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### Estimation of the alpha decay half-lives

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**Résumé.** — Des versions améliorées des formules utilisées pour estimer les durées de vie de la désintégration  $\alpha$  (Fröman, Wapstra *et al.*, Viola-Seaborg, Hornshøj *et al.*, Taagepera-Nurmia et Keller-Münzel) ont été obtenues en modifiant les paramètres additifs pour annuler la valeur moyenne des erreurs absolues. Une nouvelle relation semiempirique est obtenue en utilisant la théorie de la fission. Elle dépend explicitement de l'écart du nombre de protons, ainsi que du nombre de neutrons, aux nombres magiques voisins et permet d'obtenir un meilleur accord avec les résultats expérimentaux.

Abstract. — Improved up to date versions of the known  $\alpha$  decay half-lives formulae (Fröman, Wapstra *et al.*, Viola-Seaborg, Hornshøj *et al.*, Taagepera-Nurmia and Keller-Münzel) are obtained by changing the additive parameters in order to have a vanishing mean value of the absolute errors. A new semiempirical relationship is derived on the grounds of the fission theory of alpha decay. It takes into consideration explicitly the dependence on the difference from magicity both of the neutron and proton numbers. allowing us to obtain better agreement with experimental data.

#### 1. Introduction.

During the last few years, the number of known alpha emitters has been increased, mainly by measuring the activity of new neutron deficient nuclei produced in heavy ion reactions [1-6]. A new island of alpha activity in the neighbourhood of the double magic nucleus 100 Sn was studied [7].

Alpha decay competes usually with fission and beta decay in the disintegration of the heaviest nuclei synthesized up to now [8]. It is expected to be often encountered in the superheavy region [9].

As far back as 1911, Geiger and Nuttall found a simple dependence of the alpha decay partial halflife, T, on the alpha particle range in air. Now the disintegration period can be estimated, if the kinetic energy of the emitted particle,  $E_{\alpha}$ , is known, by using semiempirical relationships [10-15]. Some of these formulae were derived only for a limited region of the parent proton and neutron numbers Z and N = A - Z. Their parameters have been determined by fitting a given set of experimental data.

Since then the precision of some measurements was increased and new alpha emitters were discovered. Consequently it is interesting to have from time to time the possibility of changing some of the parameter values. A better agreement with experimental results can be obtained by changing the parameters {  $C_k$  } of the various formulae presented below. In an attempt to improve the description of data even in the neighbourhood of the magic neutron and proton numbers, where the errors of the other relationships are large, a new formula with six parameters  $\{B_k\}$ , based on the fission theory of alpha decay [16] have been derived and was briefly presented in reference [17].

A corresponding computer program [18] allows us to improve automatically the parameters  $\{C_k\}$ and  $\{B_k\}$  mentioned above, each time a better set of experimental data is available.

The purpose of this paper is to present the derivation of our formula and to compare the results of estimations with the measured half-lives.

Our set of 376 data on the strong (favoured) alpha transitions of 123 even-even, 111 even-odd, 83 odd-even and 55 odd-odd nuclei, are presented elsewhere [19].

The released energy Q, is related to  $E_{\alpha}$  by the equation

$$Q = E_{\alpha} A/A_{\rm d} , \qquad (1)$$

where  $A_d = A - 4$  is the mass number of the daughter nucleus. Except in a small number of cases (some isotopes of Te, I, Xe, Cs, Hf, Ta, Os, Ir and No), the *Q*-values were determined from the masses tabulated by Wapstra and Bos [20]. To find the partial decay life time of the most probable alpha transition

$$T = \frac{100}{b_{\alpha}} \cdot \frac{100}{i_{p}} T_{\iota}, \qquad (2)$$

we have used the total disintegration period,  $T_{i}$ , the alpha branching ratio,  $b_{\alpha}$ , and the intensity of the considered alpha transition  $i_{p}$  (in percent) compiled by Rytz [21] and also references [1-4, 6, 7, 22, 23]. The experimental values of T will be denoted by  $T_{exp}$ .

#### 2. New additive parameters in the known formula.

The formula given by Fröman [10]

$$\log T = [139.8 + 1.83(Z - 90) + 0.012(Z - 90)^2] / \sqrt{Q} - 0.3(Z - 90) - 0.001(Z - 90)^2 + C_F, \quad (3)$$

is limited to the region of even-even nuclei with  $Z \ge 84$ . Q-values are expressed in MeV and T in seconds throughout in this work.

Almost all parameters  $\{C_k\}$  are negative. Hence the values  $-C_k$  are given in table I. The original parameter value is called « old » and the new one is obtained from the condition that the mean value of the absolute error  $(1/n) \sum_{i=1}^{n} \log (T_i/T_{iexp})$  vanishes in each group of the nuclei mentioned above. The very simple

relationship of Wapstra et al. [11] :

$$\log T = (1.2 Z + 34.9) / \sqrt{Q} + C_{\rm w}, \qquad (4)$$

is also valid for even-even nuclei with  $Z \ge 85$ .

The formula presented by Taagepera and Nurmia [12]

$$\log T = 1.61(Z_{\rm d}/\sqrt{E_{\alpha}} - Z_{\rm d}^{2/3}) + C_{\rm T}, \qquad (5)$$

(where  $Z_d = Z - 2$  is the atomic number of the daughter nucleus and  $C_T$  was allowed to vary for different groups of nuclei) was improved by Keller and Münzel [14] to

$$\log T = H_{\rm K}(Z_{\rm d}/\sqrt{Q} - Z_{\rm d}^{2/3}) + C_{\rm k}, \qquad (6)$$

Table I. — Initial and improved values of the parameters  $(-C_{\rm t})$ .

where  $H_{\rm K} = 1.61$  for even-even (e-e); 1.65 for evenodd (e-o); 1.66 for odd-even (o-e) and 1.77 for odd-odd (o-o) nuclei.

The equation given by Viola and Seaborg [13] is of the form

$$\log T = (a_1 Z - a_2) / \sqrt{Q - b_1 Z - b_2 + C_V}, \quad (7)$$

where  $a_1 = 2.42151$ ;  $a_2 = 62.3848$ ;  $b_1 = 0.59015$ ;  $b_2 = 4.2109$  for N < 126 and  $a_1 = 2.11329$ ;  $a_2 = 48.9879$ ;  $b_1 = 0.39004$ ;  $b_2 = 16.9543$  for N > 126, Z > 82.



Fig. 1. — The errors of life-time predictions with Taagepera-Nurmia's (a), Keller-Münzel's (b), Viola-Seaborg's (c) and Hornshøj *et al.* (d) formulae for even-odd nuclei.

«k»	even-even		odd-even		even-odd		odd-odd	
	old	new	old	new	old	new	old	new
F W T K V H	52.3 53. 21.02 20.2 0 20.279	51.699 52.400 20.789 20.226 0.043 20.347	52.3 53. 20.64 20.7 - 0.772 20.279	51.317 52.026 20.470 20.643 - 0.196 20.051	52.3 53. 20.86 20.5 - 1.066 20.279	51.299 51.940 20.346 20.383 – 0.339 19.922	52.3 53. 20.33 20.8 - 1.114 20.279	50.705 51.377 19.758 20.571 - 0.962 19.355

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$$\log T = 0.803 \ 07 \left(\frac{A_d^{4/3} Z_d}{A}\right)^{1/2} \times \left(\frac{\arccos\sqrt{x}}{\sqrt{x}} - \sqrt{1-x}\right) + C_H, \quad (8)$$

in which  $x = 0.538 \ 243 \ QA_d^{1/3}/Z_d$  and  $C_H$  is not changed in various groups of nuclei, like  $C_F$  of equation (3) and  $C_W$  of equation (4).

In spite of the strong influence of the neutron shell effects, in equations (3)-(8), the Z dependence was stressed. The neighbourhood of the magic number of nucleons is badly described by all these formulae; in  $T/T_{\rm exp}$  diagrams there are negative peaks larger than one order of magnitude at N = 126 for even N and at N = 127 for odd N nuclei (see as an example Fig. 1 for even-odd alpha emitters).

Extremely large negative errors are obtained for Z = 83, N = 127 (5.6; 5.2; 6.4 and 5.8 orders of magnitude). For clarity, the consecutive isotopes of a given element in figures 1, 3 and 4 are connected by a line segment, and a dashed line is used if one or more isotopes of a sequence are missing. From N = 60 to 82 there is a gap in the yet discovered  $\alpha$ -emitters or nuclides which are stable against alpha decay. Up to now only a few components of the new island of alpha activity, close to the double magic <sup>100</sup>Sn, have been found.

#### 3. A new formula.

By applying the phenomenological fission theory with the Myers-Swiatecki [24] variant of the liquid drop model to the alpha decay, it was shown [16] that the potential barrier, for the split of a particular parent nucleus into its daughter component and an alpha particle, is of the shape shown in figure 2, where  $E' = Q + E_{vib}$  and  $E_{vib}$  is the zero point vibration frequency. For  $E_{vib} = 0.4$  MeV one has

$$T = 3.58 \times 10^{-21} \exp(K) \,\mathrm{s} \,. \tag{9}$$

The WKB formula of penetrability leads to

$$K = \frac{2}{\hbar} \int_{R_{\rm a}}^{R_{\rm b}} \left[ 2 \,\mu(E(R) - E') \right]^{1/2} \,\mathrm{d}R \,, \qquad (10)$$



Fig. 2. — The barrier shape for alpha decay.

in which  $\mu = (4 A_d/A) m$  is the reduced mass, m is the nucleon mass and  $\hbar$  is the Planck constant.

By choosing two intervals of integration  $(R_a, R_t)$  and  $(R_t, R_b)$  the action integral is split in two terms  $K = K_i' + K_s'$  corresponding to the overlapping and the separated fragments respectively. The main contribution,  $K_s'$ , comes from the separated fragments, where the potential energy is the Coulomb interaction

$$E(R) = \frac{2 Z_{\rm d} e^2}{R}, \ R \ge R_{\rm t} = R_{\rm d} + R_{\alpha}, \ (11)$$

in which e is the electron charge and  $R_d = r_0 A_d^{1/3}$ ,  $r_0 = 1.2249$  fm. With the substitution  $R = R_b' \cos^2 \xi$ , the integration of  $K_s'$  is performed easily and leads to an analytical relationship. It is approximated by a larger quantity obtained by replacing E' by Q and consequently  $R_b'$  by  $R_b = 2 Z_d e^2/Q$ . Now  $K = K_i + K_s$ and

$$K_{\rm s} = \frac{8 \ e^2}{\hbar} \sqrt{2 \ m} \ Z_{\rm d} \left(\frac{A_{\rm d}}{AQ}\right)^{1/2} \times \\ \times \left[ \arccos \sqrt{x} - \sqrt{x(1-x)} \right], \quad (12) \\ x = R_{\rm t}/R_{\rm b} = r_0 (A_{\rm d}^{1/3} + 4^{1/3}) \ Q/(2 \ Z_{\rm d} \ e^2).$$

The contribution of the nuclear potential is given by  $K'_i$  which has been computed numerically [16]. It can be approximated as a small percentage of  $K_s$  giving us

$$K = \chi K_{\rm s}, \tag{13}$$

where  $\chi$  is different for various nuclides, and it can be either greater or less than unity. For each of the nuclei of our set of experimental data one can determine an « experimental » value

$$\chi_{exp} = \ln 10(\log T_{exp} + 20.446)/K_s,$$
 (14)

where, after replacing the numerical constants, one has from equation (12)

$$K_{\rm s} = 2.529\,56\,Z_{\rm d} \left(\frac{A_{\rm d}}{AQ}\right)^{1/2} \times \\ \times \left[\arccos\sqrt{x} - \sqrt{x(1-x)}\right], \quad (15)$$
$$x = 0.425\,3\,Q(1.584\,7 + A_{\rm d}^{1/3})/Z_{\rm d}.$$

Equation (9) becomes

$$\log T = \chi K_{\rm s} / \ln 10 - 20.446 \,. \tag{16}$$

The figures 3a, b, c, d, show the variation of the quantity  $\chi_{exp}$  for e-e, o-e, e-o and o-o groups of nuclei respectively. For e-e nuclei (Fig. 3a), there is a systematic saw-tooth variation :  $\chi_{exp}$  increases slowly when N is increased between two successive magic numbers and decreases steeply from magic to magic-plus-two neutron numbers. The same thing happens for o-e nuclei (Fig. 3b), though the dispersion of data is more pronounced. For e-o (Fig. 3c) and o-o (Fig. 3d)



Fig. 3. — The experimental values of the coefficient  $\chi = K/K_s$  for even-even (a), odd-even (b), even-odd (c) and odd-odd (d) nuclei.

nuclei the maximum values of  $\chi_{exp}$ , reached at the magic-plus-one number of neutrons is greater. The e-o nuclei (Fig. 3c) show a very sharp peak at N = 127. The very special behaviour of the data for Z = 83, N = 127 which was observed when the known formula had been analysed, is also present in the figure 3d.

The variation of  $\chi_{exp}$ , plotted in figure 3, suggests that the rising part of  $\chi$  could be approximated by some simple laws of variation with Z and N : e.g. a constant value, a first order polynomial or a second order polynomial. The saw-tooth is obtained if N and Z are replaced by the reduced variables y and z :

$$\chi = B_1 + B_2 y + B_3 z + B_4 y^2 + B_5 zy + B_6 z^2,$$
(17)

expressing the distance from the closest magic-plusone number  $N_i$  (or  $Z_i$ ):

$$y \equiv (N - N_i)/(N_{i+1} - N_i); \quad N_i < N \le N_{i+1}$$
  

$$N_i = \dots, 51, 83, 127, 185, \dots$$
(18)

$$z \equiv (Z - Z_i)/(Z_{i+1} - Z_i); \quad Z_i < Z \le Z_{i+1}$$
  

$$Z_i = ..., 29, 51, 83, 115, ...$$
(19)

The parameters  $\{B_k\}$  are found from the fit with our set of experimental data.

#### 4. The fit with experimental data.

The value of the parameter  $B_1$  for the simple constant approximation of  $\chi = B_1$ , can be obtained straightforwardly by using the least squares method. The sum  $\sum_{i=1}^{n} \log (T_{i \exp}/T_i)$  is minimized with respect to  $\chi$ giving

$$B_1 = \ln 10 \sum_{i=1}^{n} (\log T_{i \exp} + 20.446) K_{si} / \sum_{i=1}^{n} K_{si}^2, (20)$$

in each of the four groups of nuclei (n = 123 for e-e, 83 for o-e, 111 for e-o and 59 for o-o nuclei). In this way the following figures have been obtained :  $B_1 = 1.002410$  for e-e;  $B_1 = 1.016046$  for o-e;  $B_1 = 1.019613$  for e-o and  $B_1 = 1.049592$  for o-o nuclei.

For the first order and the second order polynomial  $\chi = \chi(y, z)$  a numerical procedure [18] has been used in order to find the parameters  $B = \{B_k\}$  minimizing

Table II. — Parameters  $B_k$  for the first order and second order polynomials.

Group of nuclei	B <sub>1</sub>	B <sub>2</sub>	B <sub>3</sub>	B <sub>4</sub>	B <sub>5</sub>	B <sub>6</sub>
even-even	0.988 675 0.987 722	0.041 423 0.021 227	- 0.000 179 0.017 407	0.023 342	 0.001 546	 - 0.016 509
odd-even	1.010 867	0.026 086	- 0.012 949			
	1.003 660	0.021 626	0.034 805	0.000 870	0.039 190	- 0.066 147
even-odd	0.995 703	0.068 843	- 0.000 178			
	1.014 620	- 0.119 094	0.030 606	0.156 507	0.228 848	- 0.109 130
odd-odd	1.008 055	0.089 698	0.008 549			
	1.008 070	- 0.184 468	0.259 041	0.231 988	0.326 171	_ 0.406 200



Fig. 4. — The errors of life-time prediction with the present formula when  $\chi$  is approximated with (a) a constant; (b) a first order polynominal; (c) a second order polynominal of 2 variables for even-even nuclei; (d) as (c) for odd-even nuclei; (e) as (c) for even-odd nuclei and (f) as (c) for odd-odd nuclei.

the sum of the squares of the deviations. The parameter values obtained in this way are given in table II.

The capacity of our formula to describe the experimental data can be judged from figure 4. For eveneven nuclei, figure 4a shows the constant approximation, figure 4b (the first order polynomial approximation) and figure 4c the second order polynomial approximation. The increased error in the vicinity of the magic number of neutrons N = 126, which is present for all known formulae and for the constant  $\chi$ (Fig. 4a) is practically smoothed out by the second order polynomial approximation. This behaviour is only partly achieved for o-e (Fig. 4d), e-o (Fig. 4e) and o-o (Fig. 4f) nuclei. Even the very large error of 5-6 orders of magnitude mentioned in section 2 for Z = 83, N = 127, is greatly reduced, below 0.4 of an order of magnitude.

An overall estimation of how well various formulae can describe the experimental data could be quantitatively obtained by introducing the standard rms deviations of  $\log T$  values :

$$\sigma = \left\{ \sum_{i=1}^{n} \log \left( T_{i} / T_{exp} \right)^2 / (n-1) \right\}^{1/2}.$$
 (21)

Even the constant  $\chi$  approximation of our formula has  $\sigma$  somewhat lower than the best of the known relationships [14]. Of course the second order polynomial approximation leads to smaller standard deviations.

#### 5. Conclusions.

A relatively reliable estimate of the alpha decay partial half-life can be made by using semiempirical relationships. After changing the additive parameters of the examined formulae in such way that the mean value of the errors  $(1/n) \sum_{i=1}^{n} \log (T_i/T_{i\exp})$  vanishes in each group of the nuclei (e-e, o-e, e-o and o-o), the standard deviation,  $\sigma$ , of log T values is usually reduced. In this way an up to date version of the known formulae has been obtained. The Keller-Münzel relationship is the best of all formulae, but even this has an increased error in the vicinity of the magic number of neutrons N = 126.

This is practically smoothed by the present formula for e-e nuclei. The shell effects have been taken into consideration by introducing explicitly the dependence on the reduced variables expressing the relative distance of neutron and proton from the closest magic-plus-one numbers. In this way one can obtain the smallest errors in all groups of nuclei. The new formula was obtained by splitting the contribution to the WKB action integral into two parts for the overlapping and for the separated fragments. The main contribution is given by the Coulomb interaction (separated fragments) and is calculated easily by a closed relationship. The nuclear force term (overlapping region) is approximated as a small percentage of the Coulomb part with the parameter values determined from a fit with experimental data. In the future when a better set of experimental data (more accurate or more complete) shall be available, the parameters  $\{B_k\}$  and  $\{C_k\}$  could be better approximated using a computer program [18].

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