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Atomic Mass Formula with Constant Shell Terms

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An atomic mass formula is constructed in the form of a sum of gross terms and empirical constant shell terms. The shell parameter values are determined by a new statistical method, whose essential point is to take account of the intrinsic error of the mass formula. This method is explained in some detail, and the properties of the resulting formula are discussed.

§ 1. Introduction

We construct an atomic mass formula paying special attention to its error. This study was motivated by the following two facts. First, the experimental masses, to which the mass formula is to be fitted by adjusting free parameters, have a great variety of errors ranging from keV to MeV. How much weight should we give to these mass data? Second, the various existing mass formulas[1),2)] do not agree well with each other when compared in the nuclidic region far from stability. How should we take this disagreement in the absence of errors[*)] attached to these formulas?

The mass formula in this paper has a form similar to that of Ref. 2), and consists of a gross part and empirical proton and neutron shell parts. In determining the values of the parameters in the shell parts, however, we apply a new statistical method. The essential point of this method is to take account of the error inherent in the mass formula. Existence of such an error is inferred from the following consideration: No matter how carefully we may adjust the parameter values and no matter how accurately the experimental masses may be measured, there will still remain some discrepancy between the mass formula and the experimental masses. We refer to this error as intrinsic errors.

The statistical weight to be used in our method turns out to be the inverse square of the error composed of the experimental and the intrinsic errors. Thus, this method includes both the ordinary weighted least-squares method and the equally-weighted least-squares method as special cases; the former corresponds to zero intrinsic errors and the latter to infinite intrinsic errors. In this paper, the intrinsic errors are determined self-consistently. The new method not only enables us to deal justly with the experimental data with a variety of errors, but also

*) There is one exception; the mass values by Comay and Kelson[3)] have errors.

gives information on the errors of mass predictions.

We describe the form of the mass formula in § 2, explain the new statistical method in § 3, give the details of the analysis in § 4, and show the results and discuss some properties of the formula in the last section.

§ 2. Mass formula

We assume the mass excess of the nuclide with Z protons and N neutrons in the following form:

$$M_E(Z,N)=M_{Eg}(Z,N)+P_Z(N)+Q_N(Z)-\Delta M_{\text{odd-odd}}(A). \qquad (1)$$

Here, $M_{Eg}(Z,N)$ is a smooth function of Z and N representing the gross feature of the nuclear mass surface. The terms $P_Z(N)$ and $Q_N(Z)$ are the proton and neutron shell terms respectively, each of which is not necessarily smooth with respect to the subscript Z or N, but is a smooth function of the variable N or Z in the parentheses. The last term is a small correction for odd-odd nuclei.

The gross part $M_{Eg}(Z,N)$ is expressed in MeV as

$$\begin{aligned}M_{Eg}(Z,N)=&7.68023A+0.39120I+a(A)\cdot A+b(A)\cdot|I|\\&+c(A)\cdot I^2/A+E_c(Z,N)-14.33\times 10^{-6}Z^{2.39}\end{aligned} \qquad (2)$$

with $A=N+Z$, $I=N-Z$,

$$a(A)=a_1+a_2A^{-1/3}+a_3A^{-2/3}+a_4A^{-1}\,,$$

$$b(A)=bA^{-2/3}\,,$$

$$c(A)=c_1+c_2A^{-1/3}+c_3A^{-2/3}/(1+c_4A^{-1/3}).$$

The Coulomb energy $E_c(Z,N)$ is taken as that of the trapezoidal charge distribution:

$$\begin{aligned}E_c(Z,N)=&\frac{0.864}{r_0}\left(\frac{R}{r_0}\right)^5\left[1+\frac{5}{6}\left(\frac{z}{R}\right)^2+\frac{1}{2}\left(\frac{z}{R}\right)^3+\frac{1}{6}\left(\frac{z}{R}\right)^4\right.\\&\left.-\frac{1}{42}\left(\frac{z}{R}\right)^5\right]\cdot\left(\frac{Z}{A}\right)^2-\frac{0.66}{r_0}\left(\frac{Z}{A}\right)^{4/3}\cdot A\end{aligned}$$

with

$$R=r_0\left\{\left[\sqrt{\left(\frac{A}{2}\right)^2+\frac{1}{27}\left(\frac{z}{r_0}\right)^6}+\frac{A}{2}\right]^{1/3}-\left[\sqrt{\left(\frac{A}{2}\right)^2+\frac{1}{27}\left(\frac{z}{r_0}\right)^6}-\frac{A}{2}\right]^{1/3}\right\}.$$

In the following numerical calculations, we use 1.13 fm for the radius parameter r_0 and 1.5 fm for the half surface thickness z.[2] The last term in Eq. (2) is the binding energy of atomic electrons. The correction term for odd-odd nuclei is, on the average, expressed as[2)]

$$\Delta M_{\text{odd-odd}}(A)=\frac{11719.21}{(A+31.4113)^2}-\frac{1321495}{(A+48.1170)^3}\,. \tag{3}$$

We take two functional forms for the shell terms $P_Z(N)$ and $Q_N(Z)$: The first is the constant form

$$P_Z(N)=P_Z\,,\qquad Q_N(Z)=Q_N\,, \tag{4}$$

and the second is the linear form

$$P_Z(N)=P_Z{}^0+(N-N_{0Z})P_Z{}^1\,,\quad Q_N(Z)=Q_N{}^0+(Z-Z_{0N})Q_N{}^1\,, \tag{5}$$

where P_Z, Q_N, $P_Z{}^0$, $P_Z{}^1$, $Q_N{}^0$, $Q_N{}^1$ are adjustable parameters, and Z_{0N} and N_{0Z} are constants representing the β-stability line. In this paper we present the results for the constant shell form. As for the linear shell form, some of the results were already reported at the Sixth International Conference on Atomic Masses;[4)] more detailed explanation will be given elsewhere.

The values of the gross-part parameters, a_i, b, c_i, are obtained through a procedure similar to that described in Ref. 2) as

$$\begin{aligned} a(A)&=-17.080+30.138A^{-1/3}-31.322A^{-2/3}+24.192A^{-1}\,,\\ b(A)&=15.0A^{-2/3}\,,\\ c(A)&=35.217-89.811A^{-1/3}+92.940A^{-2/3}/(1+0.56912A^{-1/3}). \end{aligned} \tag{6}$$

The values of the shell parameters, P_Z and Q_N, whose number amounts to about 250, are determined by a new statistical method explained in the next section.

In determining the values of the shell parameters, we use only even-even and odd-A nuclei excluding odd-odd ones in consideration of the somewhat more irregular nature of the odd-odd mass surface. The mass excesses of even-even and odd-A nuclei are, in the case of the constant shell form, simply written as

$$M_E(Z,N)=M_{Eg}(Z,N)+P_Z+Q_N\,. \tag{7}$$

§ 3. A new statistical method

Our next step is to determine the values of the shell parameters P_Z and Q_N, the sum of which should reproduce the difference between the experimental mass and the gross-part value, $M_{E\text{exp}}(Z,N)-M_{Eg}(Z,N)$, as accurately as possible. Here, we encounter a problem. The errors of the input mass data range from keV to MeV, and if we apply the ordinary weighted least-squares method, in which inverse squares of experimental errors are used as the statistical weights, our formula will be governed by a rather small number of input data with very small errors. This seems to be unfair to the input data with larger errors because our formula will have errors of 100 keV or more. Confronting such a problem a certain author added a constant quadratically to all the experimental errors and another treated all the data with equal weights. In this paper, however, we overcome this difficulty by devising a new statistical method.

In order to divide the "experimental shell energies", $M_{E\mathrm{exp}}(Z, N)-M_{Eg}(Z, N)$, into the proton and neutron shell terms as P_Z+Q_N, we use an iteration method. We first assume some initial values for Q_N's, and compute

$$y_Z(N)=M_{E\mathrm{exp}}(Z, N)-M_{Eg}(Z, N)-Q_N\ , \tag{8a}$$

which we call "experimental" proton shell energy. Then, we adjust the values of P_Z's so as to fit to the "experimental" proton shell energies. We call this procedure step (I). Using these values of P_Z's, we compute

$$M_{E\mathrm{exp}}(Z, N)-M_{Eg}(Z, N)-P_Z\ , \tag{8b}$$

which we call "experimental" neutron shell energy. Then, we adjust the values of Q_N's so as to fit to the "experimental" neutron shell energies. We call this procedure step (II). The values of Q_N's thus obtained are generally different from those used in the previous step (I). We repeat these steps (I) and (II) until the parameter values converge.

The essential point of our new statistical method lies in each of the steps (I) and (II). We explain it for the case of step (I).

As mentioned in § 1, we introduce intrinsic errors in our formula. More specifically, we attach intrinsic errors to the shell terms P_Z and Q_N, and denote them by α_Z and β_N respectively in the sense of standard deviation. In general, these intrinsic errors are different if Z's or N's are different. No intrinsic error is attached to the gross part in this paper. The meaning of the intrinsic error will become clearer if we plot $y_Z(N)$ as a function of N. Usually, one tries to draw a line through these plots, but we try to draw a stripe with an approximate width of α_Z. In our special case in which P_Z does not depend on N and Q_N does not depend on Z, the stripe is horizontal. In addition to the intrinsic errors we take account of the errors coming from the uncertainties in the determination of the values of the shell parameters P_Z and Q_N. We call them extrinsic errors, and denote them by $\Delta P_Z^{\mathrm{ext}}$ and $\Delta Q_N^{\mathrm{ext}}$ for the proton and neutron respectively. Then, the error of the "experimental" proton shell energy $y_Z(N)$ is written as

$$\eta_Z(N)=[\varepsilon_Z(N)^2+\beta_N{}^2+(\Delta Q_N{}^{\mathrm{ext}})^2]^{1/2}\ , \tag{9}$$

where $\varepsilon_Z(N)$ stands for the error of $M_{E\mathrm{exp}}(Z, N)$. Thus, our problem is reduced to the determination of P_{Z0} (the best value of P_Z), $\Delta P_Z^{\mathrm{ext}}$, and α_Z by comparison with $y_Z(N)$ and $\eta_Z(N)$.

In order to proceed further, we consider the true value of the quantity that corresponds to $y_Z(N)$, and denote it by $Y_Z(N)$. This true value is known only probabilistically, and this probability is governed by the two informations that we have. The first information is that the true values $Y_Z(N)$ in which Z is fixed and N is varied are distributed around P_Z with the standard deviation α_Z. This is expressed in the form of error function as

$$\Phi_{\text{th}}(Y_Z(N); P_Z, \alpha_Z) = \frac{1}{\sqrt{2\pi}\,\alpha_Z} \exp\{-[Y_Z(N)-P_Z]^2/2\alpha_Z^2\}. \tag{10}$$

The second information is that the experimental value $y_Z(N)$ tends to be distributed around $Y_Z(N)$, or equivalently $Y_Z(N)$ tends to be distributed around $y_Z(N)$, with the standard deviation $\eta_Z(N)$. This is expressed as

$$\Phi_{\text{exp}}(Y_Z(N); y_Z(N), \eta_Z(N)) = \frac{1}{\sqrt{2\pi}\,\eta_Z(N)} \exp\{-[Y_Z(N)-y_Z(N)]^2/2\eta_Z(N)^2\}. \tag{11}$$

Then, the probability of finding the true value in $Y_Z(N) \sim Y_Z(N)+dY_Z(N)$ as deduced from these two informations is given by the product $\Phi_{\text{th}} \cdot \Phi_{\text{exp}} dY_Z(N)$. Accordingly, we can write the probability with which the set of experimental data $y_Z(N)$'s is obtained as

$$\Psi = \Psi(y_Z(N)\text{'s}; \eta_Z(N)\text{'s}, P_Z, \alpha_Z) = \prod_N \left[\int_{-\infty}^{\infty} \Phi_{\text{th}}(Y_Z(N); P_Z, \alpha_Z)\Phi_{\text{exp}}(Y_Z(N); y_Z(N), \eta_Z(N))\,dY_Z(N)\right], \tag{12}$$

where the product is taken over all isotopes with the fixed Z. A straightforward calculation leads to

$$\Psi = \left[\prod_N \frac{1}{[2\pi(\eta_Z(N)^2+\alpha_Z^2)]^{1/2}}\right] \cdot \exp\left[-\sum_N \frac{(y_Z(N)-P_Z)^2}{2(\eta_Z(N)^2+\alpha_Z^2)}\right]. \tag{13}$$

The most probable (best) value of P_Z is given as that maximizing the probability Ψ, and is explicitly written as

$$P_{Z0} = \left[\sum_N \frac{y_Z(N)}{\eta_Z(N)^2+\alpha_Z^2}\right] \cdot \left[\sum_N \frac{1}{\eta_Z(N)^2+\alpha_Z^2}\right]^{-1}. \tag{14}$$

The extrinsic error ΔP_Z^{ext} is obtained from Ψ by regarding it as a distribution function of P_Z. Equation (13) is rewritten as

$$\Psi = \left[\prod_N \frac{1}{[2\pi(\eta_Z(N)^2+\alpha_Z^2)]^{1/2}}\right] \times \exp\left[-\sum_N \frac{1}{2(\eta_Z(N)^2+\alpha_Z^2)}(P_Z-P_{Z0})^2 - \sum_N \frac{(y_Z(N)-P_{Z0})^2}{2(\eta_Z(N)^2+\alpha_Z^2)}\right], \tag{15}$$

and we get

$$\Delta P_Z^{\text{ext}} = \left[\sum_N \frac{1}{\eta_Z(N)^2 + \alpha_Z^2} \right]^{-1/2} . \tag{16}$$

As for the intrinsic error α_Z, one might also try to determine it by maximizing Ψ, but this method does not work in our problem. Therefore, we adopt the following procedure starting from the original meaning of α_Z. First, we calculate the expectation value of $\sum_N (Y_Z(N) - P_Z)^2$ as

$$\langle \sum_N \Delta Y_Z(N)^2 \rangle = \frac{\int_{-\infty}^{\infty} dP_Z \int_{-\infty}^{\infty} \cdots \int_{-\infty}^{\infty} \sum_N (Y_Z(N) - P_Z)^2 H \prod_{N'} dY_Z(N')}{\int_{-\infty}^{\infty} dP_Z \int_{-\infty}^{\infty} \cdots \int_{-\infty}^{\infty} H \prod_{N'} dY_Z(N')} \tag{17}$$

with

$$H = \prod_{N'} [\Phi_{\text{th}}(Y_Z(N'); P_Z, \alpha_Z) \Phi_{\text{exp}}(Y_Z(N'); y_Z(N'), \eta_Z(N'))].$$

Here, we note that the average is taken over P_Z also, because it is determined only probabilistically. It may be suggested that the quantity $\langle \sum_N \Delta Y_Z(N)^2 \rangle$ is equal to α_Z^2 multiplied by the number of the data $n_Z (= \sum_N 1)$:

$$\langle \sum_N \Delta Y_Z(N)^2 \rangle = n_Z \alpha_Z^2 . \tag{18}$$

In this expression we do not subtract the number of the parameters (one in our case of constant shell form) from n_Z though it is often done in statistical analyses like this. We could not find any reasonable foundation of such a prescription, at least for our problem. To take the average over P_Z in the calculation of $\langle \sum_N \Delta Y_Z(N)^2 \rangle$ is a more reasonable prescription.

Equation (18) includes α_Z on both sides, and can be regarded as an equation for α_Z. It always has a solution $\alpha_Z = 0$, but this solution should be abandoned as a trivial one if there is another solution satisfying $\alpha_Z > 0$. Actually there are many Z values for which we cannot find any positive solution, and after all we have obtained a set of solutions α_Z's, some of which are positive and some are zero. It is the simplest to accept this set as it is, but from a physical consideration we somewhat modify it in the present case.

The reason for the modification is as follows. Our mass formula is an approximate one, and it is difficult to think of the true intrinsic error to be zero even for a single Z value; the actual occurrence of zeros is probably due to insufficiency of input data.

Our modification is as follows. Denoting the solution of Eq. (18) by α'_Z, we adopt α_Z which is defined by

$$\alpha_Z^2 = 0.9 \alpha_Z'^2 + 0.1 \bar{\alpha}_Z^2 , \tag{19}$$

where $\bar{\alpha}_Z$ is the root mean square of α'_Z (and consequently of α_Z). Even Z and odd

Z are treated separately. The coefficients on the right-hand side of Eq. (19) have been fixed by comparison with the following simulation. We first produce simulated "experimental" mass data by assuming the probability distribution (13) in which the value of α_Z given by Eq. (19) is used. Then, using these data we calculate α_Z''s by the same procedure as mentioned above, and finally we compare the frequency distribution of these α_Z''s with that obtained by using the real experimental data. The coefficient 0.1 in Eq. (19) is the approximate maximum that produces no remarkable difference between the two frequency distributions.

This procedure is also followed in determining the values of Q_N, $\Delta Q_N^{\mathrm{ext}}$ and β_N for neutron shell terms.

After the iteration converges, we have a mass formula which provides masses together with their theoretical errors expressed as

$$\delta M(Z,N)=\left\{\alpha_Z^2+\beta_N^2+[\Delta P_Z^{\mathrm{ext}}]^2+[\Delta Q_N^{\mathrm{ext}}]^2 + \left[\frac{1}{3}\Delta M_{\mathrm{odd\text{-}odd}}(A)\right]^2\right\}^{1/2}, \tag{20}$$

where the last term in curly brakets should be added for odd-odd nuclei only.

§ 4. Input data and additional condition

We use the mass values and their errors in the mass table of Wapstra and Bos[5] by neglecting the correlation among the errors. Although they are not the original experimental data, we refer to them as experimental values. We also use the systematics mass values, which are denoted by SYST in the table, together with errors estimated in the following manner.

First, we estimate the errors of the nucleon separation energies related to the SYST nuclei in the light of the systematics of Yamada and Matumoto.[6] They are approximately expressed in MeV as

$$\Delta S_p=(C+1.8k_p+0.18k_p^2)A^{-2/3}, \tag{21a}$$

$$\Delta S_n=(C+1.8k_n+0.18k_n^2)A^{-2/3}. \tag{21b}$$

Here, ΔS_p is the error of the proton separation energy (S_p) of the nucleus in question, and k_p is the difference between the neutron numbers of that nucleus and its nearest even-N isotope that has experimental S_p data. Equation (21b) has a similar meaning with proton and neutron interchanged. In this counting of k_p or k_n we put aside the experimental errors of some S_p or S_n data and treat those S_p or S_n as SYST values if their original errors are larger than those estimated as SYST values. Furthermore, when a SYST S_p (or S_n) is located between two experimental S_p (or S_n) data, we compute the error of S_p (or S_n)

from both sides, and add the two errors inverse-quadratically. The constant C in Eq. (21a) is taken to be 6 for even-N S_p and 12 for odd-N S_p, and that in Eq. (21b) is taken to be 6 for even-Z S_n and 12 for odd-Z S_n.

Next, we estimate the error of the SYST mass of the nucleus that lies just next to both the isotope and the isotone having experimental mass data. We compute two (or sometimes three, or four) kinds of errors, each of which is obtained by quadratically adding the appropriate ΔS_p (or ΔS_n) to the error of the experimental mass of the adjacent isotone (or isotope). Then, by adding these errors inverse-quadratically, we get the estimate of the error in the mass of the SYST nucleus. This procedure is repeated to get the estimates of the errors of the SYST nuclei that are farther from the nuclei having experimental mass data. Thus, for example, we assigned the errors of 752 keV to the mass of ^{51}Co and 360 keV to ^{163}Yb as cases with small k_p and k_n, and 1591 keV to ^{162}W as a case with large k_p and k_n.

In order to avoid too slow convergence in our iteration procedure we have imposed an additional condition that the absolute values of P_Z and Q_N should not be too large. Thus, we have actually modified Eqs. (13)~(16) as follows:

$$\Psi=\left[\prod_N \frac{1}{[2\pi(\eta_Z(N)^2+\alpha_Z{}^2)]^{1/2}}\right]\cdot\exp\left[-\sum_N \frac{(y_Z(N)-P_Z)^2}{2(\eta_Z(N)^2+\alpha_Z{}^2)}-\lambda P_Z{}^2\right], \tag{22}$$

$$P_{Z0}=\left[\sum_N \frac{y_Z(N)}{\eta_Z(N)^2+\alpha_Z{}^2}\right]\cdot\left[\sum_N \frac{1}{\eta_Z(N)^2+\alpha_Z{}^2}+2\lambda\right]^{-1}, \tag{23}$$

$$\Psi=\left[\prod_N \frac{1}{[2\pi(\eta_Z(N)^2+\alpha_Z{}^2)]^{1/2}}\right]\exp\left\{-\left[\sum_N \frac{1}{2(\eta_Z(N)^2+\alpha_Z{}^2)}+\lambda\right](P_Z-P_{Z0})^2 -\sum_N \frac{(y_Z(N)-P_{Z0})^2}{2(\eta_Z(N)^2+\alpha_Z{}^2)}-\lambda P_{Z0}{}^2\right\}, \tag{24}$$

$$\Delta P_Z^{\mathrm{ext}}=\left[\sum_N \frac{1}{\eta_Z(N)^2+\alpha_Z{}^2}+2\lambda\right]^{-1/2}. \tag{25}$$

Here, λ is the constant representing the strength of the condition. We have used $\lambda=0.1$ in the present work, which is so small that the increase in the root mean square of α_Z and β_N is only 2%.

§ 5. Results and discussion

Making about 5000 times the iteration explained in § 3, we have reached a satisfactory convergence, and show important features of the final formula in the following.*) The root mean square of α_Z is 319 keV for even Z and is 398 keV for odd Z, and that of β_N is 326 keV for even N and is 341 keV for odd N.

*) The computer code to compute various quantities will be sent upon request.

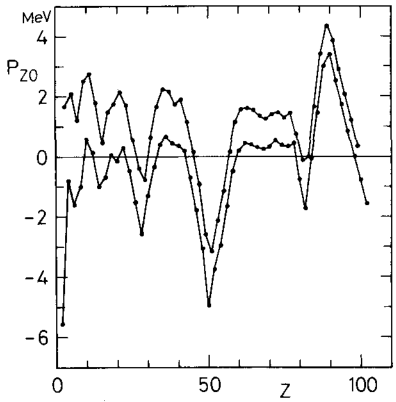

Fig. 1. The most probable (best) value of the proton shell term P_{Z0} plotted against Z.

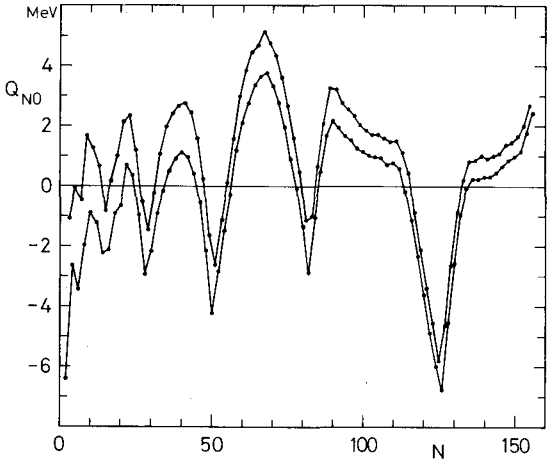

Fig. 2. The most probable (best) value of the neutron shell term Q_{N0} plotted against N.

We give the most probable (best) values of the shell parameters, P_{Z0} and Q_{N0}, in Figs. 1 and 2. In these figures we see marked dips at the j-j magic numbers, 6, 14, 28, 50, 82 and 126, but not at the L-S magic numbers, 8 and 20. The separation of the P_{Z0} plot (and also of the Q_{N0} plot) into two lines is simply due to the even-odd effect; note that our shell terms, P_Z+Q_N, include the usual even-odd term. As regards the charge symmetry of nuclear forces, which should manifest itself as an approximate equality of P_Z and Q_N for $Z=N$ in the region of light nuclei (Z, $N<20$), our new formula is somewhat inferior to the old one reported in Ref. 2). It remains to be seen to what degree we can restore the charge symmetry without increasing the errors of the mass formula appreciably. The tendency that the P_Z rapidly decreases with Z and the Q_N rapidly increases with N in the heaviest region is similar to that of Ref. 2), and is a shortcoming of the constant-shell-term formula.

In Figs. 3 and 4, in which the intrinsic errors α_Z and β_N are shown, we see fairly large fluctuations. Namely, α_Z and β_N become large for some particular values of Z and N, most of which tend to lie near magic numbers. At present, we do not know whether this tendency is caused by the true nature of the mass surface or by our particular choice of the input data.

The extrinsic errors are shown in Figs. 5 and 6. They are rather small for all Z and N, and do not show any remarkable tendency.

In § 3 we have obtained a formula (Eq. (20)) to calculate the errors of masses. In case of interpolation this formula is reasonable, but in case of extrapolation the errors given by this formula seem to be too small. In our results there is a tendency that α_Z and β_N are large when the numbers of the input data, n_Z and n_N, are large. Taking it into account, we have devised a prescription to increase the

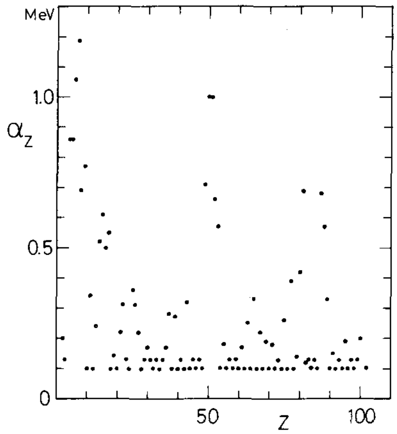

Fig. 3. The intrinsic error of the proton shell term α_Z plotted against Z.

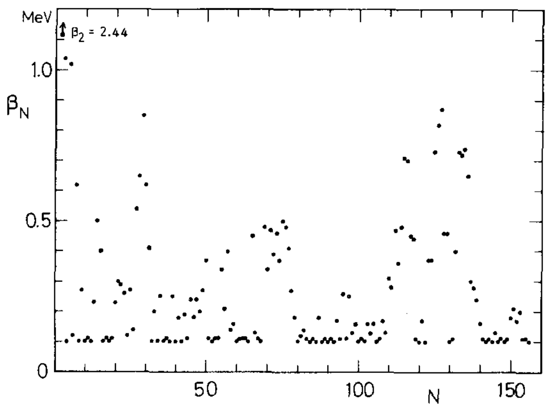

Fig. 4. The intrinsic error of the neutron shell term β_N plotted against N.

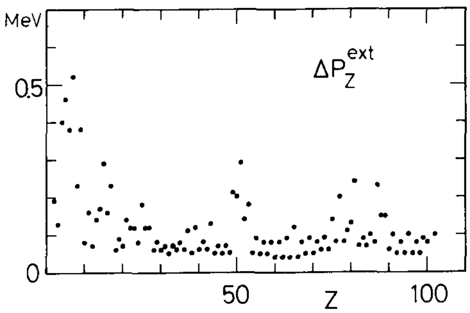

Fig. 5. The extrinsic error of the proton shell term ΔP_Z^{ext} plotted against Z.

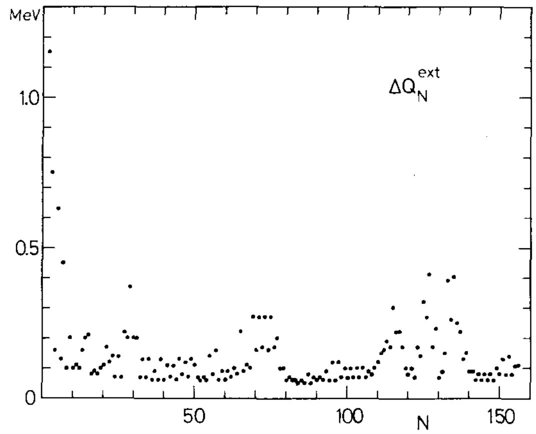

Fig. 6. The extrinsic error of the neutron shell term ΔQ_N^{ext} plotted against N.

calculated error. As it is closely related to the formula with linear shell terms, it will be given in the paper dealing with the linear-shell-term formula. The comparison of our calculated masses of Rb and Cs isotopes with the recent experimental data[7] was already reported at the Sixth International Conference on Atomic Masses.[4]

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