Generalized Atomic Mass Law*

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Least-squares analyses have been performed on a set of atomic masses using standard and generalized semiempirical mass laws. Presumably because of errors in the assumed form of the standard mass law, its least-squares coefficients can be determined at best to an accuracy of about 10%, and masses are predicted with an uncertainty of several Mev/c^2 . The standard mass law has been generalized by addition of shell effect and deformation terms. While the least-squares fitting of the generalized mass law is better than for the standard mass law, it is still not possible to predict atomic masses to an accuracy better than a few Mev/ c^2 . The nuclear deformations and the well depth of the nuclear interaction obtained from the additional mass-law terms are in reasonable agreement with more accurate determinations by other methods. A similar statement applies to the nuclear radius constant as obtained from the least-squares coefhcient of the Coulomb energy term. A study has also been made of the effects of additional terms propertional to the absolute value of the isotopic spin, exchange and surface corrections to the Coulomb energy, and the surface correction to the normal isotopic term.

[~] 'HE Weizsacker semiempirical mass law'' was developed in an attempt to correlate observed atomic masses with theoretical estimates obtained by computing the average potential and kinetic energies of the A nucleons in a nucleus, taking into account the difference between the Z protons and N neutrons. This computation resulted in the following mass law (hereafter referred to as the standard mass law).

$$
M(Z,A) = ZM_H + NM_n - \alpha A
$$

$$
+\beta\frac{I^2}{A}+\gamma A^{\frac{2}{3}}+\epsilon\frac{Z(Z-1)}{A^{\frac{1}{3}}}+\frac{1}{2}\delta_A, (1)
$$

where $M(Z,A)$ = atomic mass of the atom containing Z protons, $N = A - Z$ neutrons, and Z electrons; M_H =atomic mass of hydrogen; M_n =neutron mass; $I=N-Z$; and

$$
\delta_A = \begin{cases} 0 \text{ for } A \text{ odd} \\ \text{positive for } A \text{ even, } Z \text{ and } N \text{ odd} \\ \text{negative for } A \text{ even, } Z \text{ and } N \text{ even} \end{cases}
$$

The coefficients α , β , γ , δ _A, and ϵ are theoretically related to independently observable physical quantities. For a uniformly charged sphere of radius $R=R_0A^{\frac{1}{3}}$, the Coulomb energy term multiplying factor is

$$
\epsilon = \frac{3}{5} \frac{e^2}{R_0} = \frac{0.928}{R_0 \text{(fermis)}} \text{mMU} = \frac{0.864}{R_0 \text{(fermis)}} \text{Mev.} \quad (2)
$$

Thus, a least-squares analysis of the standard mass law yields a value of the nuclear-radius constant as well as estimates of masses of unknown nuclei. Many

I. INTRODUCTION recent analyses along these lines are reported in the literature.³ The present paper will discuss one such investigation with the standard law and with generalized forms of the Weizsacker semiempirical mass law.

II. ANALYSIS OF THE STANDARD SEMI-EMPIRICAL MASS LAW

Least-squares analyses of the standard semiempirical mass law have been performed on an electronic computer using the experimental odd- A masses tabulated by Wapstra and Huizenga.⁴ The results are listed in Table I. Figure ¹ compares the absolute value of $\Delta M = \lceil M(\text{calculated}) - M(\text{experimental}) \rceil$ with the experimental uncertainties of the input masses for the standard mass-law analysis listed as the third case of Table I. Figure 2 gives a plot of this ΔM versus mass number for the stable isotopes. The other curves of Fig. 1 and Fig. 2 will be discussed below. From the local smoothness of the curve of Fig. 2 as well as the small uncertainty in experimental masses relative to least-squares computed masses in Fig. 1, it may be concluded that the largest source of uncertainty in such mass law calculations arises from fundamental errors in the assumed form of the mass law. This conclusion is emphasized by the variation in Table I of the least-squares determined coefficients as a function of input data and its weighting. While the 5 to 20% standard deviations from the mean of the least-squares coefficients listed in this table are at best a coarse measure of uncertainties arising from an incomplete or incorrect mass law, it seems reasonable that the least-squares coefficients and, hence, the nuclear radius constant cannot be determined to better than about 10% from analyses of experimental masses by the standard semiempirical mass law. It follows from this conclusion that the standard mass law cannot be

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' C. F. Von Weizsäcker, Z. Physik 96, 431 (1935).
' H. A. Bethe and R. F. Bacher, Revs. Modern Phys. 8, 16.

^{(1936).}

³ A. E. S. Green, Revs. Modern Phys. **30**, 569 (1958).

⁴ A. H. Wapstra, Physica **21**, 367 (1955), and J. R. Huizenga

Physica **21**, 410 (1955).

Input data	Least-squares analysis on	α (mMU) (Mev)	(mMU) (Mev)	(mMU) (Mev)	(mMU) (Mev)	R_0 $(10^{-13}$ cm)
300 odd-A nuclei with $Z>28$	$f = (M - A)/A$	16.839 15.679	24.667 22.968	19.259 17.933	0.75730 0.70515	1.225
93 stable odd-A nuclei with $Z > 28$	$f = (M - A)/A$	16.809 15.652	24.936 23.219	19.236 17.911	0.75005 0.69840	1.237
300 odd-A nuclei with $Z>28$	M	17.798 16.572	27.266 25.388	22.253 20.721	0.82146 0.76489	1.130
162 odd-A nuclei with $N>82$	M	15.173 14.128	22.738 21.172	12.448 11.591	0.67957 0.63277	1.365

TABLE I. Analyses of the standard semiempirical mass law.

applied to the prediction of unknown masses to an energy, accuracy better than several Mev/c'.

III. INCLUSION OF SHELL EFFECTS IN THE MASS LAW

In Fig. 2 it is seen that the largest apparently uncorrected effect contributing to an uncertainty in the assumed form of the standard mass law is that due to nuclear shell structure. Shell effects contribute a relatively greater stability to the nuclear system both at closed shells, due to the shell closing, and between closed shells, due to nuclear deformation. Two terms have been added to the standard mass law in an attempt to account for these effects and thereby deduce more accurate least-squares coefficients and mass values. The resultant mass law will be referred to as the generalized mass law. Its form is as follows

$$
M(\text{general}) = M(\text{standard}) + S(Z,A) + D(Z,A), \quad (3)
$$

where $M(\text{standard})$ is given by Eq. (1), $S(Z,A)$ is the shell function term, and $D(Z,A)$ is the deformation energy term.

The deformation energy term will be obtained by computing the maximum energy gain with quadrupole deformation from a spherical shape. This maximum results from the competition between the first order decrease in kinetic energy with deformation, and the second and higher order changes in the surface and Coulomb energies.

The shell-function term will be computed from the sum over all nucleons of the difference of the nucleon separation energies S_s , computed on a shell model, and S_f , computed in terms of the maximum statistical Fermi kinetic energy, $T_{f \text{max}}$. Thus, the shell-function term is

$$
S(Z,A) = \sum_{A} (S_f - S_s), \tag{4}
$$

where the sum is over all of the nucleons, A in number, in the nucleus.

Weisskopf⁵ has shown that, neglecting surface and isotopic effects, the equality of the average binding

$$
P = \frac{1}{2}V - T,\tag{5}
$$

and the separation energy,

$$
S = V_{\text{max}} - T_{\text{max}},\tag{6}
$$

implies a velocity dependence of the nucleon interaction potential V. The quantity V_{max} is the interaction potential of the nucleon having the maximum kinetic energy T_{max} . Assuming the same velocity dependence in the Fermi and shell models, the shell-function term becomes

$$
S(Z,A) = A(\bar{T}_s - \bar{T}_f). \tag{7}
$$

The average Fermi kinetic energy \bar{T}_f is related to the maximum Fermi kinetic energy $T_{f~\text{max}}$ by

$$
\bar{T}_f = \frac{3}{5} T_f \text{ max}.
$$
 (8)

FIG. 1. Least-squares deviations of masses. (A) Experimenta uncertainties in measured masses. (B) $|\Delta M| = |M|$ (calculated) $-M$ (experimental) | for analysis using the standard mass law (C) $[\Delta M]$ for analysis using the generalized mass law with square-well nuclear interaction potential.

^s V. F. Weisskopf, Nuclear Phys. 3, 423 (1957).

where

FIG. 2. Deviations between calculated and experimental masses for the stable nuclei with odd atomic mass. Dashed curve: masses calculated by least-squares methods, using the standard mass law. Solid curve: masses calculated by least-squares methods using the generalized mass law with square-well nuclear interaction potential.

The shell-model kinetic energy for a nucleon in the ith level is given by

$$
T_s = C(\nu_i l_i) A^{-n/3} = C(\nu_i l_i) (R/R_0)^{-n}, \tag{9}
$$

where $n=1$ for a harmonic oscillator potential and $n\approx 2$ for a deep square well. The symbols v_i and l_i represent the quantum numbers associated with the ith level. The quantity $C(v_i l_i)$ is model dependent. It will be evaluated in an approximate manner below.

Combining Eq. (7) , (8) , and (9) yields

$$
S(Z,A) = -\frac{3}{5}AT_{f\text{ max}} + A^{-n/3}\sum_{A} C(\nu_{i}l_{i}).
$$
 (10)

To evaluate $C(v_i l_i)$ approximately, it is assumed that at some fraction through the kth shell at mass number A_k , the Fermi and shell-model separation energies of the last nucleon are equal. Thus

$$
T_{f \max} = C(\nu_j l_j) A_k^{-n/3},\tag{11}
$$

where the subscript j denotes the highest level containcontaining a particle. Substituting the volume coefficient α for the separation energy S in Eq. (6) and combining Eqs. (6) , (10) , and (11) gives

$$
S(Z,A) = (V_{\text{max}} - \alpha) \{-\frac{3}{5}A + \sum_A (A_k/A)^{n/3} \}. (12)
$$

The mass number A_k , at which the Fermi and shellmodel separation energies of the last nucleon are equal, is assumed to be given by the stable nucleus exactly

TABLE II. A_k values for the *mth* nucleon.

	A_k (protons)	A_k (neutrons)	
∠ m			
\lt{m}			
$20 \le m \le 28$	53		
$28 \le m \le$	89	70	
$50 \le m \le 82$ ٠	161	l 16	
82 < m < 126	269	176	
$126 \le m \le 184$.	255	

intermediate between two closed shells with respect to the nucleon type of interest. For example, for the proton shell with $50\leq Z \leq 82$, the stable nucleus with $Z=66$ is $_{66}Dy^{161}$, hence $A_k=161$. In Table II, the A_k values corresponding to the above definition are listed.

In mass law analyses involving the shell-function term $S(Z,A)$, the factor $(V_{\text{max}}-\alpha)$ has been left as an arbitrary coefficient determined by least-squares analysis. Values of this coefficient will be discussed following the derivation of the nuclear deformation energy term that gives rise to increased stability and decreased atomic mass between closed shells. A detailed plot of the shell-function term for a particular least-squares analysis is given in Fig. 3.

In terms of the deformation parameter λ defined from

$$
r = R[1 + \lambda P_2(\cos \theta)], \qquad (13)
$$

$$
P_2(\cos\theta) = \frac{1}{2}(3\cos^2\theta - 1),
$$

Frc. 3. Deformation and shell-function terms for the stable odd-A nuclei using a square-well nuclear interaction potential.

and r is the radius vector to the point (r, θ, ϕ) on the surface of a nucleus, the nuclear deformation' energy change ΔE may be written

$$
\Delta E = -C_1 \lambda + C_2 \lambda^2 - C_3 \lambda^3 + 0(\lambda^4),\tag{14}
$$

where the coefficients C_i will be defined and discussed below. The extremum of ΔE gives the deformation energy term $D(Z,A)$ and equilibrium deformation λ_m as

$$
D(Z,A) = -C_2 \lambda_m^2 \xi \left\{ 1 - \frac{2C_3}{C_2} \lambda_m \right\},
$$
(15)

$$
\sum_{n=1}^{\infty} \frac{(C_2^2 - 3C_1 C_3)^{\frac{1}{2}}}{(16)}
$$
(16)

$$
\lambda_m = \frac{C_2}{3C_3} - \frac{(C_2^2 - 3C_1C_3)^{\frac{1}{2}}}{3C_3},\tag{16}
$$

where ξ is an arbitrarily inserted, least-squares deter mined coefficient, whose value should be near one. The preceding equations assume both that the deformation energy change is determined solely by quadrupole-type deformations and that this energy change is adequately described by neglecting higher than cubic terms in the expansion of Eq. (14). Since 'both of these assumptions are probably wrong, $6,7$ the coefficients C_1 , C_2 , and C_3 will have to be arbitrarily adjusted to give equilibrium deformations in agreement with nuclear quadrupole moments and Coulomb excitation cross sections.

The coefficients C_2 and C_3 are assumed to arise from Coulomb and surface energy changes of a uniformly charged spheroid relative to a uniformly charged sphere. Hence, '

$$
C_2 = \frac{2}{5}\gamma' A^{\frac{2}{3}}(1-x),\tag{17}
$$

$$
C_3 = (2/21)FC_2,\tag{18}
$$

$$
x = \frac{\epsilon'}{2\gamma'}Z(Z-1)/A,\tag{19}
$$

$$
F = (1+2x)/(1-x),
$$
 (20)

where ϵ' and γ' would be the coefficients of the Coulomb and surface energy terms in the semiempirical mass law, except that they are adjusted somewhat to compensate for the poor assumptions discussed previously.

The coefficient C_1 results from a determination of ΔT_m , the change in average kinetic energy of the mth nucleon in the i th level due to deformation.⁹ From Eq. (9)

$$
T_m + \Delta T_m = C(\nu_i l_i) \left(\frac{R + \Delta R_m}{R_0}\right)^{-n}
$$

$$
\approx T_m \left(1 - n \frac{\Delta R_m}{R}\right), \quad (21)
$$

where T_m is the kinetic energy in a spherical potential and ΔR_m is the average radial change in the position of the *m*th nucleon due to deformation.

Assuming no change in the nucleon potential energy with deformation, the energy gain due to the single particle level shift following deformation is, from Eq. (21),

Eq. (21),
\n
$$
\Delta E_1 = -\sum_A nT_m \frac{\Delta R_m}{R}
$$
\n
$$
= -n(V_{\text{max}} - \alpha) \sum_A \frac{\Delta R_m}{R} \left(\frac{A_k}{A}\right)^{n/3}, \quad (22)
$$

where use has been made of Eqs. (6) , (9) , and (11) . From Eq. (13) it is seen that $\Delta R_m/R$ is proportional to λ . Thus

$$
\Delta R_m/R = f_m \lambda, \qquad (23)
$$

FIG. 4. Deformation function parameter f_{mp} between two magic numbers.

where f_m is the deformation parameter for the mth nucleon in a shell. It is discussed below. Hence

$$
C_1 = n(V_{\text{max}} - \alpha) \sum_A f_m(A_k/A)^{n/3}.
$$
 (24)

The coefficient f_m is the sum of the contributions f_{mp} and f_{mn} , which arise from the deformed orbits of protons and neutrons, respectively. From the experimental fact that magic nuclei are spherical

$$
\sum \text{closed shell} f_{mp} = \sum \text{closed shell} f_{mn} = 0. \tag{25}
$$

The plot of f_{mp} or f_{mn} versus the number of protons or neutrons in a shell should be independent of the shell under consideration. This requirement is inconsistent with Eq. (16) and the facts that nuclear deformations between closed shells are generally small for $Z<50$ and large for $Z > 50$. To circumvent this dilemma, it is assumed that $f_{mp} = f_{mn} = 0$ for $N < 82$. For $N > 82$, the assumed form of the sum of f_{mp} over the protons in the sth shell is given in Fig. 4, in which Z_{CS1} and Z_{CS2} are the atomic numbers at the beginning and end of the sth proton shell. For consistency between computed λ_m values and observed quadrupole moments, the coefficients a and b of Fig. 4, should be about 0.3 and 0.7, respectively. The quality of least-squares analyses was relatively insensitive to $\pm 20\%$ changes in a and b. The maximum ordinate of the curve of Fig. 4 was determined from the requirement that λ_m ($_{67}$ Ho¹⁶⁵) = 0.26. The above remarks apply equally to the deformation due to neutrons and to the quantity f_{mn} . A detailed plot of the deformation energy term for a particular least-squares analysis is given in Fig. 3.

IV. RESULTS OF ANALYSES OF THE GENERALIZED MASS LAW

The results of two typical least-squares analyses of the generalized mass law on the masses of 300 odd-A nuclei are given in Table III. In these analyses, it was assumed that $a=0.3$, $b=0.7$, $\epsilon'=0.775$ mMU, and $\gamma' = 25$ mMU. A comparison of calculated and empirical¹⁰ deformation parameters is made in Fig. 5.

⁶ S. K. Moszkowski (private communication).
⁷ K. Kumar and M. A. Preston, Phys. Rev. **107**, 1099 (1957).
⁸ W. J. Swiatecki, Phys. Rev. **104**, 993 (1956). ⁹ James Rainwater, Phys. Rev. **79**, 432 (1950).

¹⁰ Alder, Bohr, Huus, Mottelson, and Winther, Revs. Modern Phys. 18, 432 (1956); see Table V3 on p. 531.

Well type					$V_{\rm max} - \alpha$ $\begin{array}{cc} \alpha & \beta \\ (mMU) & (mMU) & (mMU) & (mMU) & (mMU) \end{array}$ (Mev) (Mev) (Mev) (Mev) (Mev) (Mev) $(fermis)$ ε	R_0	
Square $(n=2)$ Harmonic	16.412 15.282	24.438 22.755	16.922 15.757	0.74120 0.69016	9.1483 8.5183	1.252	1.2226
oscillator $(n=1)$	19.310 17.980	24.592 22.899	17.144 15.963	0.74545 0.69412	18.752 17.461	1.245	1.2930

TABLE III. Analyses of the generalized mass law $(1 \text{ fermi} = 10^{-13} \text{ cm}).$

Figures 1 and 2 illustrate the improved agreement between the general semiempirical mass law and experimental masses, as compared to the standard mass law analysis illustrated in the same figures. While this agreement is better for the generalized mass law, it is not sufficient for the quantitative determination of nuclear masses or mass law parameters. Because of the local smoothness of the curve of Fig. 2, as well as the small uncertainty in experimental masses relative to least-squares computed masses of Fig. 1, it may still be concluded that the largest source of uncertainty in the general mass law calculations arises from fundamental errors in the assumed form of the mass law. It is probable that no simple fundamental law will be applicable to a wide range of Z and \overline{A} for quantitatively predicting nuclear masses.

Nevertheless, it is still interesting to inquire into the qualitative validity of the generalized mass law in terms of the least-squares determined coefficients and the calculated deformations. From Fig. 5, it is seen that the computed deformations agree with measured values to within experimental errors. From Tables I and III, it is seen that the magnitudes of the least-squares coefficients are relatively unchanged by addition of the closed-shell and deformation terms. The fact that $\xi \approx 1.25$, indicates that deformation energy changes are in general agreement with the deformations that produce them. The well depth is 23.8 Mev for a square well, and 35.4 Mev for a harmonic oscillator, as determined by the least-squares coefficient of the shellfunction term. This result is in general agreement with other determinations and with the fact that one might expect the maximum harmonic oscillator well depth to be greater than that for a square well, since something like the average, not the maximum, well depths must be equal in the two models.

In summary, the generalization of the standard semiempirical mass law by the inclusion of terms that account for shell closings and nuclear deformation, results in an improved agreement between experimental and least-squares determined masses. The least-squares coefficients are qualitatively reasonable and yield a nuclear radius constant in the neighborhood of 1.25 Fermis, a well depth between 20 and 40 Mev, and qualitatively accurate nuclear deformations. However, neither the standard nor the generalized mass law is sufficiently accurate to allow for quantitative estimation Of nuclear masses to within a few Mev, or quantitative

analyses of the least-squares coefficients to better than about 10% .

An attempt was also made to calculate the deformation energy term on an electronic computer using Nilsson's model.¹¹ This attempt was discontinued because the calculated deformations failed to agree with experiment (i.e. , magic nuclei were badly deformed) unless large changes were made in Nilsson's parameters.

IV. INCLUSION OF ADDITIONAL SURFACE AND EXCHANGE EFFECTS IN THE STANDARD MASS LAW

The results of the previous sections indicate that the probability of a simply generalized mass law yielding quantitative estimates of nuclear masses is small. However, it is still interesting to determine the qualitative effects on the least-squares coefficients of the addition of further surface and exchange terms in the mass law. An isotopic exchange term¹² proportional to $|I|/A$ as well as isotopic surface correction terms proportional to $|I|/A^*$ and I^2/A^* have been studied. The least-squares coefficients of the terms involving the absolute value of the isotopic spin fluctuated widely in sign and magnitude with variations of input data and its weighting. Hence, further discussion of these terms will be omitted. The effect of an isotopic surface correction term, $-\eta I^2/A^{\frac{4}{3}}$, in the standard mass law of Eq. (1) will be discussed below.

The surface and exchange effects on the Coulomb energy modify the Coulomb term of Eq. (1) into the

FIG. 5. Comparison of calculated deformations (λ) with experimental measurements. \bullet Calculated for a square-well nuclear interaction potential. X from Coulomb excitation measurements, ' $+$ from hyperfine structure measurements.¹⁰

¹¹ B.R. Mottelson and S. G. Nilsson, Phys. Rev. 99, 1615 (1955). ¹² J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physic*
(John Wiley & Sons, Inc., New York, 1952), pp. 223 ff,

following form:

$$
E_C = \epsilon \left\{ \frac{Z^2}{A^{\frac{1}{4}}} - 0.764 \frac{Z^{\frac{1}{4}}}{A^{\frac{1}{4}}} - \frac{7}{24} \left(\frac{t}{R_0} \right)^2 \frac{Z^2}{A} + \cdots \right\}
$$
 (26)
= $\epsilon \frac{Z^2}{A^{\frac{1}{4}}} - 0.764 \epsilon_1 \frac{Z^{\frac{1}{4}}}{A^{\frac{1}{4}}} - 3.05 \epsilon_1 \frac{Z^2}{A} + \cdots$,

where the surface thickness t is chosen as 3×10^{-13} centimeters and R_0 in the surface correction term is written in terms of ϵ_1 using Eq. (2). The quantity ϵ_1 is an estimate of the Coulomb coefficient ϵ obtained by an iterative procedure. The results of several analyses of the standard mass law generalized by the additional Coulomb terms and the isotopic surface correction term are listed in Table IV.

The deviations between experimental and leastsquares masses were essentially independent of any of the additions to the standard mass law listed in Table IV. This fact, as well as the near constancy of the conventional coefficients and larger variation of the isotopic surface term coefficient η , indicates that masses are not particularly sensitive to the magnitude of η . The ratio η/γ can be determined¹³ from the atomic

weight of that isotope of a heavy element which has the maximum spontaneous fission lifetime. One finds $\eta/\gamma = (1-4Z^2/A^2)^{-1}$, where Z and A apply to the isotope with the maximum lifetime. Examples among the heavy elements are $_{90}Th^{232}$, $_{92}U^{236}$, $_{94}Pu^{240}$, and $_{96}Cm^{246}$. From these cases, $\eta/\gamma \approx 2.6$. It is particularly satisfying that this is just the value obtained when η is determined by least-squares analysis on the packing fraction f for the case with $\epsilon_1=0$ in which uncertainties arising from the additional parameter in the Coulomb term are avoided. In the other cases tested, η/γ reaches a value as high as 4.4, indicating, in general, that this ratio can be determined only to order of magnitude by analysis of observed masses.

An arbitrary "best" choice of the solutions in Table IV is $\epsilon_1 = 0.724$, $\alpha = 15.297$, $\beta = 29.188$, $\gamma = 18.342$, η =46.354, ϵ =0.69614, all in Mev. Note that R_0 =1.24 fermis for this case. Again, it must be emphasized that these parameters in the Weiszacker mass law give atomic masses only to an accuracy of several ${\rm MeV}/c^2$.

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¹³ W. A. Fowler (private communication). J. A. Wheeler (private communication to W. A. Fowler).