). However, decay similar to double alpha decay can be expected for ^{143}La. The accuracy in atomic-mass measurements in the region of this nucleus enables us to speak of possible ^{143}La decay into ^{135}I by simultaneous emission of two alpha particles with total energy release 730±30 keV, $^{143}La=^{139}I+2\cdot^{4}He$, with strongly inhibited emission of a single alpha particle by the same nucleus, $^{143}La=^{139}Cs+^{4}He$, due to the very small energy release 80±20 keV. This relationship between the energies of the two successive alpha decays is due to filling of the shell $N=82$ in the region of these nuclei, which leads to possible alpha decay of neutron-enriched nuclei, in contrast to the known alpha conversion of neutron-deficient nuclides. Calculations using the simplest expression for the probability for a charged particle to penetrate the Coulomb barrier

$$P\sim\exp(-K), \quad K=\int_{r_1}^{r_2}[2M(V-Q)]^{1/2}dr$$

show that the probability for the simultaneous emission of two alpha particles in this case is several hundred times higher than that for the emission of a single alpha particle. An analogous situation holds for the passage of a virtual cluster ^{8}Be bound in the nucleus ^{143}La by the pairing energy of two alpha particles.

Since the exact conditions for double alpha decay [like the conditions (30)] are not satisfied in this case, while it is expected that the simultaneous emission of two alpha particles will dominate over single alpha decay, this possible type of decay can be termed quasi-double.

A possible scheme for experiments to seek quasi-double alpha decay might be the following. A thin ^{235}U target is bombarded by an intense beam of thermal neutrons in a nuclear reactor. Nuclei of ^{143}La ($T_{1/2}=14.3$ min) as fission fragments are collected at the collector of a mass spectrograph composed of a dipole magnet and an electrostatic analyzer. The collected fission fragments are then analyzed by a sensitive mass spectrometer for content of nuclides with mass numbers 143, 139, and 135 and by a laser for content of the chemical elements neodymium, lanthanum, and barium, which are the final products of chains of beta decays of the initial nuclide and the daughter nuclides from alpha decay.

Bound-state beta decay

The recent discovery of beta decay into a bound state[55]

$$^{163}Dy^{66+}=^{163}Ho^{66+}+\nu_e \qquad (35)$$

imposes new requirements on the accuracy of atomic-mass measurements.

The mass difference of the neutral atoms ^{163}Dy and ^{163}Ho known from Ref. 32 leads to capture by the holmium nucleus of an electron from an atomic shell of dysprosium with capture energy $Q_{ec}=2.6-2.7$ keV. The possibility of the inverse reaction is determined in this case by the difference between the electron binding energies in shells of the holmium and dysprosium atoms, which determine the mass ratio in the process (35). The table of atomic masses in Ref. 32 gives the experimental values of the masses of, as a rule, neutral atoms or, in some cases, singly charged ions. The difference between the masses of the bare dysprosium nucleus circulating in a storage ring and of the hydrogen-like holmium ion leading to the inverse process—beta decay into a bound state—can be calculated from the expression

$$M(N,Z)^{Q+}=M(N,Z)-QM_e+E_3(q)/c^2,$$

where $M(N,Z)^{Q+}$ is the mass of the ionized atom with charge number $Q=q/q_0$, M_e is the free electron mass, $E_e(Q)$ is the binding energy of Q electrons in the atomic shell, and $M_e=(548579.903\pm0.013)$ nu$=(510999.06\pm0.15)$ eV/c^2.

The calculated values of the electron binding energies in atoms can be found in the table of Ref. 56, but the accuracy with which these values is determined is not known. Therefore, for example, the difference of the total binding energies of 66 electrons in the Dy and Ho atoms can be determined with an error considerably larger than the error with which the atomic masses are determined. A calculated value of the energy of bound-state beta decay equal to 50 keV is quoted in Ref. 55 without any error.

Using an ion storage ring as a mass spectrograph, the authors of Ref. 55 measured the half-life $T_{1/2}^{\beta b}=48+3$ d for the bound-state beta decay of ^{163}Dy, but could not resolve the $^{163}Dy^{66+}$ and $^{163}Ho^{66+}$ mass doublet ($\Delta M=50$ keV/c^2, $\Delta M/M=3.3\times10^{-7}$) in order to experimentally confirm or reject the calculated decay energy, the exact value of which is very important for understanding this new process.

The authors of Ref. 55 assume that bound-state beta decay is a two-body process, the final participants in which are only the daughter nucleus and a monoenergetic neutrino. However, it is impossible to exclude the possibility that in some fraction of cases, determined by the electromagnetic interaction constant, internal bremsstrahlung analogous to that in electron capture will arise in bound-state beta decay. This follows from the fact that the electron acceleration inducing the bremsstrahlung in electron capture is identical but opposite in sign to the "acceleration" in the electron deceleration in the Coulomb field of the nucleus in bound-state beta decay. Then, instead of a two-body process, in a small fraction of cases of bound-state beta decay a kinematically less well defined three-body process must occur. This phenomenon can be studied by measuring the mass difference of the participants in bound-state beta decay and the recoil momentum distribution of the daughter nucleus. In the limiting case in such an experiment it may be possible to measure the rest mass of the neutrino emitted in bound-state beta decay.

A possible experiment to study internal bremsstrahlung in bound-state beta decay and perhaps to measure the neutrino mass is the following. The parent and daughter nuclei of bound-state beta decay (or electron capture) orbiting in a storage ring undergo momentum selection, and their mass doublet is resolved. Then the selection is stopped and the line-broadening of the daughter nucleus in the frequency spectrum of the ion circulation, which arises from the storage of new daughter nuclei from bound-state beta decay (or electron capture), is recorded. This provides the momentum spread as a result of the emission of bremsstrahlung photons and neutrinos.

In the relativistic treatment taking into account terms through second order in the small parameter, the momentum broadening is

$$\Delta P/P=\pm(Q/Pc)\{1-(M_\nu^2c^4/2Q^2)[1-(Q/Mc^2)]\}, \tag{36}$$

where $M=M(N-1, Z+1)^{Z+}$ is the mass of the daughter nucleus after beta decay of the nucleus $M(N,Z)^{Z+}$ into a bound state, M_ν is the neutrino rest mass, and $Q=Q_{\beta b}$ is the energy released in bound-state beta decay.

It follows from Eq. (36) that the half-width of the momentum distribution of $^{163}Ho^{66+}$ ions, which have energy $300A$ MeV in the storage ring, is $\Delta P/P=\pm3.8\times10^{-7}$, owing to the recoil momenta from the emission of internal bremsstrahlung photons and massless neutrinos. This spread is about Q/Mc^2.

The shape of the recoil momentum distribution makes it possible to judge whether only a neutrino or a neutrino and internal bremsstrahlung photons accompany bound-state beta decay.

If we assume that the neutrino emitted in bound-state beta decay has nonzero rest mass, the momentum broadening must be decreased. For a neutrino rest mass $M_\nu=5$ eV/c^2 the decrease of the momentum spread is $\Delta P/P=10^{-4}$ of the momentum spread arising from the bremsstrahlung-photon momenta. In this case a principle analogous to the well-known nonius (vernier) principle makes it possible to perform measurements with a relative error of 10^{-11}.

We note that according to the recently published experimental data of Ref. 57, the neutrino mass is estimated to be $M_\nu<7.2$ eV/c^2 at the 95% confidence level.

Similar investigations can be carried out for nuclides undergoing electron capture from the shell of a hydrogen-like ion. For example, let us consider the electron capture $^{37}Ar^{17+}=^{37}Cl^{17+}+\nu$. The decay kinematics in this case will be the two-body kinematics owing to the absence of electrons in the shell of the daughter nucleus and, accordingly, the absence of characteristic x-ray emission, and also owing to the absence of gamma-ray emission from the daughter nucleus (in the case of electron decay of ^{37}Ar).

The actual manifestation of the frequency spread of the ion circulation as a function of the momentum spread depends on the storage-ring parameters and, in particular, on the broadening coefficient of the equilibrium orbit when the momentum is changed.

The known methods of electron cooling of ions in a storage ring produce velocity-monochromatized ions with an error of at least 10^{-6}. The laser cooling method, which ensures a monochromatization several orders of magnitude better, is not applicable to bare nuclei (if we do not have in mind intranuclear gamma transitions and gamma lasers) and in practice is not applicable also to hydrogen-like heavy ions. This problem of momentum selection of stored ions can apparently be solved by using a superconducting and, accordingly, time-independent solenoid. The ion selection in this case can be based on resonance buildup of betatron oscillations of secondary ions in the storage ring, analogous to the buildup of oscillations in the well known high-frequency quadrupole mass spectrometer.

Cluster decay

It follows from the behavior of the packing fraction as a function of mass number (see Fig. 1) that heavy nuclei release energy in decays with the emission of not only alpha particles, but also heavier clusters. Theoretical calculations[58] and experiments[59] have shown that this new type of decay exists in the region of heavy nuclei with ^{208}Pb produced as the daughter nucleus. New calculations[60] have demonstrated the possibility of cluster decay also in another nuclear region, namely, the possibility of decay into daughter nuclei close to the doubly magic ^{100}Sn nucleus. One example of the decay in this new region of nuclides might be ^{114}Ba decay with emission of ^{12}C, which has been the object of experimental searches.[61] However, these experiments require more accurate values of the masses of nuclides near ^{100}Sn.

The ^{100}Sn mass value quoted in the table of Ref. 32 is that obtained from systematic regularities in the behavior of the experimental masses and has an absolute error ±0.45 MeV ($\Delta M/M=\pm5\times10^{6}$), which introduces a large

uncertainty into the calculations of the half-lives of cluster decay. An increase of the relative accuracy of measuring the masses of ^{100}Sn and atoms in this region of nuclides by at least a factor of 10 would significantly improve the basis for designing and performing experiments to search for new cluster radioactivity.

4.5. Synthesis of the nuclide ^{100}Sn

The study of nuclei like ^{100}Sn with two filled nucleon shells is of special interest for checking and improving nuclear models. According to shell models, such a nucleus should have an energy gap between levels of neighboring shells equal to 6.5 MeV, and should have the clearly manifested properties of doubly magic nuclei and also the properties of superfluid nuclei.

The study of nuclei very far from the beta-stability line is important also for astrophysical theories. In particular, neutron-deficient nuclei can, under the conditions prevailing in stars, give rise to so-called rapid proton capture.

This has stimulated the authors of Refs. 62 and 63 to perform experiments on the synthesis of nuclei near ^{100}Sn, the heaviest doubly magic nucleus with $Z=N$, lying 18 neutrons from the beta-stability line and the fifth such nucleus after ^{4}He, ^{16}O, ^{40}Ca, and ^{56}Ni.

The experiments of Ref. 62 were carried out at the UNILAC SIS accelerator complex in Germany. In these experiments, ^{124}Xe ions of energy 1095A MeV were used to obtain ^{100}Sn nuclei.

The experiments of Ref. 63 were performed at the GANIL accelerator with a beam of ^{112}Sn ions accelerated to 63A MeV. The enriched isotope ^{112}Sn from the store created at the initiative and under the direction of Academician I. V. Kurchatov was used in the experiments. The nuclear-reaction products produced in bombardment of the ^{58}Ni (68.3%) target were separated and identified in Z, A, and q, using a doubly achromatic magnetic mass spectrograph with a Wien filter placed at the end of the mass spectrograph and with a solenoidal superconducting lens and an additional magnetic spectrograph placed in front of the main mass spectrograph. A total of 24 events of ^{100}Sn production were detected. The results are summarized in Fig. 12.

The authors of Ref. 63 view the production of tin isotopes and isotopes of other close nuclides as the results of the fragmentation of the bombarding particles at intermediate energies. However, another interpretation of this reaction, described in connection with other experiments in Refs. 64 and 65, is not excluded.

The authors of Ref. 64 studied practically the same reaction ^{124}Sn+^{58}Ni, but in the direct kinematics (the reverse of the so-called inverse kinematics of the preceding reaction), for energy of the ^{58}Ni ions equal to 249 MeV. The authors of Ref. 65 studied the reaction ^{120}Sn+^{112}Sn for energy of the ^{112}Sn ions equal to (4.25−4.8)A MeV. In both experiments multineutron transfers of up to six neutrons were observed. The authors of Refs. 64 and 65 view these reactions as a possible manifestation of the nuclear Josephson effect.

The nuclear Josephson effect. In a review on double proton decay,[66] Gol'danskiĭ writes, "When two-proton radioactivity is compared with other physical phenomena, it is easy to observe a very close analogy with electron tunneling between superconducting and normal metals under the influence of a potential difference exceeding the superconducting gap width." In a subsequent study by Gol'danskiĭ and Larkin[67] entitled "An analog of the Josephson effect in nuclear transformations," the phenomenon of the tunneling transition of paired nucleons from one nucleus to another in a near head-on Coulomb collision is analyzed in more detail.

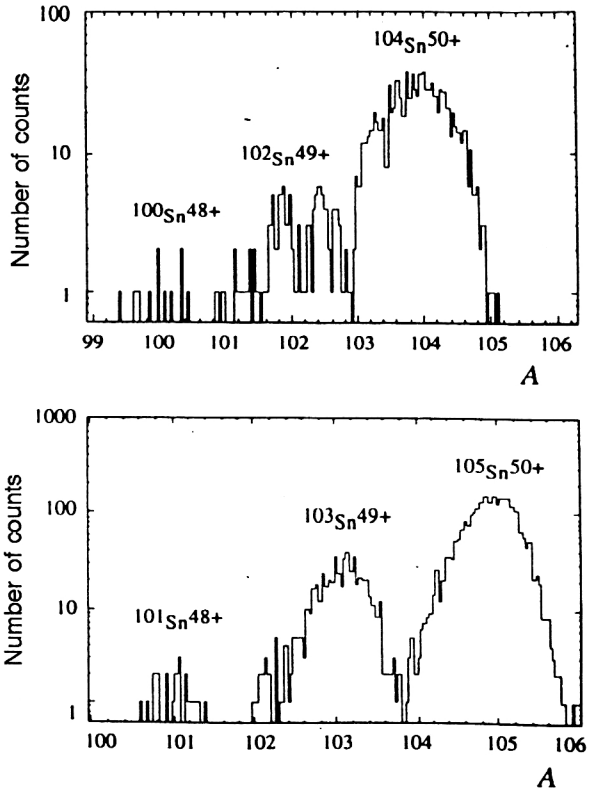

FIG. 12. Yields of tin isotopes in the experiments of Ref. 63. The masses were obtained by using the data from magnetic analysis of the reaction products, time-of-flight measurements, and identification in Z.

We recall that in 1962 Josephson showed theoretically[10] that a superconducting current of electron Cooper pairs is possible through a dielectric sandwiched between two superconductors. The appearance of this electron current through the dielectric is due to the enhancement of the tunneling probability for bound pairs as a result of the coherence of the superconducting states of two superconductors. The superconducting current through the dielectric appears in two forms: in the form of a stationary (direct) current under the action of an electromotive force due to the change of the magnetic flux, and in the form of a variable current with frequency given by Eq. (7) under the action of an applied potential difference.

The stationary Josephson effect in conjunction with the phenomenon of magnetic-flux quantization is used in the SQUID, a supersensitive device for measuring magnetic flux. The sensitivity of the device is 10^{-15} Wb, which is sufficient for magnetic cardiography. The nonstationary Josephson effect has been used in metrology, in particular, as men-

tioned above, to maintain the voltage standard on the basis of Eq. (7). It has also provided the basis for the design and development of supersensitive electromagnetic-radiation detectors.

Josephson and coworkers were awarded the Nobel Prize in 1973 for their discovery related to the phenomenon of tunneling in solids.

The authors of Ref. 67 considered a close encounter, without contact, of one heavy nucleus with another along a classical Rutherford trajectory and calculated the probability for a tunneling quantum-mechanical exchange of one or two protons. The effect of the superfluidity of the colliding nuclei on the proton tunneling probability was taken into account. In the end it was shown that when there is only one level near the Fermi surface with small angular momentum and when the change of energy of the tunneling nucleons is small (2−3 MeV), the ratio of the paired-nucleon transition probability and the product of the probabilities of two one-nucleon transitions becomes exponentially large. This by itself initiated a deeper theoretical treatment and experimental searches for a possible nuclear Josephson effect.

In the present review it is not possible to analyze the numerous theoretical studies of the nuclear Josephson effect and the attempts to discover it experimentally which were initiated by Refs. 66 and 67. We only note one feature of the superfluid transfer of paired nucleons which is important for a possible interpretation of the results quoted above on the synthesis of neutron-deficient isotopes of tin.

An important constraint on the superfluid transfer of pairs of nucleons in the collision of two nuclei is the need to emit or absorb the electromagnetic quanta in order to cancel the energy difference Q of the initial and final superfluid states. The well known process of energy dissipation via thermal excitation of a nucleus does not apply here, owing to the requirement that the nuclear superfluidity be preserved, which would be violated by thermal and vibrational excitations. The process of energy dissipation via the emission of electromagnetic radiation occurring over a time 10^{-18} sec is of low probability during the short time that the collision of two heavy ions takes place ($\sim 10^{-21}$ sec).

In the case of proton transfer the cancellation of the energy difference of the initial and final states of the colliding nuclei and, accordingly, the increase of the reaction probability can be ensured by changing the electrostatic nuclear interaction energy. This was discovered experimentally in Ref. 68, but in reactions not of the superfluid type, and has also been studied in Ref. 64 in connection with reactions of superfluid nucleon transfer.

In the general case of neutron or proton transfer the energy difference of the states can be canceled both by changing the electrostatic energy and by the effect of the change of the nuclear moment of inertia in the nucleon transfer, which leads to a change of the orbital rotation energy in the collision. This was first pointed out in Ref. 69 and independently again in Ref. 65.

According to Ref. 69, the general condition for the cancellation of the energy difference of the initial and final superconducting states Q due to the difference of the electrostatic energy before and after the proton transfer, $\Delta B = B_{12} - B_{34}$, and due to the change of the energy in the orbital motion has the form

$$\frac{(A_1/A_2)[1+(A_1/A_2^{1/3})]^2(1+A_3/A_4)^{8/3}}{(A_3/A_4)[1+(A_3/A_4)^{1/3}]^2(1+A_1/A_2)^{8/3}} = 1+(Q+\Delta B)/(E-B_{12}). \quad (37)$$

Equation (37) was obtained by using energy and momentum conservation and conservation of the location of the center of mass of the colliding nuclei, which interact only via internal forces in the nucleon-pair transfer. In the derivation we have assumed that during the time of the nucleon-pair transfer there is a weak sliding contact between the nuclei, which does not excite rotational perturbations which would destroy the nuclear superfluidity.

Some of the results of the classical treatment of the effect of the change of the orbital energy of two colliding nuclei as a result of the transfer of several pairs of neutrons according to Eq. (37) are given in Table II.

TABLE II. Comparison of the parameters of two alternative multineutron transfer reactions in the interaction ^{58}Ni+^{112}Sn at the energy 63A MeV.

Type of reaction	Reaction products	Q of the reaction, MeV	ΔE_{orb}, MeV	$Q_{\text{eff}}=Q+\Delta E_{\text{orb}}$, MeV
$-12n$	^{70}Ni+^{100}Sn	−33.0	+198	+165
$+6n$	^{52}Ni+^{112}Sn	−34.6	−162	−197

In this table we show the reaction considered above and another possible reaction with roughly the same value of Q. In the fourth column we give the calculated values of the change of the orbital energy of the two colliding nuclei ^{58}Ni+^{112}Sn as a result of neutron transfer from one nucleus to the other. Here we have assumed that all the excess kinetic energy of the bombarding nucleus above the Coulomb barrier goes into the orbital motion of the two colliding nuclei (2252 MeV). Such an energy transformation is possible if the impact parameter in the collision is roughly equal to the sum of the radii of the colliding nuclei, corresponding to weak sliding contact. In the last column we give the total effect, which shows that the removal of six pairs of neutrons from ^{112}Sn can be accompanied by the required transformation of initial energy in the orbital motion into internal energy of the nucleus. When three pairs of neutrons are captured by the same nucleus, the required expenditures of intranuclear energy not only are not compensated by the energy of the orbital motion, but, conversely, the neutron transfer requires an increase of the kinetic energy of the orbital motion in addition to the intranuclear expenditures of energy. This can be manifested in the value of the cross sections for the two branches of the nuclear reaction, which can be measured directly in experiments using a mass analyzer to measure the yield of tin nuclei with $A>112$ in comparison with the yield of tin nuclei with $A<112$ (see Fig. 12).

An achromatic magnetic mass analyzer of products of nuclear reactions in the beam of the U-400M cyclotron with very large angular and momentum acceptances is presently being built at the JINR Flerov Laboratory of Nuclear Reac-

tions. This can be used to perform experiments to explain the role of the hypothetical Josephson effect in the direct nuclear reaction $^{112}Sn+^{58}Ni$.

We note that the validity of the classical treatment of the Coulomb collision of two charged nuclei is determined by the well known criterion

$$\kappa = b/\lambda\!\!\!^{-} = 2q_1q_2/\hbar v \gg 1, \tag{38}$$

where b is the minimum distance between the colliding nuclei in a near head-on collision, or the collision diameter, equal to the two maximum impact parameters at which the incident nucleus is scattered into the backward hemisphere in the c.m. frame. Also, $\lambda\!\!\!^{-}$ is the reduced de Broglie wavelength, q_1 and q_2 are the charges of the colliding nuclei, and v is the relative velocity of the nuclei. To actually estimate the parameter in (38) it is more convenient to write it in the form

$$\kappa = kZ_1Z_2[(A_1+A_2)/A_2][A_1/E_1]^{1/2}, \tag{39}$$

where $k=0.314=(\text{“}\pi\text{”}/10)$ MeV$^{-1/2}$ (here “π” is a mnemonic, not a mathematical quantity), E_1 is the kinetic energy of the bombarding particles in the lab frame expressed in MeV, and Z and A are the atomic numbers and mass numbers of the colliding nuclei.

For the case considered above of a $^{58}Ni+^{112}Sn$ collision for energy of the tin ion equal to $63A$ MeV it follows from Eq. (39) that $\kappa=160$, which means that the classical treatment of this collision is valid.

The criterion (38) was formulated by Bohr in Ref. 70 and at first was referred to as the Bohr criterion. For example, in Ref. 71 we read: “Transparency 1 shows that we shall be treating the motion of the charged particles classically... . It is well known that the value of the parameter κ is very important in this regard. According to the well known argument of Niels Bohr, if this parameter is much larger than unity, the motion must be essentially classical...”

However, after some time this parameter became known as the Sommerfeld parameter without any justification. For example, this occurs in Ref. 72, which contains a series of lectures that the author has been giving to students since 1967.

A more detailed discussion of the history of this question, and also derivations of intermediate results related to this criterion but not presented by Bohr can be found in Ref. 73.

4.6. Other problems requiring atomic-mass measurements

An important application of magnetic mass separators is in the separation and identification of radioactive nuclides in nuclear spectroscopic studies. In such studies mass separators perform isotope separation in the mass number A (class-C identification), and in some special cases they also separate an isobar mixture in the atomic number Z (class-A identification), but sometimes using a mechanical conveyer.

As noted in Ref. 74, new measurements of the masses of the ^{35}Cl and ^{37}Cl atoms are important for metrology, as these atoms together with ^{12}C are reference nuclides in atomic-mass measurements.

Improvement of the unified mass unit u requires more accurate measurement of the mass of the ^{28}Si atom.

Improvement of the Faraday constant in electrolysis requires more accurate knowledge of the mass of ^{109}Ag.

Nuclides whose masses disagree with the values following from systematic trends are given in Tables B and C of Ref. 32. Naturally, here the masses need to be measured more accurately.

Testing the principle of CPT symmetry requires measurement of the proton–antiproton mass difference with an accuracy of at least $\Delta M/M=10^{-9}$. There are plans to carry out such measurements.[75]

CONCLUSION

In this review we have discussed basic concepts in the measurement of nuclide masses: the atomic mass, the mass number, the mass excess and deficit, the mass defect, and the nuclear binding energy in a historical context. We have described the evolution of the energy equivalent of the unit of atomic mass.

We have given a detailed discussion of the basic mass formulas of three classes: liquid-drop models and droplet models, shell models, and phenomenological models. We have commented on the recent table of atomic masses.

We have discussed the most important problems in nuclear physics requiring the use of mass analyzers for their solution. We have analyzed the requirements on the measurement accuracy imposed by these problems.

The most important of these problems are the identification of far transuranium elements by mass-analytic methods, the study of double neutron decay, the search for quasi-double alpha decay, the search for manifestations of the nuclear Josephson effect in nuclear reactions, and the study of beta decay into a bound state and the related measurement of the neutrino mass. We have given the physical bases for solving these problems and have sketched several possible types of methodological solution.

In discussing methodological questions we have intentionally omitted literature references to particular planned or existing experimental setups, since this will be done in detail in a subsequent review being prepared for publication.

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