



## Cluster Radioactivity

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### ABSTRACT

Cluster radioactivity (spontaneous emission of heavy particles from nuclei) is presented from a theoretical point of view in good agreement with experimental results. After a brief historical account, we give details about the analytical superasymmetric fission (ASAF) model extensively used for predicting the half-lives of heavy and superheavy ( $Z \geq 104$ ) elements. For the already measured 26 cluster decays (from  $^{14}\text{C}$  to  $^{32,34}\text{Si}$  of parent nuclides with  $Z = 87-96$ ) it is clear that cluster radioactivity is a rare phenomenon in the best case about 9 orders of magnitude weaker than the competing alpha decay. Then we show the theoretical possibility of a strong cluster decay compared to alpha decay for some superheavy nuclei with  $Z \geq 122$ , e.g.  $^{306}122$ ;  $^{310-314}122$ ;  $^{306-324}124$ , and  $^{311-323}124$ .

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## 1. Introduction

The spontaneous emission of heavy particle from nuclei Cluster Radioactivity (CR) was predicted in 1980 [1], when Aureliu Săndulescu (1932-2019) was Vice Director of the Joint Institute for Nuclear Research (J.I.N.R.) in Dubna, near Moscow, Soviet Union, and Prof. Dr. H.C. mult. Walter Greiner was Director of the Institut für Theoretische Physik der J.W. Goethe University, Frankfurt am Main, Germany.

The first experimental confirmation was reported from Oxford University in 1984 [2]; 11 events of  $^{14}\text{C}$  emitted from  $^{223}\text{Ra}$  have been obtained in a long run of about six months; they have used the standard technique of identification with semiconductor telescope detectors. With a magnetic spectrometer SOLENO at Orsay [3-5] and an Enge-split pole spectrometer at Argonne National Laboratory [6-7] a similar result was seen in a few hours because a much stronger source could be used. An alternative detection technique was based on solid state nuclear track detectors. The difficulty of experiment was due to the very low intensity (about nine orders of magnitude) of  $^{14}\text{C}$  in a strong background of alpha particles. The shortest measured

half-life of  $T_c = 10^{11.01}$  s corresponds to  $^{14}\text{C}$  radioactivity of  $^{222}\text{Ra}$  and the largest branching ratio relative to  $\alpha$ -decay,  $b_\alpha = T_\alpha/T_c$ , of  $10^{-8.9}$  was observed for  $^{14}\text{C}$  radioactivity of  $^{223}\text{Ra}$ .

In the first three publications [3, 6, 8] the predictions [1] have not been mentioned. The situation changed after publication of a comment [8]; shortly after the scientific community acknowledged the predictions and the article [1] is now one of the most cited in the field.

One of the greatest honour is a short presentation in Encyclopaedia Britannica [9] of the predictions. There are only 4 romanian scientists and 2 born in Romania: 1. N.C. Paulescu (1869-1931) who conducted groundbreaking research on the antidiabetic hormone insulin; 2. Henri Marie Coandă (1886-1972) Romanian inventor, aerodynamics pioneer, and builder of an experimental aircraft. He discovered the Coandă effect of fluid dynamics; [3-4]. Aureliu Săndulescu (1932-2019) and Dorin N Poenaru (1936-) Romanian theoretical physicists. Two romanian born: George E. PALADE (1912-2008) (Nobel Pr. for Medicine 1974, SUA) for discoveries concerning the functional organization of the cell that were seminal events in the development of modern cell biology, and Stefan W. HELL (1962- ) (Nobel Pr. for Chemistry 2014, Germany) for an ultrasensitive microscope.

In the article published in Phys. Rev. C 92 (2015) 064301 by Y.Z. Wang et al. [10] it is shown that from 18 formulae used to calculate  $\alpha$ -decay half-lives the best results are obtained with semFIS2 and UNIV2 developed by D.N. Poenaru et al. We continued to be active, e.g. [15-25].

The following types of radioactivities have been experimentally confirmed worldwide (Oxford, Moscow, Orsay, Berkeley, Geneva, Dubna, Argonne, Milano, Viena, Lanzhou, Beijing and Livermore) [26]:  $^{14}\text{C}$ ,  $^{20}\text{O}$ ,  $^{23}\text{F}$ ,  $^{22, 24-26}\text{Ne}$ ,  $^{28, 30}\text{Mg}$  and  $^{32, 34}\text{Si}$  from parent nuclides with  $Z = 87 - 96$ . There are about 26 different successful experiments. The measured half-lives are in good agreement with theoretical predictions within the analytical superasymmetric fission (ASAF) model developed by D. N. Poenaru et al. Among the most active experimentalists we should mention: Svetlana Tretyakova [27], P. Buford Price [28]; Eid Hourany and Michel Hussonnois [4, 29]; Roberto Bonetti [26], A. Ogloblin [30] and others. Among the books there are [31-35]; chapters in books: [36-41]. Many theoretical works have been performed by other colleagues, e.g. [42-46].

In the following we shall briefly present the ASAF model, as well as our predictions concerning the possibility of a cluster radioactivity stronger than alpha decay for some superheavy nuclei with atomic numbers  $Z \geq 121$  [48].

## 2. The ASAF Model

In a systematic search for cluster radioactivity one has to investigate about 2000 nuclides (with known experimental masses) against spontaneous emission of 200 isotopes of the elements with  $Z_e = 2-28$ , i.e. almost  $10^5$  combinations. We could do the calculations in a reasonable time by using an analytical relationship for the half-life. The analytical superasymmetric (ASAF) model was developed in order to meet this goal. The starting point was the Myers-Swiatecki liquid drop model [49] with a phenomenological correction in the spirit of Strutinsky method [50].

Initially we published [51-57]. We give some informations concerning the ASAF model, see e.g. [58]. More details can be found in the e-print [59]. The half-lives is expressed in decimal logarithm of the values in seconds,  $T = \log_{10} T_{1/2}(s)$ .

The half-life of a parent nucleus  $AZ$  against the split into a cluster  $A_c, Z_c$  and a daughter  $A_d, Z_d$  is calculated by using the WKB quasiclassical approximation; the action integral is expressed as [60]

$$T = [(h \ln 2) / (2E_v)] \exp(K_{ov} + K_{si}) \quad (1)$$

$$K = \frac{2}{\hbar} \int_{R_a}^{R_b} \sqrt{2B(R)[E(R) - Q]} dR \quad (2)$$

with  $B = \mu$  — the reduced mass,  $K = K_{ov} + K_s$  (overlapping and separated fragments), and  $E(R)$  is the total deformation energy.  $R_a, R_b$  are the turning points, defined by  $E(R_a) - Q = E(R_b) - Q = 0$ .

For ASAF we replace in eq. 2  $E(R) - Q$  by  $[E(R) - E_{corr} - Q]$ .  $E_{corr}$  is a correction energy similar to the Strutinsky shell correction [50]. In order to get a smaller number of parameters we took  $E_v = E_{corr}$ . The turning points of the WKB integral are:  $R_a = R_i + (R_t - R_i) \left[ (E_v + E^*) / E_b^0 \right]^{1/2}$ ;  $R_b = R_t E_c \left\{ 1/2 + \left[ 1/4 + (Q + E_v + E^*) E_t / E_c^2 \right]^{1/2} \right\} / (Q + E_v + E^*)$  where  $E^*$  is the excitation energy concentrated in the separation degree of freedom.  $R_i = R_0 - R_c$  is the initial separation distance,  $R_t = R_c + R_d$  is the touching point separation distance,  $R_j = r_0 A_j^{1/3}$  ( $j = 0, e, d$ ;  $r_0 = 1.2249$  fm) are the radii of parent, emitted and daughter nuclei, and  $E_b^0 = E_i - Q$  is the barrier height before correction.

The two terms of the action integral corresponding to the overlapping ( $K_{ov}$ ) and separated ( $K_s$ ) fragments, are calculated by analytical formulas (approximated for  $K_{ov}$  and exact for  $K_s$  in case of separated spherical shapes within the LDM (Liquid Drop Model [49])). Since 1984, the ASAF model results have been used to guide the experiments and to stimulate other theoretical works.

$$K_{ov} = 0.2196 (E_b^0 A_e A_d / A)^{1/2} (R_t - R_i) \times \left[ \sqrt{1 - b^2} - b^2 \ln \frac{1 + \sqrt{1 - b^2}}{b} \right] \quad (3)$$

$$K_s = 0.4392 \left[ (Q + E_v + E^*) A_e A_d / A \right]^{1/2} R_b J_{rc}; \quad (4)$$

$$b^2 = (E_v + E^*) / E_b^0$$

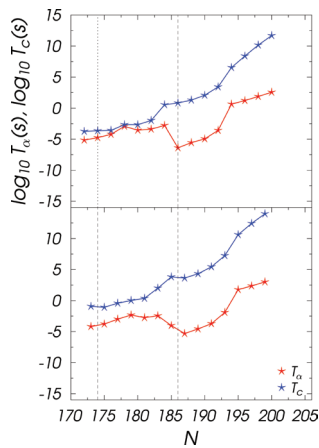
$$J_{rc} = (c) \arccos \sqrt{(1 - c + r) / (2 - c)} - [(1 - r)(1 - c + r)]^{1/2} + \sqrt{1 - c} \ln \left[ \frac{2\sqrt{(1 - c)(1 - r)(1 - c + r)} + 2 - 2c + cr}{r(2 - c)} \right] \quad (5)$$

where  $r = R_t / R_b$  and  $c = r E_c / (Q + E_v + E^*)$ . When there is no centrifugal contribution ( $l = 0$ ), one has  $c = 1$ .

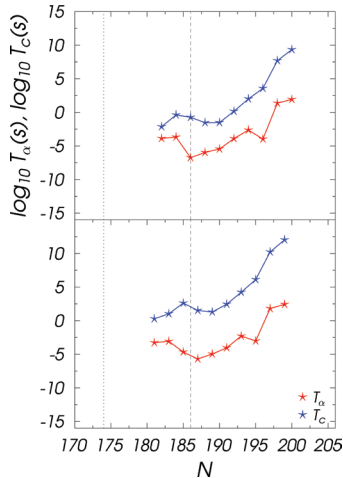
The potential barrier shape similar to that we considered within the ASAF model was recently calculated by using the macroscopic-microscopic method [61], as a cut through the PES at a given mass asymmetry, usually the  $^{208}\text{Pb}$  valley or not far from it.

For even-even parent nuclei, one has

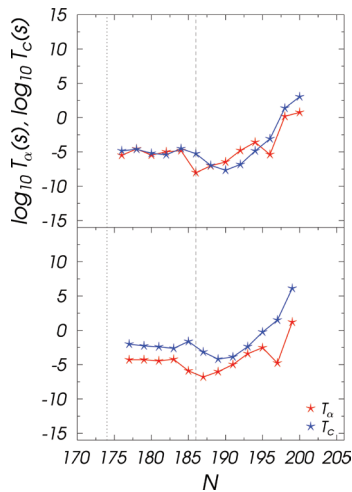
$$\log T = -\log P_s + c_{ee} \quad (6)$$



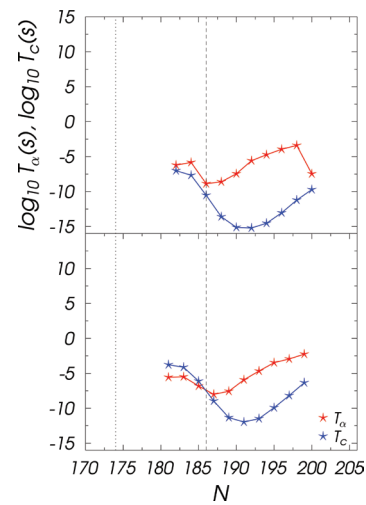
**Figure 1:** Comparison of the  $\log T_{\alpha}(s)$  and  $\log T_c(s)$  for  $Z = 120$ .  $N$  is the neutron number of the parent nucleus.



**Figure 2:** Comparison of the  $\log T_{\alpha}(s)$  and  $\log T_c(s)$  for  $Z = 121$ .  $N$  is the neutron number of the parent nucleus.



**Figure 3:** Comparison of the  $\log T_{\alpha}(s)$  and  $\log T_c(s)$  for  $Z = 122$ .  $N$  is the neutron number of the parent nucleus.



**Figure 4:** Comparison of the  $\log T_{\alpha}(s)$  and  $\log T_c(s)$  for  $Z = 124$ .  $N$  is the neutron number of the parent nucleus.

where  $c_{cc}$  may be used as a parameter which can be determined using a given set of  $n$  experimental data on cluster radioactivity; the optimum value of  $c_{cc}$  is the one which gives the minimum value of the standard RMS deviation  $\sigma$ :

$$\sigma = \left\{ \sum_{i=1}^n [\log(T_i / T_{\text{exp}})]^2 / (n-1) \right\}^{1/2} \quad (7)$$

The larger  $n$  would be the better choice.

We estimated the half-lives for more than 150 decay modes, including all cases experimentally confirmed until now [62, 63].

A very important quantity in calculations is the released energy,  $Q$ :

$$Q = [M - (M_e + M_d)]c^2 \quad (8)$$

obtained as a difference between the parent,  $M$ , and the two decay product masses,  $M_e$  and  $M_d$  in units of energy;  $c$  is the light velocity. Besides the existing measured masses [11] (used whenever available), we also considered the theoretical Wp4 [10, 12, 13] and Koura-Tachibana-Ueno-Yamada (KTUY05) [14] models.

### 3. Cluster Radioactivity of the Heaviest Superheavies

The measurements have shown that very frequently the daughter nucleus was the doubly magic  $^{208}_{82}\text{Pb}_{126}$  or one of its neighbors. We extended the study of cluster decays to very heavy superheavy nuclei ( $Z \geq 110$ ) in order to study emitted clusters with  $Z_c > 28$  from parents with  $Z > 110$  and daughter around  $^{208}\text{Pb}$  [17, 48]. We considered emitted clusters with

$$Z_c^{\max} = Z - 82 \quad (9)$$

In very difficult experiments with cold fusion reactions [64, 65], or hot fusion induced by  $^{48}\text{Ca}$  projectiles [66, 67], superheavy elements with atomic numbers  $Z = 104 - 118$  have been synthesized. The identification method was mostly based on  $\alpha$ -decay chains. Theoretical works may be exemplified by [32, 39, 68-74].

We present in Figures 1-4 variation of the  $\log T_\alpha$ (s) and  $\log T_c$ (s) with the neutron number of the parent nucleus. In the upper part of every figure there are the even N components; at the bottom - the odd ones.

While for both Figures 1 and 2 one has always the alpha decay as the dominant mode  $T_\alpha < T_c$ , for even N components in Figure 3 there are many cases with  $T_\alpha > T_c$ . Even better situation can be seen in Figure 4 where we got  $T_\alpha > T_c$  for many parent nuclides both with even- and odd- N.

## Conclusion

In conclusion, for  $Z \geq 122$  we predict many examples of superheavy nuclei in which cluster radioactivity could be the dominant decay mode:  $T_c \leq T_\alpha$ , e.g.:  $^{306}_{122}\text{Z}$ ;  $^{310-314}_{122}\text{Z}$ ;  $^{306-324}_{124}\text{Z}$ , and  $^{311-323}_{124}\text{Z}$ .

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