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E4-84-811

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ATOMIC NUCLEI DECAY MODES BY SPONTANEOUS EMISSION OF HEAVY IONS

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1984

1. INTRODUCTION

Toward the end of the 19^{th} century, H.Becquerel discovered the radiations emitted by some atomic nuclei. This phenomenon has been called by Marie and Pierre Curie-radioactivity. After some years of research it was established that there are three modes of nuclear decay: α, β, and γ. Now we know other modes of nuclear disintegration from the ground state: spontaneous fission (f); proton (p) and neutron (n) radioactivities, and there are predictions for the emission of two protons (2p) and two neutrons (2n). One should not forget also the β-delayed modes: α ; f ; n ; 2n; 3n; p ; 2p ; ^{3}H and the predicted ^{3}He. In these two-step processes a β-decaying precursor, far from the β-stability line[1], populates emitter excited levels, from which the quantum-mechanical tunneling goes faster. Details, including references, about historical development were given elsewhere[2].

In a series of previous papers[2-8] we have studied a new phenomenon intermediate between fission and α-decay, showing that a great variety of heavy ions (HI) with mass number $A_2 > 4$ and atomic number $Z_2 \geq 2$ could be emitted from heavy nuclei (A,Z) leading to the daughter (A_1, Z_1). Some of our predictions published since 1980, concerning these "unexpected"[9] novel decay modes, have been mentioned in a review paper[10].

At the beginning of 1984 Rose and Jones[11], from Oxford University, reported the discovery of the ^{14}C radioactivity of ^{223}Ra. This experiment, confirmed in Moscow[12] and Orsay[9], gives the first evidence for one of many kinds of the new decay modes.

Our starting point[3,4] was the examination of a typical (for example ^{238}U) fission fragment mass distribution curve[13] where we could notice two important things. First is the well known asymmetry, which was a longstanding puzzle of the theory. We know that it is essentially due to shell effects as it was explained[14-16] by using potential energy surfaces[17] computed with the Strutinsky[18,19] shell correction method. Alternatively the fragmentation theory[20], in which a Schrödinger equation in the asymmetry parameter $\eta = (A_1 - A_2)/A$ is solved, was successful in both regions of low[21] and high[3,22] mass asymmetry. Second: nothing was mentioned about the low mass region from $A_2 = 4$ to $A_2 \simeq 60$–70, except a shoulder[23] which corresponds to the new asymmetric peak computed in ref.[22].

By studying the nuclear stability against new superasymmetric fission modes, we have seen that in this only apparently empty region, one can explore a real new world of many kinds of exotic radioactivities. We have shown[3] on the basis of penetrability calculations, that some even-even nuclei like $^{12,14}C$,

^{24}Ne, ^{28}Mg , $^{32,34}Si$, ^{46}Ar , and ^{48}Ca , could be preferentially emitted from isotopes of Ra, U, Th, Pu, Cm, Cf, Fm, and No.

Unfortunately, the penetrability calculations, qualitatively useful to illustrate the possibility of finding such new decay modes and which have been used also in refs. [9,11,12], could not give the full information necessary to plan an experiment in which a lifetime or a branching ratio can be determined. One can find for example other clusters emitted from ^{223}Ra or from other parent nucleus for which the penetrability value is much closer to that of the α-decay than for ^{14}C. The main ideas of our research program allowing to estimate the half-lives have been presented in ref.[4]. As we have been interested to know what happens beyond the low asymmetric fission region, it was natural to assume that the new phenomenon, which is also a quantum-mechanical process of tunneling through the potential barrier, could be interpreted as a spontaneous fission process [4] and studied with the Strutinsky method [18] adapted for a superasymmetric splitting. In the attempt to unify the description of these decay modes with that of the fission, the first step [24-26] was the extension of three variants of the liquid drop model (LDM [27], FRNFM [28] and Y+EM [29]) for systems with charge asymmetry different from the mass asymmetry. Due to the large wealth of experimental data, we have tested the model firstly on alpha decay [30,31,3,4]. The half-lives computed with the WKB method, successfully used in fission [32], were in good agreement with experimental results extended over a range of 24 orders of magnitude (from 10^{-7} to 10^{17} s).

Another model describing in a unified way the light particle emission from highly excited states above the barrier and the fission, was developed by Moretto [33]. To compute the energy and angular distributions of the emitted particles based on statistical considerations, Moretto extended the transition state formalism, used in the induced fission, to the evaporation process from excited compound nucleus. This model and that developed by Swiatecki [34] were recently tested [35] on the production of ^{4}He, Li , $^{7,9}Be$, B , C , N , O , and F in the reaction 90 MeV ^{3}He on ^{nat}Ag. Unlike these models dealing with states excited above the barrier, we are concerned with ground states or low excited states well below the barrier, where quantum-mechanical tunneling and shell effects are extremely important.

The numerical methods used for the computation of the barrier shape and the WKB penetrability are too slow to be used for a systematic search of new decay modes, where we have to consider more than 10^{5} combinations parent-emitted HI.

Consequently an analytical relationship was derived [4] and was extended [5] to account for the angular momentum and small excitation energy effects. The predicting power of the model was illustrated [5] for ^{5}He -radioactivity and β-delayed ^{5}He -

radioactivity. We have seen[4] that all ~2000 nuclei with masses tabulated by Wapstra and Bos[36] are stable (the released energy $Q < 0$) with respect to the spontaneous emission of $^{2,3}H$, $^{3,6-9}He$, ^{4}Li, ^{7}B, and ^{9}C. After He isotopes, the lifetimes for other possible candidates with $Z_2 = 4-10$ have been calculated[6-8]. In this paper we report a more comprehensive list[2] of the decay modes and an improved accuracy due not only to the utilization of the new mass tables[37], but also to the introduction of some shell effects in the zero point vibration energy which gives better fit with experimental data for 380 α-emitters. In this way we expect also to estimate better the lifetime for the new decay modes, especially in the neighbourhood of the magic neutron and proton numbers N_1, Z_1 of the daughter.

II. CALCULATION METHODS FOR THE BARRIER AND THE LIFETIME

We are looking for metastable states[4], for which both the released energy

$$Q = M(A,Z) - [M(A_1,Z_1) + M(A_2,Z_2)], \qquad (1)$$

and the barrier height are positive quantities. At the beginning, the mass excees $M(A,Z)$ for about 2000 nuclides tabulated by Wapstra and Bos[36] have been used[3-8], but now[2] we work with more complete (2200 nuclides) updated tables[37].

A. Numerical Method

In order to find the barrier shape by adding a shell correction δE to the liquid drop model deformation energy E_{LD}

$$E(R) = E_{LD}(R) + \delta E(R) \qquad (2)$$

two parametrizations for nuclear shapes have been adopted: intersected spheres[30], or a spheroid interesected with a small sphere[31]. In the first case, the deformation parameter, R, the separation distance between centers, was varied from an initial value $R_i = R_0 - R_2$ to the infinity. The radii $R_0 = r_0 A^{1/3}$, $R_j = r_0 A_j^{1/3}$ ($j = 1,2$) are calculated with different values of the radius constant parameter r_0, when E_{LD} is computed in the framework of various variants of the liquid drop model: LDM[27], FRNFM[28] and Y+EM[29]. These models have been extended[24-26] for different charge densities of the two fragments because, for example, the alpha particle has $N_2/Z_2 = 1$, but N_1/Z_1 for ^{208}Pb is as large as 1.54.

By choosing as the origin of the potential energy the value at infinite separation distance between fragments and a suitable[30] phenomenological correction term δE, from eq. (2) one has initially $E(R_i) = Q$ – the experimental Q-value given by eq. (1). In the overlapping region of the two fragments, for $R_i \le R < R_t = R_1 + R_2$ numerical methods[25,38] are used to compute the barrier shape $E(R)$,but when they are separated ($R > R_t$), analytical relationships are avaiable.

Following the method successfully used to calculate the spontaneous fission lifetimes[32], the half-life of a metastable system is given by

$$T = \frac{\hbar \ln 2}{\Gamma} = \frac{\ln 2}{\nu P}, \qquad (3)$$

where Γ is the disintegration width, $\nu = \frac{\omega}{2\pi} = \frac{2E_v}{h}$ represents the number of assaults on the barrier per second – the characteristic frequency of the collective mode and $E_v = \hbar\omega/2$ is the zero point vibration energy. According to the WKB theory, the probability per unit time of penetration through the barrier

$$P = [1 + \exp(K)]^{-1} \simeq \exp(-K), \qquad (4)$$

where the action integral

$$K = \frac{2}{\hbar} \int_{R_a}^{R_b} \{2\mu[E(R) - Q']\}^{1/2}\, dR, \qquad (5)$$

and the initial energy

$$Q' = Q + E_v. \qquad (6)$$

The mass parameter, μ, can be replaced by the reduced mass $\mu = m_n A_1 A_2 / A$, in which m_n is the nucleon mass. R_a and R_b are the entrance and exit points calculated from the equations $E(R_a) = E(R_b) = Q'$.

If we compare the decay width $\Gamma = \frac{E_v}{\pi} P$ with the relationship $\Gamma = 2\gamma^2 \cdot P'$ used in the R-matrix theory of α-decay, it seems that the role of the reduced width γ^2 – a quantity which is proportional with the cluster preformation probability, is played in our case by the zero point vibration energy. Nevertheless, the penetrability P' is not dependent on γ^2, but P is function of E_v through the eqs. (5) and (6). Also in our P there is a contribution of the deformation energy $E(R)$ of the system of two extended objects, not only a point in a potential obtained from scattering as it is usually considered when P' is calculated. Hence E_v is not a preformation probability.

Consequently we found that the variation of E_v with the neutron number N_1 of the deughter nucleus for alpha decay is much smaller than that of γ^2. This "stability" of the parameter E_v is a useful property when the method is employed to predict the lifetime for a new decay mode.

For clusters heavier than α particle, it is not known up to now how one can calculate the preformation probability. This is another reason to use the fission theory in which E_v is obtained by fitting the experimental data. One has to mention in this respect that due to the fact that the reduced width $\gamma^2 \not< \gamma^2_\alpha$ and is unknown, the argument used by some experimenters [9,11,12] that they found a good ratio P'/P'_α, does not imply necessarily that the branching ration $\frac{\Gamma}{\Gamma_\alpha} = \frac{\gamma^2}{\gamma^2_\alpha} \cdot \frac{P'}{P'_\alpha}$ will be also good. So it is desirable to calculate the lifetimes not only the penetrabilities.

One has to do a numerical quadrature to find the half-life by using the barrier $E(R)$ computed numerically. This method has been used [30,3,4] for alpha decay of some 58 even-even α emitters. Due to the fact that the potential barrier is different when E_{LD} is given by LDM, FRNFM and Y+EM, the best overall agreement with experimental results ranging over 24 orders of magnitude has been obtained with $E_v = 0.37$; 0.37 and 0.3 MeV respectively.

It is worth mentioning that a TDHF calculation [39] for the system ${}^4He + {}^{208}Pb$ leads to a value of $E_v \simeq 0.4$ MeV - very close to the preceding numbers.

For ${}^{14}C$ radioactivity of ${}^{223}Ra$, when E_{LD} is computed in the framework of the Y+EM, one needs $E_v = 0.025$ MeV, to reproduce the T.

B. Analytical Formula

To solve systematically the dynamical problem of the new decay modes, we need to consider a huge number of combinations parent nucleus - emitted HI. For example, only for the 100 isotopes of the emitted elements with $Z_2 = 2-10$ which have been considered in ref. [5-8] as possible candidates to be emitted from ~ 2000 nuclides listed by Wapstra and Bos [36], we have more than 10^5 cases. But of course, the real number is much larger because we went [2] further with Z_2 up to $Z_2 = 24$ and there are 2200 nuclides in the new mass table [37].

For such a large amount of calculations, necessary to find the most important peculiarities of the new decay modes, the numerical procedure is too slow. One needs an analytical relationship for the lifetime.

A first version of an analytic formula has been developed [4] since 1980, approximating the potential barrier in the overlapping region by second order polynomial in R. This was improved [5] in 1983, to account for the contribution of the angular momentum, $\hbar\ell$, and of a small excitation energy. As a consequence of the spin and parity conservation, one has a centrifugal barrier when the parent or/and the emitted HI are not even-even nuclei or the process is going from an excited state with a finite spin. The excitation is useful for enhanced emission processes from excited states populated for example by β-decay of a precursor [5] or by a thermal neutron capture, etc.

After replacing all the constants, by expressing the time in s, the energies in MeV and the lengths in fm, the decimal logarithm of the half-life is given by the following relationships:

$$\log T = 0.43429\,(K_{ov} + K_s) - \log E_v - 20.8436\,, \tag{7}$$

$$K_{ov} = 0.2196\,(E_b^o A_1 A_2 / A)^{1/2} \{ (b^2 - a^2)^{1/2} - \frac{a^2}{b} \ln [\frac{b + (b^2 - a^2)^{1/2}}{a}] \}, \tag{8}$$

$$K_s = 0.4392\,(\frac{Q' A_1 A_2}{A})^{1/2} R_b J_{mc}\,. \tag{9}$$

$$E_i = E_c + E_\ell\ ; \quad E_c = \frac{1.44}{R_t} Z_1 Z_2\ ; \quad E_\ell = \frac{20.735\,\ell(\ell+1)\,A}{R_t^2 A_1 A_2}\,, \tag{10}$$

$$R_b = \frac{R_t E_c}{Q'} [\frac{1}{2} + (\frac{1}{4} + \frac{Q' E_\ell}{E_c^2})^{1/2}]\ ; \quad r = \frac{R_t}{R_b}\,, \tag{11}$$

$$Q' = Q + E_v + E^*\ ; \quad a = b[\frac{Q' - Q}{E_b^o}]^{1/2}\ ; \quad b = R_t - R_i\,, \tag{12}$$

$$J_{mc} = (c+m-1)^{1/2} - [r(c-r)+m]^{1/2} + \frac{c}{2} [\arcsin \frac{c - 2r}{(c^2+4m)^{1/2}} -$$

$$- \arcsin \frac{c-2}{(c^2+4m)^{1/2}}] + \sqrt{m} \ln \{ \frac{2\sqrt{m}[r(c-r)+m]^{1/2} + cr + 2m}{r[2\sqrt{m}(c+m-1)^{1/2} + c + 2m]} \}, \tag{13}$$

$$m = \frac{r^2 E_\ell}{Q'}\ ; \quad c = \frac{r E_c}{Q'}\ ; \quad r_0 = 1.2249\ fm\,, \tag{14}$$

where E^* is the fraction of the excitation energy, concentrated in the collective mode leading to separation.

The zero point vibration energy, $E_v = 0.51$ MeV, used at the beginning [5,6] was obtained by a fit with a selected set of experimental data [40] T_i^{exp} on 376 α emitters. Recently [41] this set was improved and completed. Now there are 380 α-decaying nuclides and as it is shown by the curve (a) from figure 1, for $E_v = 0.51$ MeV, the standard deviation of log T

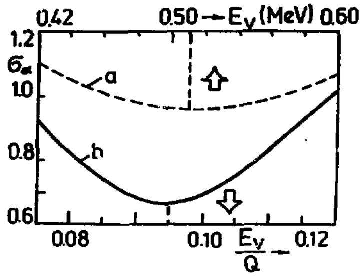

Fig. 1. Choosing the zero point vibration energy E_v *, by minimization of the standard deviation of logT values for 380* α *-emitters.* E_v *= constant (a) and* E_v *proportional with* Q_α *, when* $Q_\alpha > 0$ *(b).*

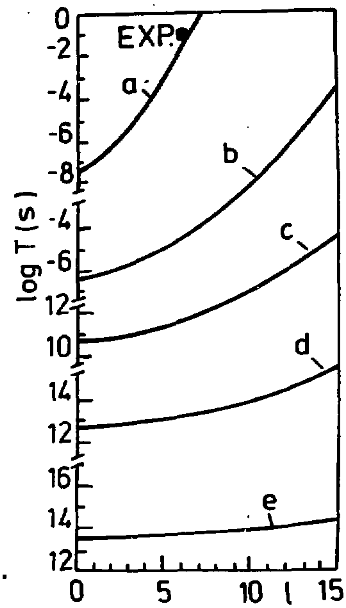

Fig. 2. The centrifugal barrier effect on the lifetime for emission of various charged particles: a) protons from ^{151}Lu *(*E_v *= 0.51 MeV); b)* α *from* ^{212}Po*; c)* ^{8}Be *from* ^{216}Rn*; d)* ^{14}C *from* ^{222}Ra *and e)* ^{48}Ca *from* ^{256}No.

$$\sigma = \left\{ \frac{1}{379} \sum_{i=1}^{380} (\log T_i - \log T_i^{exp})^2 \right\}^{1/2}, \qquad (15)$$

takes the minimum value $\sigma = 0.96$.

When the result of the first measurement for the ^{14}C radioactivity of ^{223}Ra was available [11], we have seen [6-8] that our predictions made by using $E_v = 0.51$ MeV have been too pessimistic by 3 orders of magnitude. In order to reproduce also the new experimental data, we have used [6-8] the law $E_v = 0.1275\ A_2$.

But with our semiempirical formula [42,41] derived for α decay lifetime by using the fission theory, in which the shell effects have been carefully taken into account, one gets $\sigma = 0.41$

for the same data. Hence it was evident that some shell effects are present in the zero point vibration energy. This fact was seen very clearly when we displayed the variation of E_v calculated to reproduce exactly the lifetime for each α emitter, versus the neutron number of the daughter nucleus. The shape of this dependence suggested a simple way to introduce shell effects in E_v, by making it proportional with Q. The curve (b) of fig. 1 shows the optimum value $E_v = 0.095\ Q$ for which one has $\sigma \approx 0.73$ - smaller than for E_v = constant. A convenient law, extended to the other decay modes, was found[2] to be

$$E_v = Q[0.056 + 0.039 \exp(\frac{4 - A_2}{2.5})]; \quad Q > 0; \quad A_2 \geq 4. \qquad (16)$$

In this way, for α one has $E_v = 0.095\ Q$ and for ^{14}C radioactivity $E_v = 31.8 \times 0.056 \approx 1.77$ MeV.

The results presented in the following sections are obtained by using this equation. We need further experimental data in order to check the accuracy of the estimations made with the present relationships. Of course we can get only rough estimations based on this simple model, but the most interesting results obtained in this way, could be studied experimentally or with more refined theoretical models.

From figure 2, one can see that a small angular momentum, up to 5 units of $\hbar$, gives an important contribution to the lifetime only for light emitted particles (especially for protons). For heavier ions one can neglect small angular momentum effects, hence in the following we should not consider the spin and parity conservation, but we can keep in mind that the centrifugal barrier could worsen the estimated values, especially for lighter HI.

III. LIFETIME PREDICTIONS

In a systematic search for new decay modes, the first step consists in finding the regions of the nuclear table where the condition of metastability is fulfilled. We have used the Wapstra and Audi[37] new version of the mass tables to compute the Q values. Up to now we have done calculations in which all isotopes of the elements with $Z_2 < 25$ are supposed to be possible candidates of emitted HI from all nuclides with $A > 2A_2$ and $Z > 2Z_2$. Usually only the 2200 nuclides tabulated by Wapstra and Audi have been consideted but sometimes we extended the region by using a computer program for mass extrapolations[43] and some of the 1975 mass predictions[44].

Since 1980 we have rejected[4] some of these candidates like $^{2,3}H$, ^{4}Li, ^{7}B, and ^{9}C for which $Q < 0$, and we devoted a special

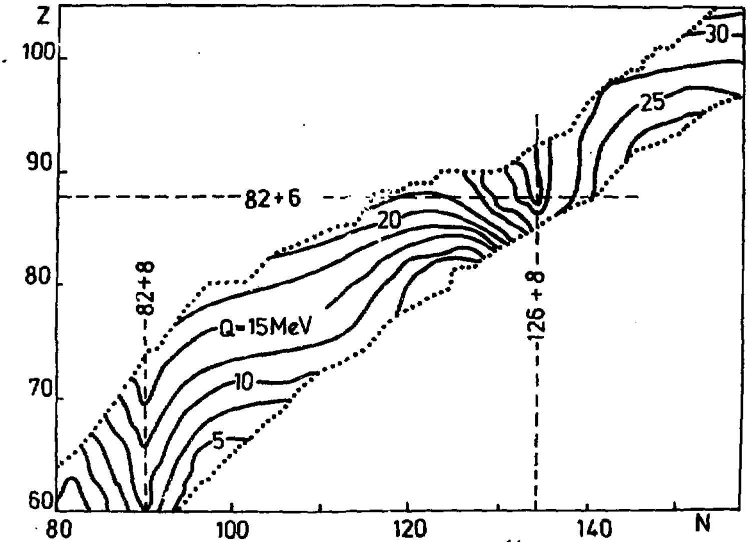

Fig.3. Contour plots of Q-*values for* ^{14}C *emission from various nuclides with* $Z \geq 60$ *and* $N \geq 80$ *having masses tabulated by Wapstra and Bos* /36/.

consideration /5/ to the He isotopes with A_2=3-9 where besides the well-known ^4He, we found ^5He -radioactivity for two islands of translead nuclei leading to daughters with neutron numbers in the neighbourhood of the shell closure $N_1 = 126$ and of the subshell at $N_1 = 152$. Like the diproton, dineutron and trineutron, ^5He and ^8Be are unstable, but one can assume a mechanism in which the components are leaving together the parent nucleus and after tunneling through the potential barrier they are moving apart.

A part of the nuclear chart, with $Z > 60$ and $N > 80$, can be seen in figure 3, where the contour plots of Q-values for ^{14}C emission are drawn. For a given mass number, $A = Z + N$, there is a smooth (liquid-drop model) trend of increasement from the neutron rich side toward the neutron deficient side, like for α-decay Q-values. The reverse is true for very neutron rich emitted HI like $^{8,9}He$. The shell effects for the daughter magic numbers $Z_1 = 82$, $N_1 = 82$, 126 are also very clearly seen, pushing down the maximum Q-value for ^{14}C emission from $^{222}_{88}Ra$ (33.05 MeV) and from $^{223}_{89}Ac$ (33.08 MeV).

By comparing figure 3 with a similar one for α decay, one can see that the condition of metastability for HI emission is fulfilled for a larger region of the nuclear chart. From this point of view these exotic radioactivities are more general phenomena. But their intensities are weaker (the lifetimes larger) than that of α-decay.

The very strong shell effect at the double magic daughter ^{208}Pb present on figure 3, which has been seen in the asymmetric fission mass distribution calculated with the fragmentation theory [22,3], was observed also for all other emitted HI. Hence, we can resume in figure 4 the Q-values for the HI emission in which the daughter is ^{208}Pb. The released energy increases with Z_2 and for $Z_2 > 16$, one has $Q > 100$ MeV. This energy is shared between the emitted particle and the daughter. One can assume that like for α decay the kinetic energy of the HI is given by $E_k = QA_1/A$ and the recoil energy of the daughter is $Q-E$. But it can happen that for heavier emitted particles, like in fission, a part of this energy is lost to excite or to deform the fragments.

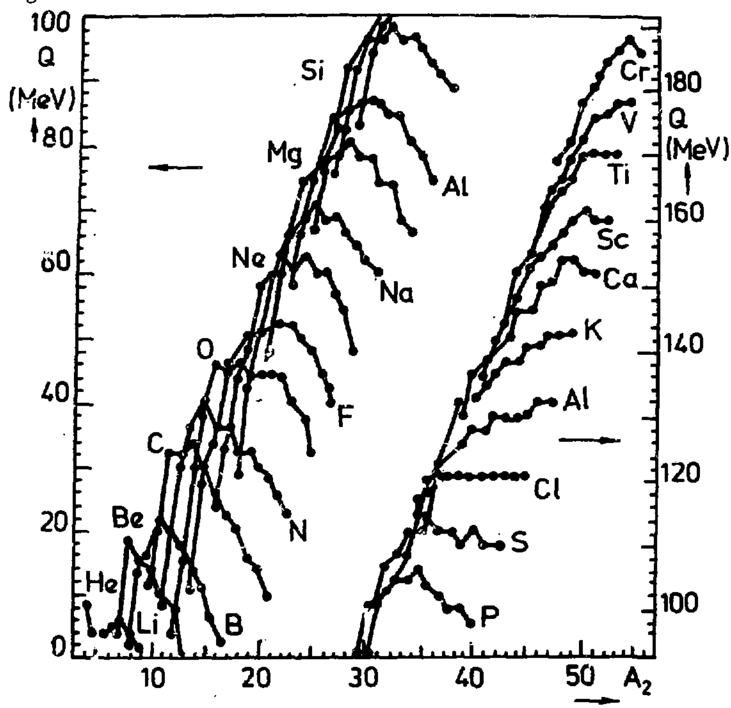

Fig.4. The released energy for the emission of various isotopes of the specified elements from the parents leading to the double magic ^{208}Pb daughter nucleus.

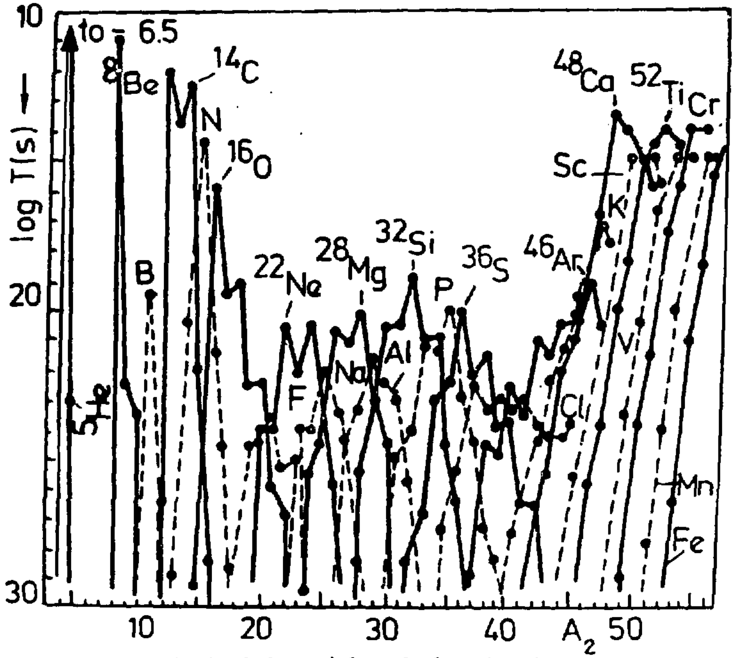

Fig.5. The decimal logarithm of the lifetimes for the same processes as in figure 4.

The corresponding predicted lifetimes are plotted in figure 5, displaying the best values expected for almost all new decay modes. To guide the eye, various isotopes of a given emitted element are joined with a heavy line if Z_2 is even, or with a dotted line if Z_2 is odd. The specified mass numbers of some emitted HI are used as reference points. For a practical reason, Z_2 is restricted to values smaller than 26(parents with $Z < 103$), but of course we can continue even for the superheavy nuclei.

Almost all nuclei are metastable with respect to several decay modes, but if the lifetimes are very long, they are stable from practical point of view. If the age of the universe is of the order of 10^{10} years (that is $10^{17.5}$ s), then the processes with a half-life much shorter than that explains why some nuclides could not be found in nature. Of course, one has to consider all the competing decay modes, the most important determining the minimum partial half-life. The available experimental techniques allow one to measure very slow processes ($T \simeq 10^{22}$ years), as for example the spontaneous fission of some actinides. Consequently we cut figure 5 at $T = 10^{30}$ s. The time scale of all figures of this paper (except figures 2 and 8) is reversed, because we want to get the impression of the intensity which is proportional with T^{-1}.

The most important shell effect is in the region of the double magic ${}^{208}_{82}Pb$ daughter. Similar predictions[2] for lighter parent nuclei leading to the double magic ${}^{132}_{50}Sn_{82}$ show longer lifetimes.

On figure 5 one can see also a pairing effect: for odd N_2 or Z_2 emitted HI, the lifetime is longer than for their even neighbours. There are few exceptions to this rule concerning Z_2, as for example ${}^{15}_{7}N_8$ and ${}^{35}_{15}P_{20}$, but in these cases the neutron numbers N_2 are magic ones, and proton numbers are almost magic.

For each A_2 on figure 5 one has a Z_2 giving the minimum lifetime. When A_2 is increased beyond $A_2 = 16$, the better emitted HI for a given A_2 becomes more and more neutron rich and the corresponding parent - more neutron deficient nuclei. For $Z_2 > 17$ there is a cut in the curves at higher values of A_2, due to the fact that the corresponding masses (on the neutron rich side of the emitted particles or the neutron deficient one of the parents) are not available.

The list of all new decay modes given on this figure is very large: ${}^{5}He$, ${}^{8-10}Be$, ${}^{11,12}B$, ${}^{12-16}C$, ${}^{13-17}N$, ${}^{15-22}O$, ${}^{18-23}F$, ${}^{20-26}Ne$, ${}^{23-28}Na$, ${}^{23-30}Mg$, ${}^{27-32}Al$, ${}^{28-36}Si$, ${}^{31-39}P$, ${}^{32-42}S$, ${}^{35-45\ldots}Cl$, ${}^{37-47\ldots}Ar$, ${}^{40-49\ldots}K$, ${}^{42-51\ldots}Ca$, ${}^{44-53\ldots}Sc$, ${}^{46-53\ldots}Ti$, ${}^{48-54\ldots}V$, ${}^{49-55\ldots}Cr$, etc. There are more than 140 kinds of exotic radioactivities with $Z_2 = 2-24$ and they continue for $Z_2 > 24$. *From these one can select a list of most intensive* processes for a given A_2: ${}^{5}He$, ${}^{8}Be$, ${}^{11}B$, ${}^{12,14}C$, ${}^{15}N$, ${}^{16-20}O$, ${}^{21}F$, ${}^{22-24}Ne$, ${}^{25}Na$, ${}^{26-28}Mg$, ${}^{29}Al$, ${}^{30-34}Si$, ${}^{35}P$, ${}^{36-38}S$, ${}^{39,41}Cl$, ${}^{40,42-44}Ar$, ${}^{45}K$, ${}^{46-50}Ca$, ${}^{51-53}Ti$, ${}^{54,55}Cr$, etc. For each element, one or two isotopes are the best emitters: ${}^{8}Be$, ${}^{11}B$, ${}^{12,14}C$, ${}^{15}N$, ${}^{16}O$, ${}^{21}F$, ${}^{22-24}Ne$, ${}^{25}Na$, ${}^{26-28}Mg$, ${}^{29}Al$, ${}^{32}Si$, ${}^{35}P$, ${}^{36}S$, ${}^{37}Cl$, ${}^{46}Ar$, ${}^{47}K$, ${}^{48}Ca$, ${}^{49}Sc$, ${}^{52}Ti$, ${}^{53,54}V$, ${}^{54,55}Cr$, etc.

A detailed illustration of the $N_1 = 126$ neutron and $Z_1 = 82$ proton shell effects in the daughter nucleus is given in figure 6, where the lifetimes for the 16 decay modes (${}^{8}Be$, ${}^{12-14}C$, ${}^{15}N$, ${}^{23}F$, ${}^{24,25}Ne$, ${}^{28}Mg$, ${}^{32}Si$, ${}^{46}Ar$, and ${}^{48}Ca$) are plotted versus the daughter neutron number N_1, for various daughter proton numbers $Z_1 = 80-87$. Almost always the minimum value of the lifetime for a given Z_1 is obtained when $N_1 = 126$. But for some light emitted HI (like ${}^{8}Be$ and ${}^{12}C$) Z_1 for minimum lifetime is not 82 like usually. Nevertheless, the improvement with respect to the values given in fig.5 is not larger than one order of magnitude. Also the even-odd effect can be better seen on figures 6 (c), (e), (f) and (h). For even-odd emitted HI

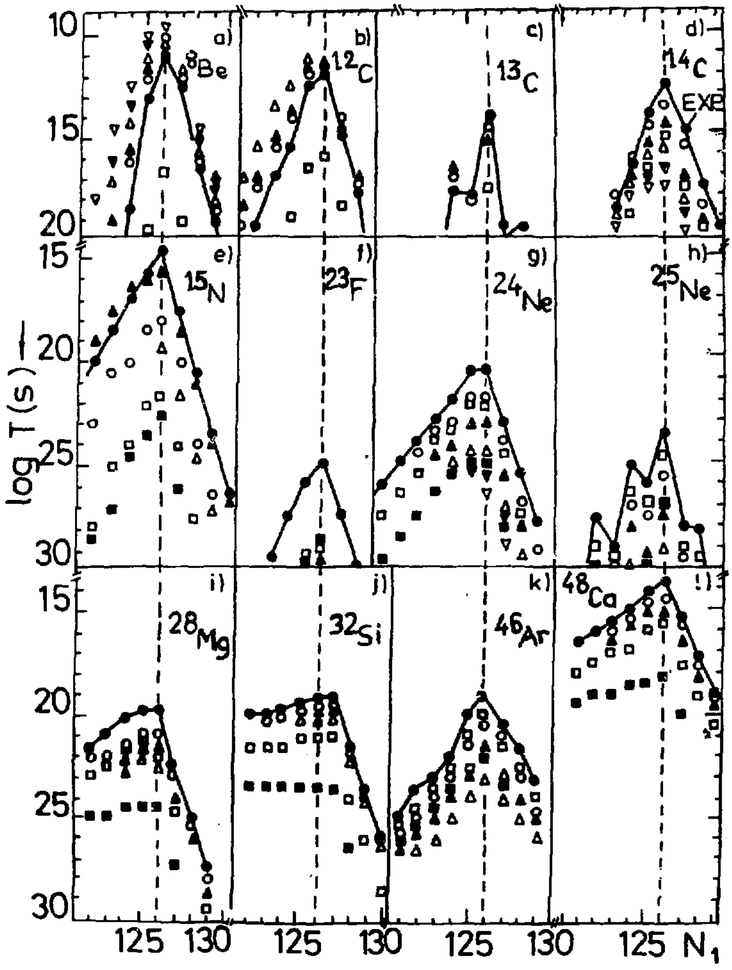

Fig. 6. Lifetimes vs daughter neutron number, N_1 *, for various decay modes by spontaneous emission of the following nuclei:* ^{8}Be *(a);* ^{12}C *(b);* ^{13}C *(c);* ^{14}C *(d);* ^{15}N *(e);* ^{23}F *(f);* ^{24}Ne*(g);* ^{25}Ne *(h);* ^{28}Mg *(i);* ^{32}Si *(j);* ^{46}Ar *(k); and* ^{48}Ca *(l). The daughter atomic numbers are:* ■ – *80;* □ – *81;* ● – *82;* ○ – *83;* ▲ – *84;* Δ – *85;* ▼ – *86; and* ∇ – *87.*

(${}^{13}_{6}C_7$ and ${}^{25}_{10}Ne_{15}$) the even N_1 (odd N) are preferred. For odd-even (${}^{15}_{7}N_8$ and ${}^{23}_{9}F_{14}$) the even Z_1 (odd Z) are the best.

In figure 7 we compare the results obtained with the numerical method (the points) and with the analytical formula (curves). One can see that not far from the magic neutron number of the daughter the agreement is good, but in general the results of the analytic formula are more optimistic than that of the numerical method. This fact may be due to the inclusion of shell effects in the parameter E_v of the lifetime analytical formula; in the numerical method we have been working with $E_v =$ = const.

The minimum value obtained for ${}^{8}Be$ emission is about 17 orders of magnitude longer than that of the best value for alpha emission. But ${}^{8}Be$ is itself an unstable nucleus fissioning in

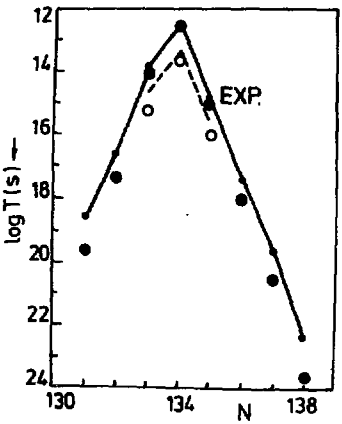

Fig.7. Comparison of lifetimes for ${}^{14}C$ *emission from various isotopes of* Ra *and* Ac*, calculated with a numerical method* (● – Ra , O – Ac) *and the analytical formula* (—— – Ra , - - - – Ac).

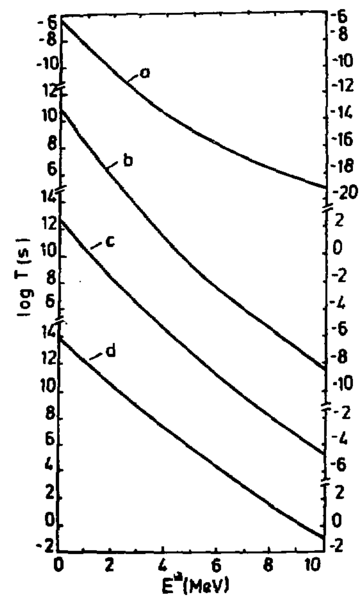

Fig.8. Lifetimes for the emission from excited levels of ${}^{4}He$ *(a);* ${}^{8}Be$ *(b);* ${}^{14}C$ *(c) and* ${}^{48}Ca$ *(d), when the daughter is* ${}^{208}Pb$. E^* *is the fraction of the excitation energy concentrated in the separation degree of freedom.*

two alpha particles, hence it will be difficult to reject the α background in order to detect the $^{8}Be \rightarrow 2\alpha$ particles. Nevertheless, in some plastic detectors , ^{8}Be is very easy identified due to the specific T-shaped track.

From figure 8 one can see that the lifetime for various decay modes can be substantially diminished by exciting the emission level above the ground state. A precursor nucleus far off the β stability line could populate the emitter excited levels. The β-delayed HI emission can be analyzed in the same way as the β-delayed ^{5}He radioactivity/5/ .In fact we can say that the right word is not β-delayed but β-enhanced HI emission, because the lifetime for HI emission from excited state is usually longer than that of the β decay of the precursor. Alternatively the feeding of excited levels could be supplied by thermal neutron capture like in (n,α) reactions.

IV. BRANCHING RATIO

Usually the most important competitor of these new decay modes is the alpha decay. Consequently for some experiments/11,12/ the absolute value of the lifetime T is not so important as the branching ratio relative to alpha decay T/T_{α}.We have shown/7,8/ that from this point of view Rose and Jones /11/ discovered the best emitter of ^{14}C - namely ^{223}Ra with a branching ratio $\frac{\Gamma}{\Gamma_{\alpha}} = T_{\alpha}/T = (8.5 \pm 2.5)\cdot 10^{-10}$ confirmed by Alexandrov et al./12/: $(7.6 \pm 3.0)\cdot 10^{-10}$ and by Gales et al./9/: $(5.5 \pm 2.0)\cdot 10^{-10}$ Of course, if the α particles could be discriminated (for

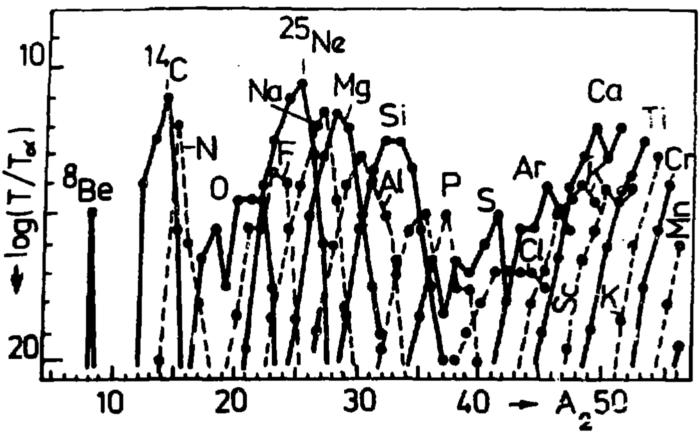

Fig.9. The branching ratios relative to alpha decay for the same processes like in figure 4.

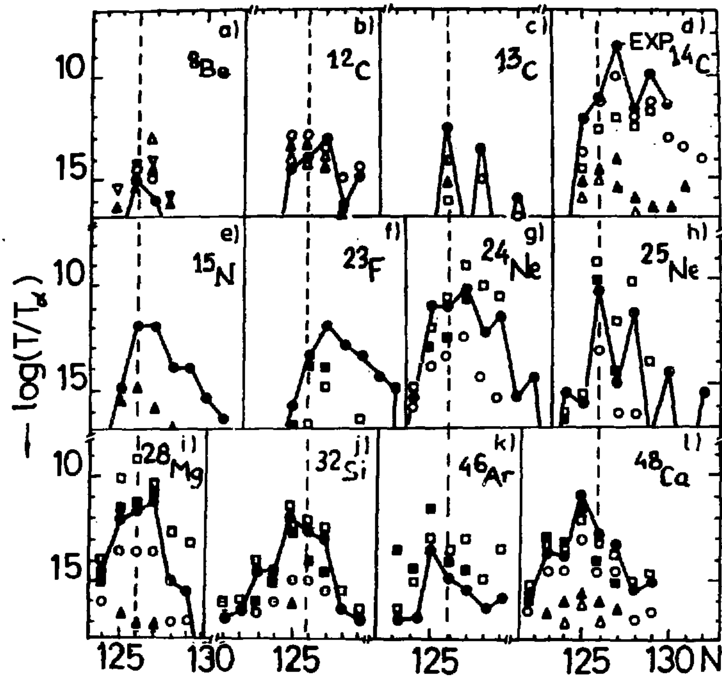

Fig.10. The branching ratios relative to alpha decay for the same processes as in figure 6.

example, deflected /9/ by the magnetic field), the most important quantity is the lifetime. From this point of view the best ^{14}C emitter is ^{222}Ra and not ^{223}Ra.

The branching ratio relative to the α decay was plotted in figure 9 for the same processes as in figure 5, leading to the double magic daughter ^{208}Pb. If a measured α-decay lifetime is not available in the tables /40,41/, we calculate T_α by using the semiempirical formula /42,41/.

One can see that figure 9 is very different from figure 5. For example ^{8}Be, which was the best in figure 5 ($T = 10^{11}$ s) is now pushed down three orders of magnitude under $\frac{T}{T_\alpha} \leq 10^{12}$ and above this level one has not only ^{14}C ($T = 12.6$ s) and ^{48}Ca ($T = 13.5$ s) but also ^{24,25}Ne and ^{28,29}Mg with $T > 10^{20}$ s.

We have shown /7,8/ on the example of ^{14}C radioactivity that the simple rule that the optimum case is obtained for the double magic daughter or not too far from it, which works quite well for the absolute value of log T, is no longer useful for the branching ratio T/T_α. Now we present much more decay modes in figure 10.

Only in a small number of cases (^{13}C, ^{15}N, ^{25}Ne and ^{28}Mg) the best branching ratio is obtained at $N_1 = 126$. Usually $N_1 = 127$ is preferred for HI lighter than ^{25}Ne and $N_1 = 125$ for emitted ions heavier than ^{28}Mg. $Z = 82$ is on the top for $^{13,14}C$, ^{15}N, ^{23}F and ^{48}Ca, but we can have $Z_1 = 85$ for ^{8}Be, $Z_1 = 80$ for ^{46}Ar and $Z = 81$ for $^{24,25}Ne$, ^{28}Mg, and ^{32}Si. The calculated numbers seem to indicate that it would be difficult to perform experiments due to the weak intensity of these processes, but we know that small branching ratios, relative to α decay, in the range 10^{-13} - 10^{-10} have been determined for the spontaneous fission of some Th, U, Np, and Am isotopes.

In the experimental search for new decay modes one can use either figures 5 and 6 for the absolute value of T or figure 10 for the branching ratio. More details about some cases in which good ratio T/T_α is obtained, are given in the Table. One has to stress that T_α is the partial lifetime only for the strongest alpha transition, hence sometimes it is different from the lifetime for all alpha transitions. In order to see if other

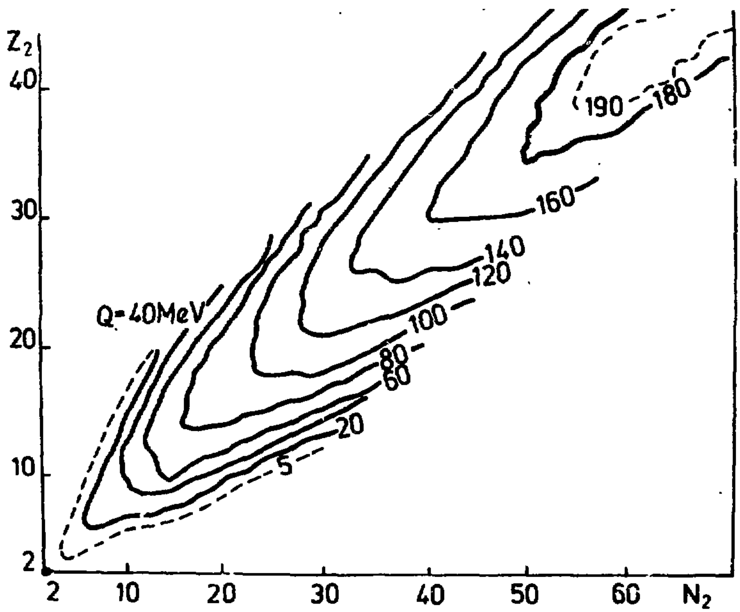

Fig.11. Q values for the emission of HI with Z_2 protons and N_2 neutrons from a source of ^{233}U.

decay modes are in competition with α-decay, not only T_α but also the total half-life T_t are given in this Table. One can see, for example, that the following nuclei $^{224}_{89}Ac$, $^{236}_{93}Np$, $^{239}_{96}Cm$ and $^{253}_{100}Fm$ have $|T_\alpha - T|$ larger than one order of magnitude. There are other cases with $T/T_\alpha \lesssim 10^{12}$ but $T_t \ll T_\alpha$, which are not mentioned in the Table. Some of the parents in this Table are not on the neutron-deficient side of the nuclear chart where, T_t is minimum, due to the fact that T_α is also small in this region.

Table. Some high branching ratio (small T/T_α) HI emissions

Emitted HI	Parent Z	Parent A	Daughter Z_1	Daughter N_1	E_k^{a} (MeV)	log T_t(s)	log T_α(s)	log T(s)	log$\left(\frac{T}{T_\alpha}\right)$
$^{14}_{6}C_8$	87	222	81	127	28.2	2.94	5.4	17.3	11.9
	88	222	82	126	31.0	1.59	1.6	12.6	11.0
		223	82	127	29.9	5.99	6.3	15.0	8.7
		224	82	128	28.6	5.55	5.5	17.4	11.9
		226	82	130	26.5	10.73	10.7	22.4	11.7
	89	223	83	126	31.0	2.12	2.5	13.4	10.9
		224	83	127	30.0	4.02	5.6	15.4	9.8
$^{15}_{7}N_8$	89	223	82	126	36.8	2.12	2.5	14.7	12.2
		224	82	127	35.2	4.02	5.6	17.7	12.1
$^{24}_{10}Ne_{14}$	91	231	81	126	54.1	12.00	12.0	22.0	10.0
	92	232	82	126	55.9	9.36	9.5	20.4	10.9
		233	82	127	54.3	12.70	12.8	23.1	10.3
		235	82	129	51.5	16.33	16.6	28.1	11.5
$^{25}_{10}Ne_{15}$	92	233	82	126	54.3	12.70	12.8	23.3	10.5
		235	82	128	51.7	16.33	16.6	28.1	11.5
$^{28}_{12}Mg_{16}$	92	233	80	125	65.3	12.70	12.8	24.5	11.7
	93	236	81	127	66.2	12.56	14.0	24.5	10.5
	94	236	82	126	70.2	7.96	8.1	19.8	11.7
$^{29}_{12}Mg_{17}$	92	235	80	126	63.7	16.33	16.6	27.4	10.8
	93	236	81	126	66.0	12.56	14.0	25.1	11.1
$^{32}_{14}Si_{18}$	96	239	82	125	84.5	4.03	7.0	18.9	11.9
$^{46}_{18}Ar_{28}$	98	251	80	125	103.0	10.45	10.9	22.6	11.7
$^{48}_{20}Ca_{28}$	100	253	80	125	118.2	5.41	6.7	18.4	11.7
	101	254	81	125	121.6	3.23	4.0	15.9	11.9
	102	255	82	125	124.7	2.30	2.8	14.1	11.2
$^{50}_{20}Ca_{30}$	101	258	81	127	119.4	6.64	6.6	17.7	11.1
	102	259	82	127	122.0	3.54	4.1	16.4	12.3

a) $E_k = Q \cdot A_1 / A$

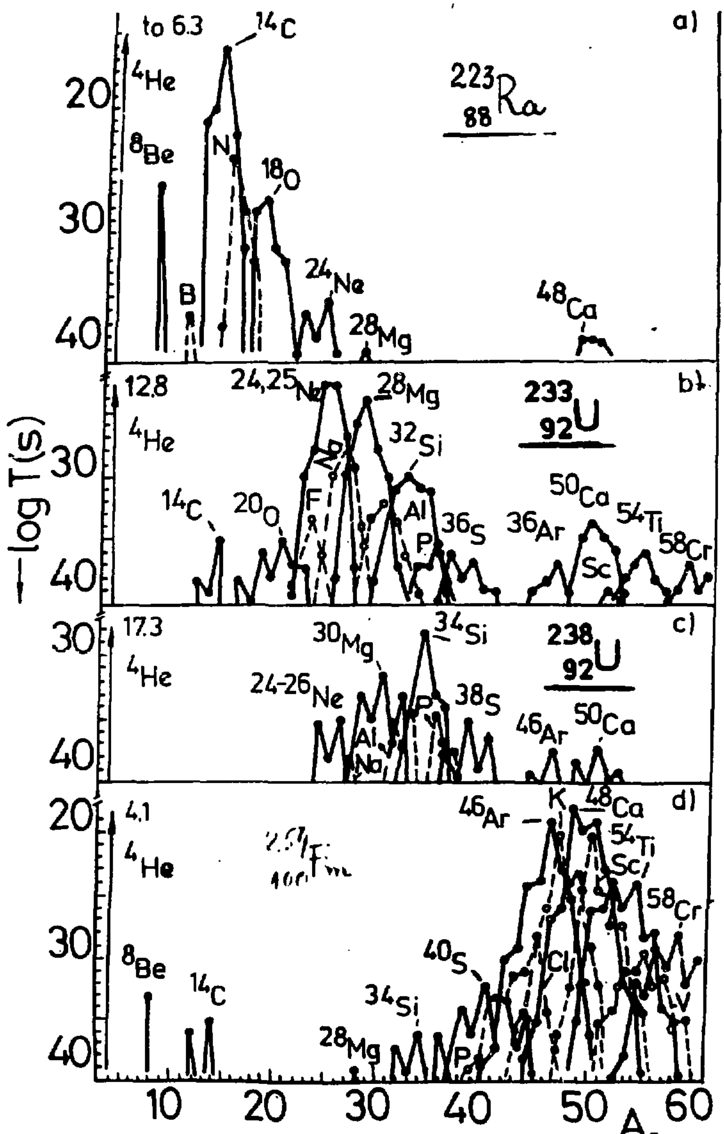

Fig. 12. Lifetime spectra for various modes of decay by EI emission of ^{223}Ra *(a),* ^{233}U *(b),* ^{238}U *(c), and* ^{254}Fm *(d).*

Other problem important in planning an experiment is to know the spectrum of, the emitted particles from a given source A,Z available. From the Q-value variation with the neutron and proton numbers N_2, Z_2 of the emitted HI, as it is shown in figure 11 for the ^{233}U parent nucleus, one cannot answer the question. These are many emitted particles with positive Q-values. The ridge shape appearing in this figure, means that usually for each A_2 there is one combination of Z_2, N_2 giving the maximum Q-value and the corresponding Q increases when A_2 increases.

In figure 12 we present the spectrum of the emitted particles from various sources: ^{223}Ra, ^{233}U, ^{238}U, and ^{254}Fm. In the first case one can see that the other HI ($^{12,13,15}C$) which are emitted from ^{223}Ra are more than 5 orders of magnitude weaker than ^{14}C, already, measured. We expect that from ^{233}U, the most probable emission will have $^{24,25}Ne$ ($T \simeq 10^{23}$ s and $T/T_{\alpha} \simeq 10^{10.5}$, as it is written in the Table). Then follows ^{28}Mg with $T = 10^{24.5}$ s and $T/T_{\alpha} \cong 10^{12}$.

^{238}U is predicted to have $T \simeq 10^{28}$ s for the emission of ^{34}Si and $T/T_{\alpha} \simeq 10^{11}$; ^{30}Mg is 4 orders of magnitude lower. But the case of $^{34}_{14}Si_{20}$ need to be examined further in order to check the validity of the approximation for the mass value of the daughter ($^{204}_{78}Pt_{126}$). For this reason, ^{34}Si was not given in the Table and there are many other similar cases. We need better accuracy in mass predictions and more experimental points to get more confidence in our estimations. From ^{254}Fm, the most likely emitted ion is ^{48}Ca with $T \simeq 10^{18}$ s and $T/T_{\alpha} \simeq 10^{16}$.

By comparing the spectra of all sources from fig. 12 one can see the trend toward heavier emitted particle when the mass number of the parent increases. This trend is similar with the one observed in the light fragment part of the asymmetric fission fragment mass distribution. In our case it is explained by the shell effect at $N_1 = 126$: $^{223}_{88}Ra_{135} \rightarrow {}^{14}_{6}C_8 + {}^{209}_{82}Pb_{127}$; $^{233}_{92}U_{141} \rightarrow {}^{24,25}_{10}Ne_{14,15} + {}^{209,208}_{82}Pb_{127,126}$; $^{238}_{92}U_{146} \rightarrow {}^{34}_{14}Si_{20} + {}^{204}_{78}Pt_{126}$ and $^{254}_{100}Fm_{154} \rightarrow {}^{48}_{20}Ca_{28} + {}^{206}_{80}Hg_{126}$. Once more one can see that the neutron shell effects at $N_1 = 126$ are usually much stronger than the proton ones at $Z_1 = 82$.

In fission for some heavy Fm isotopes the mass distribution is symmetric because the light fragment has approximately a magic neutron number 82 (or not too far from it) like the heavy fragment. But the HI emission is essentially an asymmetric process because, at least at present, it is hopeless to believe that the superheavy element $^{416}164$, for which one has symmetric fission in two fragments ^{208}Pb, will be ever available.

V. CONCLUSIONS

The asymmetric distribution of the fragment masses in the spontaneous fission of actinides is due to the shell effects in the region of magic numbers $N = 82$ and $Z = 50$ for spherical shape or the corresponding ones for deformed shapes.

By using the fragmentation theory we found a new peak in this distribution at a higher asymmetry, due to the shell effects in the region of the magic numbers $N = 126$, $Z = 82$.

The shell effects in the region of the double magic ^{208}Pb are responsible for many new decay modes of trans-lead nuclei in which HI are spontaneously emitted. But the phenomenon is also present, with weaker intensity, in the trans-tin region where ^{132}Sn plays the major role. Almost all nuclei are metastable with respect to several decay modes but if the lifetime is very long (for example longer than 10^{30} s) one can say that from the practical point of view, the corresponding nucleus is stable.

In 1980 we have made predictions for some of the new decay modes by calculating only the penetrabilities /45/. One can see for example that ^{14}C has a good probability to escape from ^{222}Ra and ^{224}Ra in comparison with its neighbours and similarly ^{34}Si from ^{238}U, etc. The same quantity has been used in 1984 by Rose and Jones who discovered the ^{14}C radioactivity of ^{223}Ra and by other two groups giving the confirmation of this experiment. For ^{14}C emission from ^{222}Ra and ^{224}Ra we predicted that the penetrability should be $10^{6.4}$ and $10^{7.1}$ times smaller than for the α-decay. The similar ratio calculated by Rose and Jones for ^{14}C emission from ^{223}Ra is in the range $10^{2.6}-10^{4.2}$ according to the adopted value of the radius constant. In the experiment they have determined a branching ratio of $\sim 10^{9}$. By comparing these numbers one can say that the branching ratio for ^{14}C emission from ^{223}Ra is better than that from $^{222,224}Ra$. But if the emitted HI from ^{238}U is ^{34}Si, it has a penetrability which is by 4 orders of magnitude higher than that of the α-decay. As we have shown in this paper, such a situation could never arise when we use our method to compute the lifetimes. We concluded that the penetrabilities do not hinder the emission of heavy clusters and in a small region of emitted nuclei, they could be used to get a rough information about the new decay modes.

But the penetrability is not a measurable quantity. Then we developed a method allowing to calculate the lifetime and the branching ratio relative to α decay determined in the experiment. This model is based on the assumption that the new phenomenon could be considered to be a superasymmetric fission. Hence the Strutinsky and the WKB methods successfully used in fission theory have been adapted for this case and used to calculate the potential barriers and the lifetimes. We learned very much by using the model in 1979-80 for alpha decay, where a large wealth (380 emitters) of experimental data is available.

The numerical method is too slow to be used in a systematic search for new decay modes where a huge amount of combinations parent nucleus-emitted HI has to be taken into account. A faster technique of computation, based on an analytical formula, was developed since 1980. This formula was improved to account for angular momentum and small excitation energy effects and used to predict in 1983 the ^{5}He and β-delayed ^{5}He radioactivities.

The lifetime predictions for the emission of various HI with $Z_2 = 4$-10, made before we knew the experimental result for ^{14}C radioactivity, by using the same zero point vibration energy as for α-decay, have been slightly pessimistic (with the 1980 variant one obtains $10^{17.7}$ s instead of $\sim 10^{15}$ s which was found in the experiment), but they were improved immediately in agreement with this unic result. Now we have introduced a shell effect also in the zero point vibration energy improving the agreement for α decay so one can have more confidence in these new estimations. Of course, we need more experimental points (new decay modes with emitted ions heavier than ^{14}C) to improve further the model. Hence the present calculations should be taken only as orientative rough estimations.

From a systematic search across the nuclear table, we have shown the great complexity and diversity of the new phenomenon: there are more than 140 emitted HI-various isotopes of the elements with $Z_2 = 2$-24, for which the lifetimes are in the range 10^{10} - 10^{30} s. For a given decay mode (A_2, Z_2) the minimum lifetime is obtained when the daughter has a magic number of neutrons $N_1 = 126$ and a magic number of protons $Z_1 = 82$ or not too far from it. When the mass number A_2 increases, for $Z_2 > 16$, the position of the optimum-optimorum emitted HI moves in the neutron rich side of the nuclear chart and the parent becomes more and more neutron deficient.

According to our calculations, the best value of the branching ratio relative to α-decay ($\sim 10^{-9}$) is obtained for ^{14}C emission from ^{223}Ra and was already measured. But in this case the absolute value of the lifetime ($\approx 10^{15}$ s) is by two orders of magnitude longer than that of ^{222}Ra for ^{14}C emission, and by 5 orders of magnitude longer than the minimum value for ^{8}Be radioactivity. The simple rule of double magic ^{208}Pb daughter which works with few exceptions for absolute value of the lifetime, is no longer valid for the branching ratio. For the optimum T/T_α, the daughter neutron number values N_1 are spread from 125 to 130 and the proton numbers Z_1 from 80 to 83 as it was shown in the Table, where there are some interesting cases deserving attention.

The increase of the lifetime due to some units of angular momentum carried by the HI order to fulfill the spin and parity conservation laws is usually negligible small, except for some very light emitted particles.

The lifetime could be substantially diminished, if it takes place from an excited level populated by β decay of a precursor far off the stability line (β-enhanced HI emission) or by a thermal neutron capture, like in (n,α) reactions (neutron induced HI emission).

Finally, we can say that the great complexity and diversity of this phenomenon opens a large field of experimental and theoretical investigations.

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45. The quantity plotted in fig.7 of the ref.[3] is the natural logarithm of the penetrability divided by 2.

Received by Publishing Department
on December 17, 1984.

Познару Д.Н. и др. Е4-84-811

Моды распада атомных ядер в результате спонтанной эмиссии тяжелых ионов

Большинство известных ядер, включая так называемые стабильные нуклиды, в действительности являются метастабильными по отношению к некоторым модам спонтанного суперасимметрического расщепления. Если время жизни этих процессов больше, чем скажем 10^{30} с, то явление не может быть обнаружено с помощью существующих экспериментальных приборов, поэтому с практической точки зрения можно считать их стабильными. Модель, выведенная из теории альфа-распада как процесс деления, позволяет оценить время жизни и относительные отношения к альфа-распаду для этих видов естественной радиоактивности. Исходя из большого количества систематических расчетов был сделан вывод, что этот процесс должен происходить с максимальной интенсивностью в ядрах более тяжелых, чем ядра свинца, где максимальное время жизни получено для комбинаций родительские ядра-тяжелые кластеры, приводящих к магическому (^{208}Pb) или почти магическому дочернему ядру. Более чем 140 нуклидов с атомным числом меньше 25 даются как возможные кандидаты, испускаемые из тяжелых ядер с величинами времени полураспада в интервале $10^{10}-10^{30}$ с. В этих новых модах распадов очень ясно проявляются оболочечная структура ядра и парные эффекты.

Работа выполнена в Лаборатории теоретической физики ОИЯИ.

Сообщение Объединенного института ядерных исследований. Дубна 1984

Poenaru D.N. et al. E4-84-811

Atomic Nuclei Decay Modes by Spontaneous Emission of Heavy Ions

The great majority of the known nuclei, including the so-called stable nuclides, are in fact metastable with respect to several modes of spontaneous superasymmetric splitting. If the lifetime against these processes is larger than let's say 10^{30} s, the phenomenon is not detectable with available experimental techniques, hence one can admit stability from the practical point of view. A model extended from the fission theory of alpha decay allows to estimate the lifetimes and the branching ratios relative to the alpha decay for these natural radioactivities. From a huge amount of systematic calculations it is concluded that the process should proceed with maximum intensity in the trans-lead nuclei, where the minimum lifetime is obtained for parent-emitted heavy ion combinations leading to a magic (^{208}Pb) or almost magic daughter nucleus. More than 140 nuclides with atomic number smaller than 25 are possible candidates to be emitted from heavy nuclei, with half-lives in the range $10^{10}-10^{30}$ s. The shell structure and the pairing effects are clearly manifested in these new decay modes.

The investigation has been performed at the Laboratory of Theoretical Physics, JINR.

Communication of the Joint Institute for Nuclear Research. Dubna 1984

30 коп.

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Набор Н.П.Боголюбовой, Л.М.Кашехлебовой.

Подписано в печать 29.12.84.
Формат 60x90/16. Офсетная печать. Уч.-изд.листов 1,95.
Тираж 465. Заказ 35667.

Издательский отдел Объединенного института ядерных исследований.
Дубна Московской области.