α-decay half-lives of superheavy nuclei

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The half-lives against α decay of transuranium nuclei including superheavies are calculated by three methods: a semiempirical formula taking into account the magic numbers of nucleons, the analytical superasymmetric fission model, and the universal curves. The calculations based on Q values determined by using the recently published compilations of atomic masses are compared to the experimental results.

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I. INTRODUCTION

Superheavy elements exist only because of their nuclear shell effects. Consequently, one may start the series of the heaviest elements with rutherfordium, $Z = 104$. Because of the extra stability from nuclear shell effects, the known isotopes of rutherfordium exhibit half-lives of up to 1 min, which is 16 orders of magnitude longer than the expected nuclear lifetime of 10^{-14} s. These isotopes would not survive without any extra shell stabilization. Spontaneous fission, the dominating decay mode in the region around Rf, becomes a relatively weaker branch compared to α decay for the majority of recently discovered nuclides [1–3]. Consequently it is important to study different methods of estimating the half-lives against α decay. Several works have been performed during the past several years, some of them also related to the fusion reactions (see Refs. [4–10] and the references therein).

As early 1911 Geiger and Nuttal found, for the members of a given natural radioactive family, a simple purely empirical dependence [11] of the α-decay partial half-life, T_α, on the mean α-particle range, $\mathcal{R}_\alpha$, in air (at 15°C and 1 atm), which may be written [12,13] as:

$$\log_{10} T_\alpha(\text{in s}) = -57.5 \log_{10} \mathcal{R}_\alpha(\text{in cm}) + C, \tag{1}$$

where C depends on the series, e.g., $C = 41$ for the ^{238}U series. One has approximately $\mathcal{R}_\alpha = 0.325 E_\alpha^{3/2}$ in which the kinetic energy of α particles, E_α, is expressed in MeV and the range in air, $\mathcal{R}$, in cm. This relationship is now of historical interest; the effect of atomic number, Z, on decay rate is obscured [14]. The one-body theory of α decay can explain it and to a good approximation produces a formula with an explicit dependence on the Z number. Nowadays, very often a diagram of $\log_{10} T_\alpha$ versus $ZQ^{-1/2}$ is called Geiger-Nuttal plot [15,16].

There are many semiempirical relationships (see for example Refs. [17–27]), which allows us to estimate the disintegration period if the kinetic energy of the emitted particle $E_\alpha = QA_d/A$ is known. Q is the released energy and A_d, A are the mass numbers of the daughter and parent nuclei. α-decay half-life of an even-even emitter can also be easily calculated by using the universal curves [28,29] or the analytical superasymmetric (ASAF) model [30]. Some of these formulas were derived only for a limited region of the parent proton and neutron numbers. Their parameters have been determined by fitting a given set of experimental data. Since then, the precision of the measurements was increased and new α emitters have been discovered [31].

The description of data in the neighborhood of the magic proton and neutron numbers, where the errors of the other relationships are large, was improved by deriving a new formula (SemFIS) based on the fission theory of α decay [32]. A computer program [33] allows us to change automatically the fit parameters every time a better set of experimental data are available. There are many α emitters, particularly in the intermediate mass region, for which both the Q values and the half-lives are well known [34–37]. Initially we used a set of 376 data [123 even-even (e-e), 111 even-odd (e-o), 83 odd-even (o-e), and 59 odd-odd (o-o)] on the most probable (ground-state-to-ground-state or favored transitions) α decays, with a partial decay half-life

$$T_\alpha = (100/b_\alpha)(100/i_p)T_t, \tag{2}$$

where b_α and i_p, expressed in percentages, represent the branching ratio of α decay in competition with all other decay modes and the intensity of the strongest α transition, respectively.

Some authors are using an effective Q value $Q_{\rm eff} = Q + \Delta E_s$, by taking into consideration a small term

$$\Delta E_s = (65.3 Z^{7/5} - 80 Z^{2/5}) 10^{-6}\ \text{MeV} \tag{3}$$

of the order of 15 to 30 keV, because of the electronic shielding. The complication introduced in such a way in the otherwise relatively simple formula is not justified by an improvement of the agreement with experimental data. Also, according to the discussion presented in Ref. [38] this contribution is practically canceled by another one of opposite sign.

The formula given by Fröman [17] is limited to the region of even-even nuclei with $Z \geqslant 84$. This formula describes well the experimental data of nuclei with $N \geqslant 128$ but fails in the region of lighter α emitters, which have not been available at the moment of its derivation. A better overall result gives a simple relationship of Wapstra *et al.* [18] also valid for even-even nuclei with $Z \geqslant 85$. In the new variant derived by A. Brown [24] for nuclei with $Z \geqslant 72$ the agreement with experimental

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TABLE I. B_k parameter values obtained by fitting the data evaluated by Rytz.

	B_1	B_2	B_3	B_4	B_5	B_6
e-e	0.993119	−0.004670	0.017010	10.045030	0.018102	−0.025097
o-e	1.000560	0.010783	0.050671	0.013919	0.043657	−0.079999
e-o	1.017560	−0.113054	0.019057	0.147320	0.230300	−0.101528
o-o	1.004470	−0.160056	0.264857	0.212332	0.292664	−0.401158

data is not bad for nuclei with $Z \geqslant 72$, but large errors are obtained for lighter parent nuclei.

The formula presented by Taagepera and Nurmia [19] remains one of the best. It is exceeded by a variant presented by Keller and Münzel [21]. Viola and Seaborg [20] introduced a relationship that shows excellent agreement in the region of actinides but underestimates the lifetimes of lighter nuclei, in contrast with overestimations obtained with the first of the above mentioned formulae.

In the region of superheavy nuclei the majority of researchers prefer to use the Viola-Seaborg formula. Very recently for nuclei with $Z = 84$–110 and $N = 128$–160, for which both $Q_\alpha^{\rm exp}$ and $T_{\rm exp}$ experimental values are available [36,37], new optimum parameter values [27] have been determined. The average hindrance factors for 45 o-e ($Z = 85$–107), 55 e-o ($Z = 84$–110), and 40 o-o ($Z = 85$–111, $N = 129$–161) nuclei were determined to be $C_V^p = 0.437$, $C_V^n = 0.641$, and $C_V^{pn} = 1.024$. In this way $T_{\rm exp}$ were reproduced by the Viola-Seaborg formula within a factor of 1.4 foe e-e, 2.3 for o-e, 3.7 for e-o, and 4.7 for o-o nuclei, respectively. For the region of mass numbers $A = 266$–294 a recently performed evaluation of experimental results [39,40] gives different values of the parameters.

Good results were obtained with a formula by Royer [26] that has 12 parameters, a,b,c, for e-e, e-o, o-e, and o-o nuclei. Shell effects were not taken into account; nuclei with neutron number close to the shell closures $N = 152$ and 162 [namely three nuclei with $N = 151 (Z = 96, 98, 100)$ one with $N = 153$ $Z = 98$, and one with $N = 161$ $Z = 110$] have been omitted in the fitting procedure. Other omission of 3 o-e nuclei with $Z = 97, N = 146, 148$ and $Z = 101, N = 154$ was motivated by a large deviation from the average behavior. A simple version of the Viola-Seaborg formula was proposed by Parkhomenko and Sobiczewski [27].

Several works on α decay of superheavy nuclei are using the relativistic mean-field model to calculate the interaction potential and/or the Q values [41–43]. Alternatively, the interaction potential may also be obtained by employing the M3Y effective interaction [44] and the binding energies can be estimated by a semiempirical shell model [45,46].

II. SEMIEMPIRICAL RELATIONSHIP BASED ON FISSION THEORY OF α DECAY

Mainly the Z dependence was stressed by all formulae, despite strong influence of the neutron shell effects. The neighborhood of the magic numbers of nucleons is badly described by all these relationships.

The SemFIS formula based on the fission theory of α decay yields

$$\log_{10} T = 0.43429 K_s \chi - 20.446, \tag{4}$$

where

$$K_s = 2.52956 Z_{\rm da}[A_{\rm da}/(A Q_\alpha)]^{1/2}[\arccos\sqrt{x} - \sqrt{x(1-x)}];$$
$$x = 0.423 Q_\alpha \left(1.5874 + A_{\rm da}^{1/3}\right)/Z_{\rm da} \tag{5}$$

and the numerical coefficient χ, close to unity, is a second-order polynomial

$$\chi = B_1 + B_2 y + B_3 z + B_4 y^2 + B_5 yz + B_6 z^2 \tag{6}$$

in the reduced variables y and z, expressing the distance from the closest magic-plus-one neutron and proton numbers N_i and Z_i:

$$y \equiv (N - N_i)/(N_{i+1} - N_i); \quad N_i < N \leqslant N_{i+1} \tag{7}$$

$$z \equiv (Z - Z_i)/(Z_{i+1} - Z_i); \quad Z_i < Z \leqslant Z_{i+1} \tag{8}$$

with $N_i = \ldots, 51, 83, 127, 185, 229, \ldots, Z_i = \ldots, 29, 51, 83, 115, \ldots,$ and $Z_{\rm da} = Z - 2$, $A_{\rm da} = A - 4$. The coefficients B_i obtained by using a high-quality selected set of α-decay data [35] are given in Table I. Better agreement with experimental results are obtained in the region of superheavy nuclei by introducing other values of the magic numbers plus one unit for protons (suggesting that the next magic number of protons could be 126 instead of 114): $Z_i = \ldots, 83, 127, 165, \ldots$.

With the SemFIS formula, Rurarz [25] have made predictions for nuclei far from stability with $62 < Z < 76$. In the variant of Ref. [23] the shell effect on the formation factor was approximated by an empirical relationship.

Practically for even-even nuclei, the increased errors in the neighborhood of $N = 126$, present in all other cases, are smoothed out by SemFIS formula using the second-order polynomial approximation for χ (see Fig. 1). They are still present for the strongest α decays of some even-odd and odd-odd parent nuclides. In fact for noneven number of

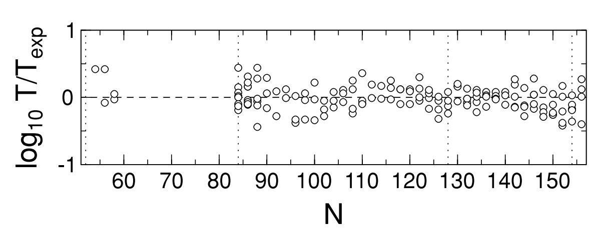

FIG. 1. The deviations of α-decay half-lives calculated with the SemFIS formula from the experimental values for even-even nuclei. The vertical bars correspond to spherical and deformed neutron magic numbers of the daughter nuclei $N_d = 50, 82, 126$, and 152.

nucleons the structure effects became very important, and they should be carefully taken into account for every nucleus, not only globally. An overall estimation of the accuracy yields the standard rms deviation of $\log_{10} T$ values:

$$\sigma = \left\{ \sum_{i=1}^{n} [\log_{10}(T_i/T_{\mathrm{exp}})]^2/(n-1) \right\}^{1/2}. \quad (9)$$

The parameters $\{B_k\}$ of the SemFIS formula could be automatically improved, for a given set of experimental data, by use of the computer program described in the Ref. [33]. We used such improved parameter values to calculate the theoretical half-lives of transuranium nuclei plotted in the lower part of Fig. 2. The partial α-decay half-lives plotted in this figure lie in the range of 10^{-7} to 10^{25} sec. One can see the effect of the spherical and deformed neutron magic numbers of the daughter nuclei $N_d = 126$, 152, 162, which are particularly clear for even-even and even-odd nuclides.

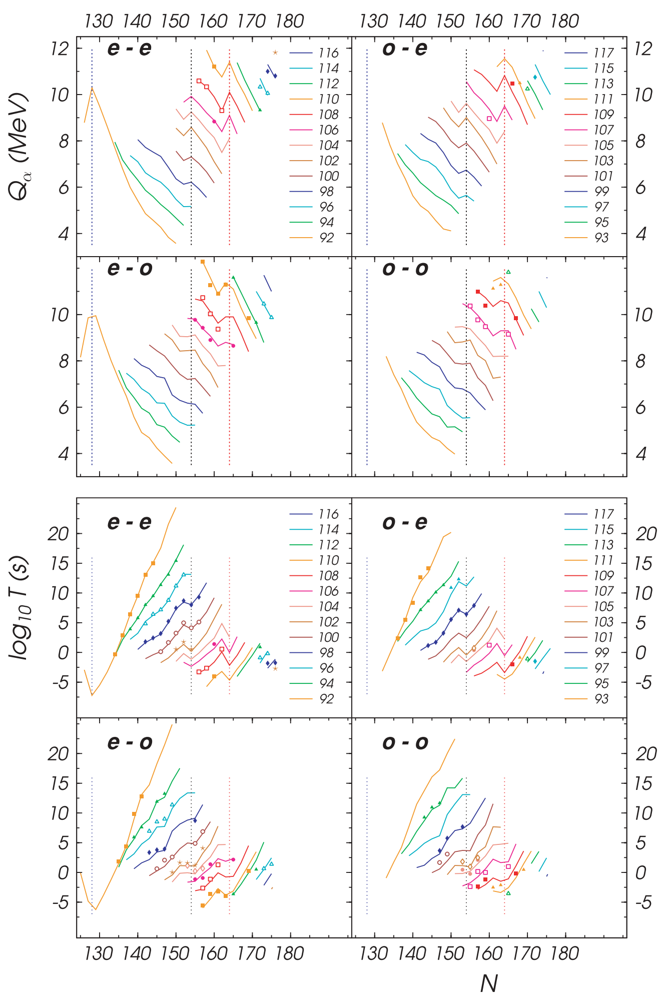

FIG. 2. (Color online) (Top) Q values for α decay of transuranium nuclei versus neutron number in four groups of even-even, even-odd, odd-even, and odd-odd nuclei. (Bottom) Partial half-lives for α decay of transuranium nuclei versus neutron number in four groups of even-even, even-odd, odd-even, and odd-odd nuclei. Calculations are performed with SemFIS formula. The vertical bars correspond to spherical and deformed neutron magic numbers of the daughter nuclei $N_d = 126$, 152, 162.

TABLE II. Standard deviations for semiempirical formula, ASAF model, and universal curves in the region of transuranium nuclei.

Group	σ-ASAF	σ-univ	σ-SemFIS
36 e-e	0.276	0.222	0.119
47 e-e	0.402	0.267	0.164
29 e-o	0.729	0.623	0.486
45 e-o	0.615	0.554	0.507
20 o-e	0.727	0.540	0.399
25 o-e	0.761	0.543	0.485
13 o-o	0.969	0.521	0.301
25 o-o	0.795	0.456	0.451

For the large set of α emitters we obtained the following values of the rms errors of $\log_{10} T$: 0.19 for SemFIS formula; 0.33 for the universal curve; 0.39 for ASAF model, and 0.43 for numerical superasymmetric (NuSAF) model [30]. The present set of transuranium nuclei is still preliminary because both the Q values and the half-lives of many nuclei need to be determined with an improved accuracy. Consequently, the standard deviations shown in Table II for the transuranim nuclei, including superheavies, are larger than they should be. The Q values at the top of Fig. 2 are calculated by use of the atomic masses from the compilation [36] where one may find either measured values or obtained from the systematics ones. The points are taken from Nuclear Data Sheets [39].

For the α decay of transuranium heavy and superheavy nuclei we took the latest values of experimentally determined half-lives from Refs. [39,40]. We also used some other sources [31,34,35]. The experimental data at the bottom of Fig. 2 are selected from these works, including the list of references of Ref. [47] used to calculate the branching ratios relative to α decay of cluster-emitting nuclei. The spherical and deformed neutron magic numbers of the daughter nuclei $N_d = 126$, 152, 162 are displayed in both systematics.

There are many parameters of the SemFIS formula introduced to reproduce the experimental behavior around the magic numbers of protons and neutrons, which could be a drawback in the region of light and intermediate α emitters. In the region of superheavies these characteristics may be conveniently used to get information concerning the next magic numbers of protons and neutrons, which were not well known until recently. When accurate experimental values of Q and T are available in the region centered on $Z = 114$–126, $N = 172$–184, the SemFIS formula may be used to estimate whether the right value of the spherical magic number is $Z = 114$, 120, 126, and $N = 172$ or 184, because of the high sensitivity of χ to the values of Z_i and N_i [see Eqs. (4)–(8)].

III. UNIVERSAL CURVES FOR α-DECAY

In cluster radioactivity and α decay the (measurable) decay constant $\lambda = \ln 2/T$, can be expressed as a product of three (model-dependent) quantities

$$\lambda = \nu S P_s, \tag{10}$$

where ν is the frequency of assaults on the barrier per second, S is the preformation probability of the cluster at the nuclear surface, and P_s is the quantum penetrability of the external potential barrier. The frequency ν remains practically constant, the preformation differs from one decay mode to another but it is not changed very much for a given radioactivity, whereas the general trend of penetrability follows closely that of the half-life. The external part of the barrier (for separated fragments), essentially of Coulomb nature, is much wider than the internal one (still overlapping fragments).

According to Ref. [29] the preformation probability can be calculated within a fission model as a penetrabilty of the internal part of the barrier, which corresponds to still overlapping fragments. One may assume as a first approximation, that preformation probability only depends on the mass number of the emitted cluster, $S = S(A_e)$. The next assumption is that $\nu(A_e, Z_e, A_d, Z_d) =$ constant. In this way one arrives at a single straight line *universal curve* on a double logarithmic scale

$$\log_{10} T = -\log_{10} P_s - 22.169 + 0.598(A_e - 1), \tag{11}$$

where

$$-\log_{10} P_s = c_{AZ}\left[\arccos\sqrt{r} - \sqrt{r(1-r)}\right] \tag{12}$$

with $c_{AZ} = 0.22873(\mu_A Z_d Z_e R_b)^{1/2}$, $r = R_t/R_b$, $R_t = 1.2249(A_d^{1/3} + A_e^{1/3})$, $R_b = 1.43998 Z_d Z_e/Q$, and $\mu_A = A_d A_e/A$. For all measurements performed until now the agreement is good, as can be seen in Figs. 3 and 4.

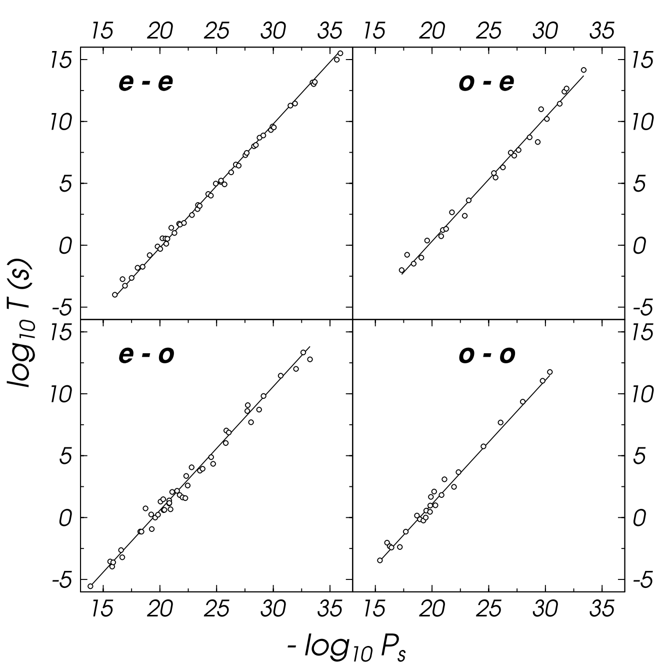

FIG. 3. Universal curves for α decay of transuranium nuclei versus neutron number in four groups of even-even, even-odd, odd-even, and odd-odd nuclei. Calculations are performed with the new constants adjusted to fit the data of transuranium nuclei.

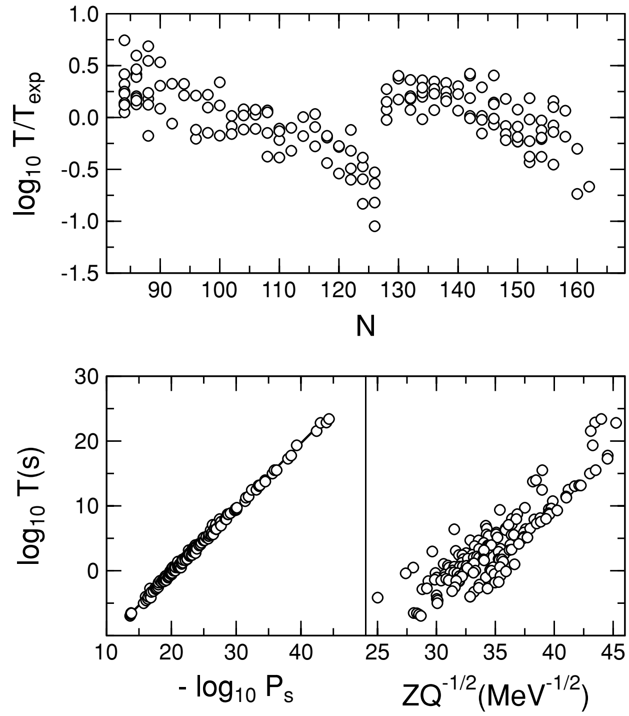

FIG. 4. (Top) The decimal logarithm of the ratio $T/T_{\rm exp}$ versus neutron number, N, showing the strong shell effect at the magic number 126. (Bottom) Comparison of the universal curve for α decay (bottom left) with the "Geiger-Nuttal" systematics (bottom right).

Sometimes this universal curve is misinterpreted as being a Geiger-Nuttal plot [see Eq. (1)]. Nowadays by Geiger-Nuttal diagram one understands a plot of $\log_{10} T$ versus $ZQ^{-1/2}$ or versus $Q^{-1/2}$. In this kind of systematics the experimental points are scattered, as shown in Fig. 4 for α decay of even-even nuclei. Nevertheless, for a given atomic number, Z, or for the members of a natural radioactive series, it is still possible to obtain a single straight line.

The strong shell effect at the magic neutron number $N = 126$, which was ignored when the approximation $S = S(A_e)$ was made, is shown in the upper part of Fig. 4 to give a pronounced underestimation of the half-lives in the neighbourhood of $N = 126$.

For α decay of even-even nuclei, $A_e = 4$, one has

$$\log_{10} T = -\log_{10} P_s + c_{ee} \tag{13}$$

where $c_{ee} = \log_{10} S_\alpha - \log_{10} \nu + \log_{10}(\ln 2) = -20.375$. We can find new values for c_{ee} and we also can extend the relationship to even-odd, odd-even, and odd-odd nuclei, by fitting a given set of experimentally determined α-decay data. The result is illustrated in Fig. 3, where the following values of constants have been used: $c_{ee} = -20.198$ for even-even nuclei, $c_{eo} = -19.412$ for even-odd, $c_{oe} = -19.680$ for odd-even, and $c_{oo} = -18.903$ for odd-odd.

In conclusion the SemFIS formula taking into account the magic numbers of nucleons, the analytical superasymmetric fission model, and the universal curves may be used to estimate the α-emitter half-lives in the region of superheavy nuclei. The dependence on the proton and neutron magic numbers of the semiempirical formula may be exploited to obtain informations about the values of the magic numbers that are still not well known.

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