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70th Anniversary

A NEW INSIGHT IN THE DECAY MODES OF HEAVY NUCLEI

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Abstract. Fission approach to the theory of heavy-particle radioactivities and α-decay, we developed in cooperation with D.N. Poenaru, is briefly reviewed. The potential energy surface for ^{222}Ra versus the separation distance between fragments and the mass asymmetry shows the fission and cluster decay valleys produced by the shell effects due to the ^{132}Sn and ^{208}Pb heavy fragments. The potential barrier for ^{14}C radioactivity of ^{222}Ra is obtained by using the macroscopic-microscopic method. The experimentally determined half-lives of cluster emitters are in good agreement with our predictions within the analytical superasymmetric fission (ASAF) model. The α-decay half-lives calculated by three fission models (ASAF, universal curve and a semiempirical formula taking into account the magic numbers) are very close to the experimental ones in the whole nuclear chart, including superheavy nuclei.

Key words: nuclear decay modes, heavy ion radioactivity, alpha decay, superheavy nuclei.

1. INTRODUCTION

The present issue of the Romanian Reports in Physics is dedicated to Dorin N. Poenaru's 70th Anniversary. In the framework of a longstanding fruitful scientific cooperation during the last 26 years we are very pleased to work together, both in Frankfurt am Main and in Bucharest. Certainly this cooperation will continue in the future. With A. Sandulescu and D.N. Poenaru we are honoured to be mentioned in the *New Encyclopaedia Britannica for calculations, published in 1980* [1], *indicating the possibility of a new type of decay of nuclei: heavy particle radioactivity.* The first successful experiment on ^{14}C radioactivity of ^{223}Ra was reported in 1984 by Rose and Jones [2].

Since 1984 the following types of radioactivities have been experimentally confirmed worldwide (Oxford, Moscova, Orsay, Berkeley, Geneva, Dubna, Argonne, Milano, Viena, Lanzhou, Beijing and Livermore): ^{14}C radioactivity of ^{221}Fr, ^{221,222,223,224,226}Ra, and ^{225}Ac; ^{20}O radioactivity of ^{228}Th; ^{22}Ne radioactivity of ^{230}U; ^{24}Ne radioactivity of ^{230}Th, ^{231}Pa, ^{232}U; ^{24,25}Ne radioactivities of ^{233,235}U; ^{28}Mg radioactivity of ^{234}U, ^{236}Pu; ^{28,30}Mg radioactivities of ^{236}U, ^{238}Pu; ^{32}Si radioactivity of ^{238}Pu; ^{34}Si radioactivity of ^{242}Cm. Also the upper limits were determined in case of ^{18}O radioactivity of ^{226}Th; ^{23}F radioactivity of ^{231}Pa; ^{24,26}Ne radioactivities of ^{232}Th, ^{234}U; ^{28}Mg radioactivity of ^{233,235}U; ^{30}Mg radioactivity of ^{237}Np, and ^{34}Si radioactivity of ^{240}Pu, ^{241}Am. The measured half-lives are in good agreement with theoretical predictions within our analytical superasymmetric fission (ASAF) model. There are still many other experiments which have a good chance to be performed in order to exploit further the strong shell effect of the doubly magic nucleus ^{208}Pb, as we had shown in a recent publication [3].

The calculations we first reported were obtained by using the following models: fragmentation theory [4, 5] and the asymmetric two center shell model [6]; alpha-decay like theory; numerical superasymmetric fission (NuSAF) model and the ASAF model. The ASAF model was particularly useful in predicting the half-lives to guide the experiments. Comprehensive tables with ASAF model predictions have been published [7, 8]. The most cited papers and chapters in books have been the Refs. [1, 9, 7, 8, 10] and [11, 12, 13], respectively. Not only scientific publications for experts, but also more general journals for popularization of science (La Recherche, Science et Vie, Physics Bulletin, Scientific American) and even newspapers from Germany, Hungary, Romania, have recognized our contributions to the foundation and development of a new branch of physics. To my knowledge no other project of scientific cooperation between Germany and Romania had ever received such a high attention.

Dorin Poenaru started to work in fission in 1964, when he begun an international cooperation with JINR Dubna on fission isomers. A very important step forward, which allowed him latter on to solve the problem of extremely asymmetric fission, was the extension of three variants of the liquid drop model for binary systems with different charge densities [14]. He gave a new interpretation of α-decay as a fission phenomenon, which was supported by the excellent agreement between the calculated halflives and the experimental data covering a range of 25 orders of magnitude. Fission theory applied to heavy paticle radioactivity and α-decay proved to be very successful. The culmination of our scientific cooperation was certainly the prediction of new kinds of nuclear decay modes.

Prof. Poenaru has been invited to present talks at the following International Scientific Meetings: Poiana Braşov, Predeal and Mamaia Summer

Schools in 1980, 1984, 1986, 1988, 1990, 1995, 1998, 2005, 2006; NATO Advanced Study Institutes in Predeal and Kemer: 1992, 1993, 2000, 2003; JINR Symposium, Dubna 1984; Conf. of the European Physical Society, Varna, 1985; Conf. on Clustering Aspects in Nuclear and Subnuclear Systems, Kyoto, 1988; Symposium on Developments in Nuclear Cluster Dynamics, Sapporo, 1988; three Confs. (Gaussig, 1988; West Berlin, 1989; Leningrad, 1989) celebrating 50 years of research in nuclear fission; Conf. on Rare Nuclear Decays and Fundamental Processes, Bratislava, 1990; Conf. on Clustering Phenomena in Atoms and Nuclei, Turku, 1991; Conf. on Nuclear Reaction Mechanisms, Varenna, 1991; Conf. on Exotic Nuclei, Foros, 1991; Conf. on Atomic Masses and Fundamental Constants & Nuclei far from Stability, Bernkastel-Küs, 1991; Conf. on Nuclear Physics at the Turn of the Millennium, Wilderness/George, 1996; Conf. on Nuclear Data for Science and Technology, Trieste, 1997; Conf. Advances in Nuclear Physics and related areas, Thessaloniki, 1997; Conf. on Fission and Properties of Neutron-Rich Nuclei, Sanibel Island and St Andrews, 1998, 1999, 2002; Symposium on Perspectives in Nuclear Physics, Atlantis Resort, Nassau, 1998; Workshop on Nuclear Theory, Rila Mountain, 1999 and 2006; Conf. on Clustering Aspects of Nuclear Structure and Dynamics, Rab Island, 1999; Symposium on Advances in Nuclear Physics, Bucharest, 1999; Symposium on Exotic Nuclear Structure, Debrecen, 2000; Symposium on Fundamental Issues in Elementary Matter, Bad Honnef, 2000; Symposium on Advances in Heavy Element Research, Tokai (Japan), 2001; Specialists Meeting on Interdisciplinary Approach to Nuclear Fission, Osaka, 2002; 4th Relativistic Ion Studies Conf. on Exotic Clustering, Catania, 2002; JLAB Workshop on Modern Sub-Nuclear Physics and Jlab Experiments, Athens, 2002; European Research and Education Networking End-user Workshop, Montpellier, 2003; Workshop on New Applications of Nuclear Fission, Bucharest, 2003; Titeica-Markov Symposium, Constanta, 2004; Symposion on Heavy Ion Physics (Gateway to the Unknown: Fundamentality–Complexity–Simplicity), Frankfurt am Main, 2006; General Conf. of the Balkan Physical Union, Istanbul, 2006.

Recently we contributed with a paper on Radioactivity [15] to the new 6 volumes Encyclopedia of Condensed Matter Physics. The most advanced asymmetric two center shell model was improved [16, 17] and applied to calculate potential energy surfaces (PES) for cluster emitters (^{222}Ra, ^{232}U, ^{236}Pu, ^{242}Cm [18]) as well as for ^{228}Th [19] and for light (^{106}Te and ^{212}Po) and superheavy (^{294}118) alpha emitters [20]. The potential barrier shape of heavy ion radioactivity obtained for the first time by use of the macroscopic-microscopic method provides further support for the particular choice of the barrier within the ASAF model. Other applications concerns the sub-barrier synthesis of $Z = 118$ isotopes [21] and the study of input channels to produce ^{286,290,298}114 [22, 23]. The dynamical calculations have been performed in a multidimensional hyperspace of deformation coordinates followed by minimization of the action integral for all possible charge and mass asymmetries. The Werner-Wheeler approximation was employed to obtain the nuclear inertia tensor.

The pairing correction energy calculated within the BCS approximation [18] was observed to give an important contribution to the deformation energy by lowering the barrier heights and smoothing the shell effects. The strong shell effect associated with the doubly magic character of the daughter ^{208}Pb, which was seen in the systematic analysis of experimental results, comes from a valley present on the PESs of cluster emitters at a relatively high value of the asymmetry parameter.

The BCS pairing has an essential contribution to the cranking inertia tensor [24] which may be expressed with an analytical relationship for a particular choice of the system Hamiltonian of a spheroidal harmonic oscillator without spin-orbit interaction. If the crossing terms P_{ij} with $i \neq j$ are not taken into account, an important error could be induced into the half-life value given by the WKB approximation. Examples for ^{240}Pu [24] illustrate the conclusions.

The α-decay life-times of superheavies and lighter emitters have been calculated [25, 20] within our ASAF model, the universal formula, and the semiempirical relationship including shell effects.

2. BARRIER SHAPE OBTAINED WITH THE MACROSCOPIC-MICROSCOPIC METHOD

In order to study a binary decay mode $^{A}Z \rightarrow {}^{A_1}Z_1 + {}^{A_2}Z_2$ the simplest nuclear shape parametrization we have used was that of two intersected spheres of radii $R_1 = r_0 A_1^{1/3}$ and R_2 with a single deformation parameter, the distance R between the two centers (by assuming the total volume conservation). One has initially $R = R_i = R_0 - R_2$ and at the touching point $R = R_t = R_1 + R_2$. R_0 is the radius of the parent nucleus ^{A}Z and the radius constant r_0 was taken to be 1.16 fm in the Yukawa-plus-exponential (Y+EM) phenomenological model. One can use a dimensionless quantity $\xi = (R - R_i)/(R_t - R_i)$ which takes values between 0 and 1 in this stage of two overlapping fragments. The mass asymmetry is defined as $\eta = (A_1 - A_2)/(A_1 + A_2)$.

The macroscopic energy E_{Y+EM} is calculated within Y+EM [26, 27] by taking into account the difference between charge and mass asymmetry [28]. The microscopic shell [29] and pairing [30] corrections $\delta E = \delta U + \delta P$ are added to the macroscpic energy leading to the total deformation energy

$$E_{def}(R, \eta) = E_{Y+EM}(R, \eta) + \delta E(R, \eta). \qquad (1)$$

Details are given in the book [12].

The two-center shell model gives at every pair of coordinates (R, η) the sequence of doubly degenerate discrete energy levels $\epsilon_i = E_i/\hbar\omega_0^0$ in units

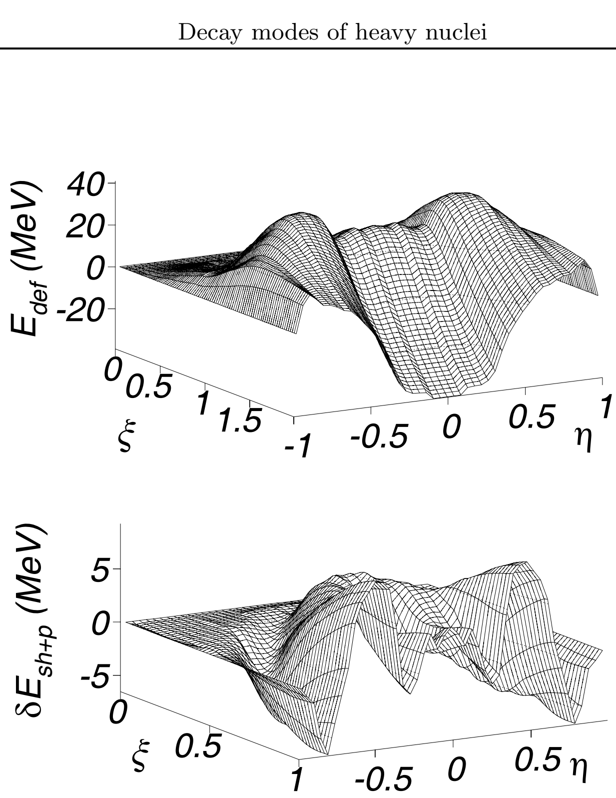

Fig. 1 – Potential energy surfaces of ^{222}Ra vs $\xi = (R - R_i)/(R_t - R_i)$ and $\eta = (A_1 - A_2)/(A_1 + A_2)$. Shell + Pairing corrections (bottom), and total deformation energy (top). The very deep valley around $\eta = 0.87$ shown at the bottom corresponds to the ^{14}C radioactivity with the daughter ^{208}Pb and the other near $\eta = 0.36$ for asymmetric cold fission with the ^{70}Ni light fragment and the ^{152}Nd daughter.

of $\hbar\omega_0^0 = 41A^{-1/3}$, arranged in order of increasing energy. The smoothed-level distribution density is obtained by averaging the actual distribution over a finite energy interval $\Gamma = \gamma\hbar\omega_0^0$, with $\gamma \simeq 1$. In units of $\hbar\omega_0^0$ the shell corrections are calculated for each pair (R, η):

$$\delta u(n, R, \eta) = \sum_{i=1}^{n} 2\epsilon_i(R, \eta) - \tilde{u}(n, R, \eta), \tag{2}$$

where $n = Z/2$ particles for the proton level scheme. Then $\delta u = \delta u_p + \delta u_n$.

Similarly, for pairing corrections we take the doubly degenerate levels $\{\epsilon_i\}$ in units of $\hbar\omega_0^0$. $Z/2$ levels are occupied with n levels below and n' above Fermi energy contributing to pairing, $n = n' = \Omega\tilde{g}_s/2$. The cutoff energy, $\Omega \simeq 1 \gg \tilde{\Delta} = 12/\sqrt{A}\hbar\omega_0^0$. The gap Δ and Fermi energy λ are solutions of the

BCS system of two eqs. Compared to shell correction, the pairing correction is out of phase and smaller in amplitude, leading for $\eta =$ constant to a smoother total curve $\delta e(R) = \delta u(R) + \delta p(R)$ where $\delta p = \delta p_p + \delta p_n$.

The potential energy surfaces (PES) of ^{222}Ra versus the normalized separation distance ξ and the mass asymmetry η are plotted in Fig. 1 (the microscopic shell plus pairing corrections at the bottom and the total deformation energy at the top). Three valleys around $\eta \simeq 0.87$; 0.36 and 0.1 can be seen at the top of Fig. 1. We count the number of valleys for $\eta \geq 0$ because the mirror $\eta \leq 0$ gives the same number for complimentary heavy fragments becoming light ones and vice-versa. I have used such valleys produced by the magic numbers of the fragments in the sixtieth to motivate the search for superheavy elements, and the development of Heavy Ion Physics worldwide and in Germany, where GSI was built. These valleys may be also seen on the total PES at the top of Fig. 1. Here the deepest valley remains that at a small value of η not far from the minimum of the macroscopic Y+EM energy at $\eta = 0$, which is responsible for the cold fission. At a large value of η, the ^{208}Pb + ^{14}C valley, favouring the ^{14}C radioactivity of ^{222}Ra, is laying on the Businaro-Gallone mountain, hence it is shallower despite the fact that it is very pronounced in the shell correction surface.

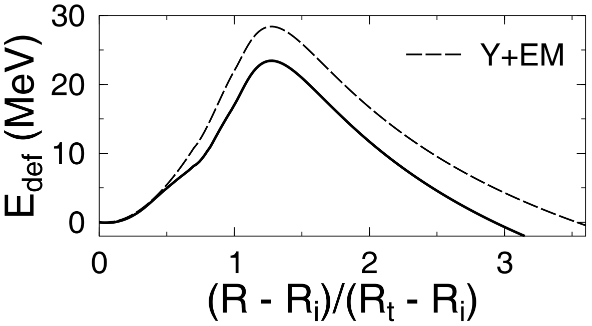

Fig. 2 – Potential barrier for ^{14}C radioactivity of ^{222}Ra obtained with the macroscopic-microscopic method. The barrier height and width are smaller compared to Yukawa-plus-exponential model (Y+EM).

A plot obtained by cutting the total PES and that of Y+EM at a given value of the asymmetry parameter is shown in Fig. 2 for ^{14}C emission from ^{222}Ra. It provides a justification for one of the basic assumption of the ASAF model, which was very successful in predicting the half-lives of cluster decay modes. It is remarkable to see for a cluster emitter a potential barrier obtained by using the macroscopic-microscopic method. Having a smaller height and width compared to the (dotted line) macroscopic Y+EM barrier, it is very similar to the barrier used in ASAF which was lower and narrower than the Myers-Swiatecki's [31] LDM barrier.

3. EXPERIMENTAL CONFIRMATIONS OF PREDICTED HALF-LIVES

The main region of emitters experimentally observed is above $Z = 86$ with daughters around ^{208}Pb. The major experimental difficulty is related to the "exotic" characteristics of heavy particle radioactivities due to the strong competition of α-decay: one has to register few rare events in a large background of many α particles. The largest branching ratio with respect to α-decay, $b = T_\alpha/T$, is about $10^{-9.2}$ and the smallest measured until now is $10^{-16.1}$.

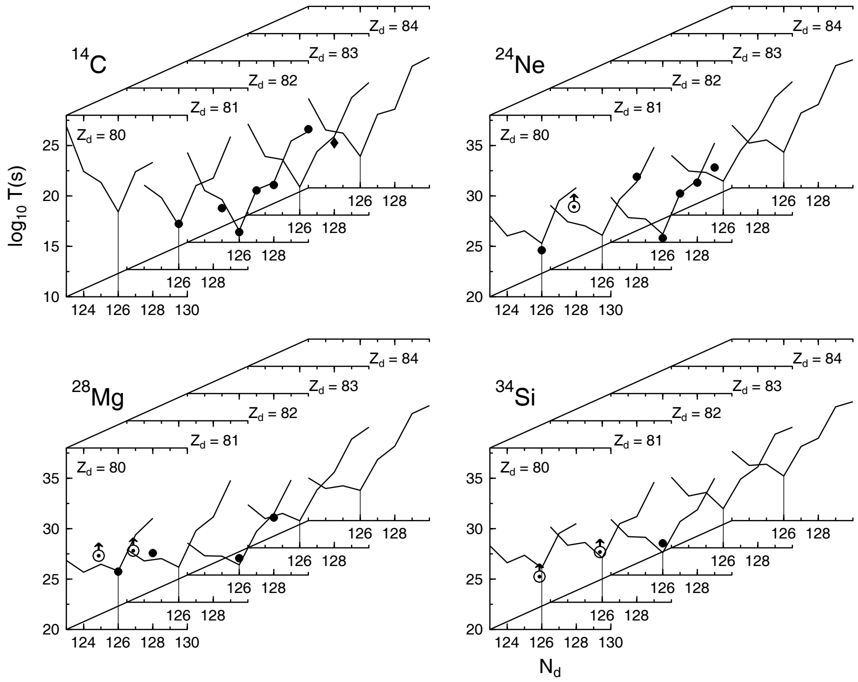

Fig. 3 – Predicted half-lives within ASAF model (lines) and measurements (points) for four kinds of cluster decay modes versus proton (Z_d) and neutron (N_d) number of the daughter nucleus. The shell effects are clearly seen as minima of life-time for magic numbers of the daughter $Z_d = 82$, $N_d = 126$.

From many decay modes with half-lives and branching ratios relative to α-decay predicted within the ASAF model, the following have been experimentally confirmed: ^{14}C, ^{20}O, ^{23}F, $^{24-26}$Ne, ^{28,30}Mg, ^{32,34}Si. The experimental data are in good agreement with predicted values (see the examples given in Fig. 3 for ^{14}C, ^{24}Ne, ^{28}Mg, and ^{34}Si decay modes). A strong shell effect can be seen: as a rule the shortest value of the half-life (maximum of $1/T$) is obtained when the daughter nucleus has a magic number of neutrons ($N_d = 126$) and/or protons ($Z_d = 82$).

By comparing the systematics of calculated values and of experimental data one can obtain many [3] other possible candidates for future experiments. Some of the possible candidates for future experiments are: ^{220,222,223}Fr, ^{223,224}Ac, and ^{225}Th as ^{14}C emitters; ^{229}Th for ^{20}O radioactivity; ^{229}Pa for ^{22}Ne decay mode; ^{230,232}Pa, ^{231}U, and ^{233}Np for ^{24}Ne radioactivity; ^{234}Pu for ^{26}Mg decay mode; ^{234,235}Np and ^{235,237}Pu as ^{28}Mg emitters; ^{238,239}Am and $^{239-241}$Cm for ^{32}Si radioactivity; ^{33}Si decay of ^{241}Cm; ^{238,239}Am and $^{239-241}$Cm for ^{32}Si radioactivity; ^{33}Si decay of ^{241}Cm; ^{238,239}Am and $^{239-241}$Cm for ^{32}Si radioactivity; ^{33}Si decay of ^{241}Cm; ^{238,239}Am and $^{239-241}$Cm for ^{32}Si radioactivity, and ^{33}Si decay of ^{241}Cm.

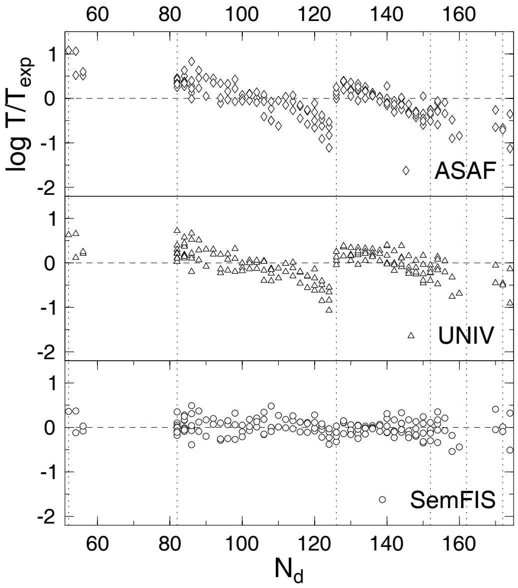

Fig. 4 – The deviations of calculated values of half-lives against α-deca from the experimental ones for 136 even-even nuclei. Three kinds of fission mode (analytical superasymmetric fission model (ASAF), universal curve (UNIV), and the semiempirical formula based on fission theory (SemFIS)) are compared.

^{14}C radioactivity may be used for spectroscopical purposes [32, 33] as it was observed when the fine structure of the ^{14}C radioactivity of ^{223}Ra, for the first time discussed by Martin Greiner and Werner Scheid [34], was discovered in an experiment with superconducting SOLENO spectrometer performed at Orsay, France [35]. The experiment was repeated in 1995 with a better resolution [32]. Surprisingly, the transition to the first excited state of the daughter ^{209}Pb was stronger than that to the ground state, despite a smaller Q-value by 0.779 MeV. A transition with an excited state of ^{14}C predicted [34] in 1986 was not observed.

4. ALPHA-DECAY OF SUPERHEAVY NUCLEI

From the liquid drop model point of view the superheavy nuclei would never exist because there would be no potential barrier against spontaneous fission. When a shell correction energy is added, a potential barrier shows up stabilizing superheavy nuclei [36, 37, 38, 39]. 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 [40, 41, 42], which are mostly proton-rich nuclei.

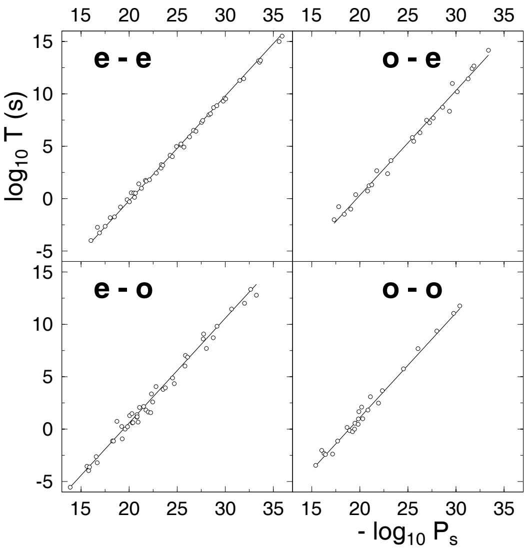

Fig. 5 – Universal curves for α-decay of transuranium nuclei in four groups of even-even, even-odd, odd-even and odd-odd nuclei. P_s is the calculated penetrability of the external Coulomb barrier.

Three relationships based on fission theory of α-decay (the ASAF model, the universal curve (UNIV) [43, 44], and the semiempirical formula (SemFIS) taking into account the magic numbers of nucleons) have been used to calculate the half-lives for α-decay of transuranian nuclei including superheavies [25, 20]. The standard deviations

$$\sigma = \left\{ \sum_{i=1}^{n} [\log(T_i/T_{exp})]^2/(n-1) \right\}^{1/2}, \tag{3}$$

from experimental values of 47 even-even nuclei was 0.402, for 45 e-o was 0.615, for 25 o-e was 0.761, and for 25 o-o $\sigma = 0.795$ within ASAF model, and 0.267, 0.554, 0.543, and 0.456, respectively, when the universal curve was used.

We used several sources for α-decay of transuranium heavy and superheavy nuclei [45, 46, 47, 48, 49, 50, 8].

The deviations of calculated values of half-lives against α-decay from the experimental ones for 136 even-even nuclei are presented in figure 4. By choosing the neutron number of the daughter nucleus in the abscisa one can see the shell effects at the following spherical and deformed magic numbers of neutrons: 50, 82, 126, 152, 162, and 172. The increased errors in the neighbourhood of $N = 126$, present in all other cases, are smoothed out by SemFIS formula.

For transuranium nuclei including superheavies, the results presented in Fig. 5 are showing good agreement between the experimental points and the calculated universal curves (lines), not only for even-even nuclei but also for even-odd, odd-even and odd-odd.

In conclusion, the fission approach of cluster radioactivities and α-decay, not only gave a new insight into the physics of nuclear decay modes, leading to our prediction of new kinds of spontaneous disintegrations, but also provided an excellent tool for calculation of the half-lives.

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