Heavy particle radioactivities of superheavy nuclei

D. N. Poenaru,[1, 2, *] R. A. Gherghescu,[1, 2] and W. Greiner[1]

[1] *Frankfurt Institute for Advanced Studies (FIAS),*
Ruth-Moufang-Str. 1, 60438 Frankfurt am Main, Germany
[2] *Horia Hulubei National Institute of Physics and Nuclear Engineering (IFIN-HH),*
P.O. Box MG-6, RO-077125 Bucharest-Magurele, Romania

(Dated:)

The concept of heavy particle radioactivity (HPR) is changed to allow emitted particles with $Z_e > 28$ from parents with $Z > 110$ and daughter around ^{208}Pb. Calculations for superheavy (SH) nuclei with Z=104-124 are showing a trend toward shorter half-lives and larger branching ratio relative to α decay for heavier SHs. It is possible to find regions in which HPR is stronger than alpha decay. The new mass table AME11 and the theoretical KTUY05 and FRDM95 masses are used to determine the released energy. For 124 we found isotopes with half-lives in the range of ns to ps.

PACS numbers: 23.70.+j, 23.60.+e, 21.10.Tg

During the last years the heaviest elements with atomic numbers up to $Z = 118$ have been synthesised [1] either with cold fusion reactions having the ^{208}Pb or ^{209}Bi target [2, 3] or with hot fusion induced by ^{28}Ca projectiles [4, 5]. Attempts to produce $Z = 120$ are reported [6] and new experiments are presently running at GSI Darmstadt [7]. The main experimental difficulty in identifying the new superheavy (SH) elements is the low probability of their formation, and the separation of the short lived compound nucleus from the very high flux of incident projectile nuclei. The lowest cross-section of 55 fb was measured at RIKEN [3] where one decay chain of ^{278}113 was observed during 79 days with beam of ^{70}Zn on ^{209}Bi target. After naming copernicium, Cn, $Z = 112$ suggested by GSI scientists, IUPAC recommends that the Dubna-Livermore collaboration be credited with discovery of new elements 114 and 116.

It is generally agreed that the term SH element, introduced [8] in 1958, is a synonym for elements which exist solely due to their nuclear shell effects. The lightest SH is $Z = 104$ Rf with half-lives of different isotopes around one minute. This is 16 orders of magnitude longer than the expected nuclear lifetime of 10^{-14} s these isotopes would survive without any shell stabilisation. 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 proton-rich nuclides. Extensive calculations of fission barriers and half-lives have been published [9].

Despite the important experimental and theoretical development there are still several unanswered questions related to the magic numbers, production cross sections, and decay modes. Besides beta decay, only alpha decay and spontaneous fission of SH nuclei have been experimentally observed up to now. We would like to take also into account heavy particle radioactivities (HPR) [10, 11].

Since 1984 [12], the following HPR have been experimentally confirmed [13] in heavy parent nuclei with $Z = 87 - 96$: ^{14}C, ^{20}O, ^{23}F, $^{22,24-26}$Ne, ^{28,30}Mg, ^{32,34}Si with half-lives in good agreement with predicted values within analytical superasymmetric fission (ASAF) model (see the review [14] and references therein). Almost always the corresponding daughter nucleus was the doubly magic $^{208}_{82}$Pb$_{126}$ or one of its neighbours. The newest measurement of ^{14}C decay of ^{223}Ac [15] was one of the possible candidates for future experiments mentioned in the systematics [16] showing that the strong shell effect due to magic number of neutrons, $N_d = 126$, and protons, $Z_d = 82$, present in order to lead to shorter half lives was not entirely exploited.

The shortest half-life of $T_c = 10^{11.01}$ s corresponds to ^{14}C radioactivity of ^{222}Ra and the largest branching ratio relative to alpha decay $b_\alpha = T_\alpha/T_c = 10^{-8.9}$ was measured for the ^{14}C radioactivity of ^{223}Ra. Consequently HPR in the region of transfrancium nuclei is a rare phenomenon in a strong background of α particles. Several attempts to detect ^{12}C radioactivity of the neutrondeficient ^{114}Ba with a daughter in the neighbourhood of the double magic $^{100}_{50}$Sn$_{50}$, predicted to have a larger b_α, have failed.

In order to check the possibility of extrapolations from $A_e = 14 - 34$ emitted clusters already measured in the region of emitters with $Z = 87 - 96$ to SHs up to 124, where one may find an emitted particle as heavy as ^{114}Mo, we estimated within ASAF model the half-life for ^{128}Sn emission from ^{256}Fm ($Q = 252.129$ MeV) and for ^{130}Te emission from ^{262}Rf ($Q = 274.926$ MeV): $\log_{10} T_{Fm}(s) = 4.88$ and $\log_{10} T_{Rf}(s) = 0.53$. They are in agreement with experimental values for spontaneous fission [17]: 4.02 and 0.32, respectively.

There are many other theoretical approaches of the HPR e.g. Refs. [18–21]. They are based on the quantum mechanical tunnelling process relationship of the disintegration constant λ, valid in both fission-like or α-like theories

$$\lambda = \ln 2/T = \nu S P_s \tag{1}$$

where T is the half life and ν, S, P_s are three model-dependent quantities: ν is the frequency of assaults on the barrier per second, S is the preformation probability of the cluster at the nuclear surface (equal to the penetrability of the internal part of the barrier in a fission theory), and P_s is the quantum penetrability of the external potential barrier. Alternatively, instead of ν one may use the zero point vibration energy $E_v = h\nu/2$ in which h is the Plank constant.

We should change the concept of HPR, previously [23] associated to a maximum $Z_e^{max} = 28$, allowing to preserve its main characteristics in the regions of SH with $Z > 110$ i.e. in a systematic search for HPR we shall consider not only the emitted particles with atomic numbers $2 < Z_e < 29$, as in previous calculations, but also heavier ones up to $Z_e^{max} = Z - 82$, allowing to get for $Z > 110$ an atomic number of the most probable emitted HP $Z_e > 28$ and a doubly magic daughter around ^{208}Pb.

Calculations are performed within ASAF model, very useful for the high number of combinations parent – emitted cluster (more than 10^5) in order to check the metastability of more than 2000 parent nuclides with measured masses against many possible decay modes. We started with Myers-Swiatecki liquid drop model (LDM) [24] adjusted with a phenomenological correction accounting for the known overestimation of the barrier height and for the shell and pairing effects in the spirit of Strutinsky method.

The half-life of a parent nucleus AZ against the split into a HP or an emitted cluster A_eZ_e and a daughter A_dZ_d is given by

$$T = [(h \ln 2)/(2E_v)] exp(K_{ov} + K_s) \tag{2}$$

and is calculated by using the Wentzel–Kramers–Brillouin (WKB) quasiclassical approximation, according to which the action integral is expressed as

$$K = \frac{2}{\hbar} \int_{R_a}^{R_b} \sqrt{2B(R)E(R)} dR \tag{3}$$

with $B = \mu$ the reduced mass, $K = K_{ov} + K_s$, and the $E(R)$ potential energy replaced by $[E(R) - E_{corr}] - Q$. E_{corr} is a correction energy similar to the Strutinsky [25] shell correction, also taking into account the fact that LDM overestimates fission barrier heights, and the effective inertia in the overlapping region is different from the reduced mass. R_a and R_b are the turning points of the WKB integral. The two terms of the action integral K, corresponding to the overlapping (K_{ov}) and separated (K_s) fragments, are calculated by analytical formulae [14].

Half-lives calculations are very sensitive to the released energy (Q value) obtained as a difference of the parent and the two decay product masses

$$Q = M - (M_e + M_d) \tag{4}$$

in units of energy. Even with the newly released tables of experimental masses, AME11 [22] as a preview for the AME13 publication, many masses are still not available for new SH, hence we shall use not only these updated tables for 3290 nuclides (2377 measured and 913 from the systematics) ending up at $Z = 118$ but also some calculated masses e.g. KTUY05 [26] and FRDM95 [27] with 9441 and 8979 masses, respectively.

In a systematic search for HPR we calculate with the ASAF model for every parent nucleus ^{A}Z the half-lives of all combinations of pairs of fragments $^{A_e}Z_e$, $^{A_d}Z_d$ with $2 < Z_d \leq Z_e^{max}$ conserving the hadron numbers $Z_e + Z_d = Z$ and $A_e + A_d = A$. Let us start with the results obtained by using the AME11 mass tables. An example of the time spectra obtained for different clusters emitted from the parent nuclei ^{222}Ra and ^{288}114 is shown in figure 1 versus the mass numbers of the light fragment. The symbols of the emitted HPR are given on the figure's legend.

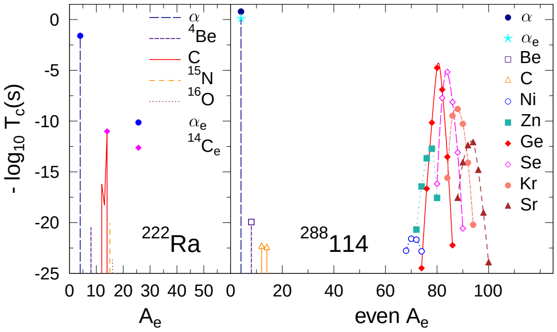

FIG. 1. (Color online) Time spectra of different cluster emissions from ^{222}Ra (left panel) and from the superheavy nucleus ^{288}114 (right panel). The most probable emitted clusters from ^{222}Ra and ^{288}114 are ^{14}C and ^{80}Ge, respectively, both leading to ^{208}Pb daughter nucleus.

From the left panel of this figure one can see that the shortest half-lives of ^{222}Ra correspond to α-decay and $^{14}_{6}C_8$ radioactivity, respectively. Both these decay modes have been experimentally observed and there is a good agreement between the calculated values and measured data. Other HPR with half-lives $T_c < 10^{25}$ s are: ^{8}Be; ^{12,13}C; ^{15}N and ^{16}O — all with much longer half-lives.

Similarly, on the right hand side of the figure we show calculated results for the SH nucleus ^{288}114. Again α decay is the strongest decay mode and there is a good agreement between our calculations and the experimentally observed half-life. The time spectrum in the region of mass numbers of emitted particles around $A_e = 80$ is more complex looking similar to a fission fragment spectrum. There are many HPR with $Z_e = 28 - 38$ having

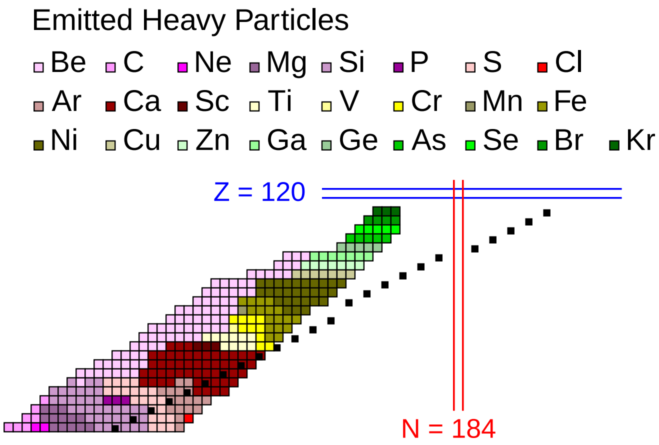

FIG. 2. (Color online) Chart of heavy and superheavy cluster emitters with atomic numbers $Z = 94 - 118$. The $Q-$values are calculated using the AME11 mass tables [22]. Black squares mark the Green approximation of the line of beta-stability. One most probable emitted cluster is given for every parent nucleus.

$T_c < 10^{25}$ s. For the sake of clarity we only plotted the results corresponding to even-even emitted HP which are leading to shorter half-lives in the same way the ^{13}C radioactivity of ^{222}Ra is less probable than both ^{14}C and ^{12}C spontaneous emissions. The most probable emitted HP from ^{288}114 is $^{80}_{32}$Ge$_{48}$ with a calculated branching ratio $b_\alpha = 10^{-5.01}$. One should also take into account a competition of $^{84}_{34}$Se$_{50}$ with a magic number of neutrons $N_e = 50$ and a branching ratio $b_\alpha = 10^{-5.42}$.

We proceed in a similar way with all parent nuclei with $Z = 94 - 118$ present on the AME11 mass table. The chart of cluster emitters from figure 2 is obtained by associating to each parent only the most probable emitted cluster. The black squares mark the Green approximation of the line of beta-stability. All superheavy nuclei present on the AME11 mass table are proton-rich nuclides with neutron numbers smaller than N_β on the line of beta stability. The experimentally determined $^{28}_{12}$Mg radioactivity of $^{236}_{94}$Pu, $^{32}_{14}$Si radioactivity of $^{238}_{94}$Pu, and $^{34}_{14}$Si radioactivity of $^{242}_{96}$Cm are fairly well reproduced.

New many types of HPR with $Z_e > 28$ may be seen on this chart: Cu, Zn, Ga, Ge, As, Se, Br and Kr. We used only one color for a given Z_e despite the fact that as the result of the calculations we obtained several isotopes of these elements, e.g. $A_e = 26, 28$ for Mg; $30, 32, 33, 34$ for Si; $36, 38, 40, 41, 42$ for S; $44, 46, 47, 48$ for Ar; $48, 49, 50, 51, 52$ for Ca; $50, 51, 52$ for Sc; $53, 54, 55, 56$ for Ti; $57, 58, 59, 60, 61$ for Cr; $60, 62, 63, 64, 66$ for Fe; $66, 68, 69, 70, 71, 72, 73$ for Ni; $69, 71, 72, 73, 74, 75$ for Cu; $72, 74, 76, 77$ for Zn; $75, 77, 78, 79$ for Ga; $78, 80, 81$ for Ge, $81, 83$ for As; $82, 84, 85$ for Se; $85, 86$ for Br, and $86, 87$ for Kr. Only one mass value was obtained for the most probable emitted particles Be, C, Ne, P, Cl, V, and Mn.

As we previously observed [23], many of the proton-rich SH nuclides are ^{8}Be emitters, but they have a very low branching ratio b_α. The general trend of a shorter half-life and a larger branching ratio when the atomic and mass numbers of the parent nucleus increases may be seen on the left hand side of the figures 3 and 4, obtained within ASAF model by using the AME11 mass tables to calculate the Q-values.

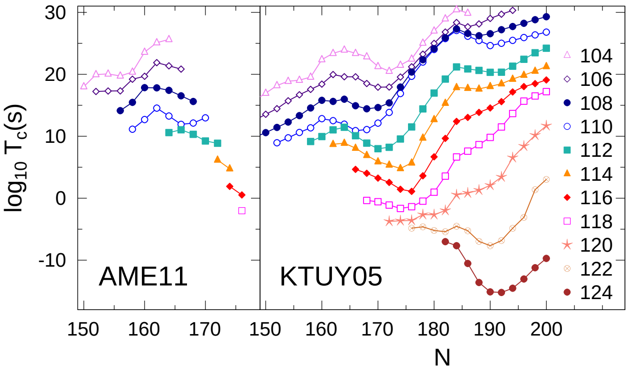

FIG. 3. (Color online) Decimal logarithm of the half-lives of superheavy nuclei against cluster radioactivities versus the neutron number of the parent nucleus. $Q-$values are calculated using the AME11 experimental mass tables [22] (left panel) and the KTUY05 [26] calculations.

One can advance toward neutron-rich nuclei by using the KTUY05 calculated mass tables, as shown in the right panels of these figures. When using KTUY05 and FRDM95 masses for parent and daughter nuclei we take into account the nuclides stable against one proton, two protons, one neutron and two neutrons spontaneous emissions. If the calculated masses are reliable, then half-lives T_c in the range of nanoseconds to picoseconds for SH nuclei with $Z = 124$ (see the right hand side of figure 3) would make difficult or even impossible any identifica-

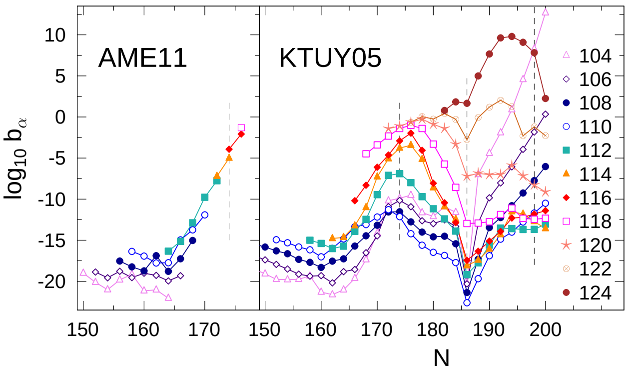

FIG. 4. (Color online) Decimal logarithm of the branching ratio relative to α decay for cluster emission from superheavy nuclei versus the neutron number of the parent nucleus. Vertical dashed lines correspond to $N = 174, 186, 198$.

tion measurement. More interesting for future experiments could be some even-even proton-rich isotopes of the 122 element with $N = 188 - 194$ for which the neutron number of the Green approximation of the line of beta stability is $N_\beta = 202$.

The pronounced minimum of the branching ratio at $N = 186$ in figure 4 is the result of the strong shell effect of the assumed magic number of neutrons $N = 184$ present in the KTUY05 masses. The half-life of α decay of a SH nucleus with $N = 186$ neutron number leading to a more stable daughter with magic neutron number $N_d = 184$ is shorter by some orders of magnitude compared to the α decay of a SH with $N = 184$. Calculated branching ratios $b_\alpha > 1$ for Rf ($Z = 104$) only occur in very neutron-rich nuclei with $N = 194-200$ compared to $N_\beta = 166$. Also their T_c half-life is extremely long. Similar results were obtained using the FRDM95 masses.

In conclusion, the concept of HPR should be changed to allow spontaneous emission of heavy particles with atomic number larger than 28 from SHs with $Z > 110$ and consequently daughter nuclei around the doubly magic ^{208}Pb. The calculated half-lives T_c against HPR and the branching ratios relative to α decay b_α are showing a trend toward shorter T_c and larger b_α for heavier SH nuclei which are not synthesised until now. If the KTUY05 and FRDM95 masses used to calculate the released energy Q are reliable, we expect to find for the element 124 many isotopes with half-lives in the range of nanoseconds to picoseconds, making practically impossible to perform any identification experiment. Nevertheless, there would be a chance to observe some proton-rich isotopes of 122 with branching ratios $b_\alpha > 1$.

We are looking forward to receive experimental information about the decay modes of SHs with $Z > 120$, hoping to confirm the present calculations. There is also a need for developing more refined decay models as well as new calculated mass tables and new mass measurements.

This work is partially supported by Deutsche Forschungsgemeinschaft Bonn and partially within the IDEI Programme under contracts with UEFISCSU, Bucharest.

---

* poenaru@fias.uni-frankfurt.de

[1] S. Hofmann, Physics **3**, 31 (2010).
[2] S. Hofmann and G. Münzenberg, Rev. Mod. Phys. **72**, 733 (2000).
[3] K. Morita *et al.*, J. Phys. Soc. Japan **73**, 2593 (2004).
[4] Y. T. Oganessian, J. Phys. G: Nucl. Part. Phys. **34**, R165 (2007).
[5] Y. T. Oganessian *et al.*, Phys. Rev. Lett. **104**, 142502 (2010).
[6] Y. T. Oganessian *et al.*, Phys. Rev. C **79**, 024603 (2009).
[7] S. Hofmann, private communication, 2011.
[8] F. G. Werner and J. A. Wheeler, Phys. Rev. **109**, 126 (1958).
[9] P. Möller *et al.*, Phys. Rev. C **79**, 064304 (2009).
[10] A. Sandulescu, D. N. Poenaru, and W. Greiner, Sov. J. Part. Nucl. **11**, 528 (1980).
[11] Encyclopaedia Britannica Online, 2011. Web http://www.britannica.com/EBchecked/topic/465998/.
[12] H. J. Rose and G. A. Jones, Nature **307**, 245 (1984).
[13] R. Bonetti and A. Guglielmetti, Rom. Rep. Phys. **59**, 301 (2007).
[14] D. N. Poenaru and W. Greiner, in *Clusters in Nuclei. Lecture Notes in Physics 1*, Ed. C. Beck (Springer, Berlin, 2010), Vol. 818, Chap. 1, pp. 1–56.
[15] A. Guglielmetti *et al.*, J. Phys.: Conf. Series **111**, 012050 (2008).
[16] D. N. Poenaru, Y. Nagame, R. A. Gherghescu, and W. Greiner, Phys. Rev. C **65**, 054308/1 (2002), erratum: C66.049902.
[17] D. C. Hoffman, T. M. Hamilton, M. R. Lane, Ch. 10 in *Nuclear Decay Modes*, (IOP Publishing, Bristol, 1996) pp. 393-432.
[18] R. Blendowske and H. Walliser, Phys. Rev. Lett. **61**, 1930 (1988).
[19] D. N. Poenaru and W. Greiner, Ch. 6 in *Nuclear Decay Modes*, (IOP Publishing, Bristol, 1996) pp. 275-336.
[20] R. G. Lovas, R. J. Liotta, A. Insolia, K. Varga and D. Delion, Phys. Rep. **294**, 265 (1998).
[21] C. Qi, F. R. Xu, R. J. Liotta, and R. Wyss, Phys. Rev. Lett. **103**, 072501 (2009).
[22] G. Audi and W. Meng, Private Communication, April 2011.
[23] D. N. Poenaru, D. Schnabel, W. Greiner, D. Mazilu and R. Gherghescu, Atomic Data Nucl. Data Tables **48**, 231 (1991).
[24] W. D. Myers and W. J. Swiatecki, Nucl. Phys., A **81**, 1 (1966).
[25] V. M. Strutinsky, Nucl. Phys., A **95**, 420 (1967).
[26] H. Koura, T. Tachibana, M. Uno and M. Yamada, Prog. Theor. Phys. **113**, 305 (2005).
[27] P. Möller, J. R. Nix, W. D. Myers, and W. J. Swiatecki, Atomic Data Nucl. Data Tables **59**, 185 (1995).