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Significance of the light clusters in exotic nuclear decay

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Some aspects of cluster radioactivity are discussed. In particular, aspects related to shell effects, as well as the importance of Q values, are addressed. A plot similar to the Geiger-Nuttal plot, which relates the half-lives of cluster emitters to Q values, is also presented. The question of why particular clusters and their associated daughter nuclei were experimentally observed is discussed and a possible answer is given.

Definite experimental data on radioactive decay with emission of ^{14}C, ^{24}Ne, ^{28}Mg, and Si isotopes has been recorded. Furthermore, for ^{233}U and ^{234}U the partial half-life for the emission of several Ne isotopes has been determined, but the relative contributions of the individual isotopes are not yet known. The same results hold for the emission of Si isotopes from ^{238}Pu. Table I gives the experimental results on cluster radioactivity. Only experiments which actually yield cluster decays are included in Table I [17]. From Table I we can see that there are four Ra isotopes (^{222}Ra, ^{223}Ra, ^{224}Ra, and ^{226}Ra) which decay with the emission of ^{14}C. There are three isotopes ^{230}Th, ^{231}Pa, and ^{232}U which decay with the emission of ^{24}Ne, and two isotopes, ^{234}U and ^{236}Pu, which decay with the emission of ^{28}Mg. Also, there are four more isotopes which decay with cluster emission. The emitted element is known, however, it is not yet clear which isotope it is.

We have calculated the Q values for different carbon isotopes which could possibly be emitted as particles for each of the radium emitters; the results are given in Table

TABLE I. Experimental results on cluster radioactivity decay.

Initial nucleus	Emitted cluster	Daughter product	$\lambda_{\text{cluster}}/\lambda_\alpha$	Reference
^{222}Ra	^{14}C	^{208}Pb	$(3.7\pm0.6)\times10^{-10}$	1
			$(3.1\pm1.0)\times10^{-10}$	2
^{223}Ra	^{14}C	^{209}Pb	$(8.5\pm2.5)\times10^{-10}$	3
			$(7.6\pm3.0)\times10^{-10}$	4
			$(5.5\pm2.0)\times10^{-10}$	5
			$(4.7\pm1.3)\times10^{-10}$	6
			$(6.1\pm1.0)\times10^{-10}$	1
^{224}Ra	^{14}C	^{210}Pb	$(4.3\pm1.2)\times10^{-11}$	1
^{226}Ra	^{14}C	^{212}Pb	$(3.2\pm1.6)\times10^{-11}$	2
			$(2.9\pm1.0)\times10^{-11}$	7
^{230}Th	^{24}Ne	^{208}Pb	$(5.6\pm1.0)\times10^{-13}$	8
^{231}Pa	^{24}Ne	^{207}Tl	$(3.8\pm0.7)\times10^{-12}$	9
^{232}U	^{24}Ne	^{208}Pb	$(2.0\pm0.5)\times10^{-12}$	10
^{233}U	^{24}Ne	^{209}Pb	$(7.5\pm2.5)\times10^{-13}$	11
	and/or	and/or		
	^{25}Ne	^{208}Pb	$(5.3\pm2.3)\times10^{-13}$	12
^{234}U	^{24}Ne	^{210}Pb	$(4.4\pm0.5)\times10^{-13}$	13
	and/or	and/or		
	^{26}Ne	^{208}Pb	$(3.9\pm1.0)\times10^{-13}$	14
^{234}U	^{28}Mg	^{206}Hg	$(1.4\pm0.2)\times10^{-13}$	13
			$(2.3\pm0.7)\times10^{-13}$	14
^{236}Pu	^{28}Mg	^{208}Pb	2×10^{-14}	15
^{238}Pu	^{28}Mg	^{210}Pb	$(5.6^{+4.4}_{-2.5})\times10^{-17}$	16
	and/or	and/or		
	^{30}Mg	^{208}Pb	$(5.6^{+4.4}_{-2.5})\times10^{-17}$	
^{238}Pu	^{32}Si	^{206}Hg	$(1.4^{+0.6}_{-0.4})\times10^{-16}$	16
	and/or	and/or		
	^{34}Si	^{204}Hg	$(1.4^{+0.6}_{-0.4})\times10^{-16}$	

TABLE II. The Q values in MeV for different C isotopes of different Ra isotopes parents.

Parent isotope	Q value for isotope (MeV)				
	^{12}C	^{13}C	^{14}C	^{15}C	^{16}C
^{222}Ra	29.05	28.81	*33.06*	25.48	. . .
^{223}Ra	27.73	28.86	*31.85*	29.14	26.01
^{224}Ra	26.37	26.17	*30.56*	26.57	26.89
^{226}Ra	23.85	23.79	*28.21*	24.28	24.72

II. In Table III we have calculated the Q values for different Ne isotopes for the three parent emitters. For the Mg isotopes the Q values were also calculated for the two parent emitters. The results are given in Table IV.

For α decay a correlation (the Geiger-Nuttal plots) was found between the decay half-lives $T_{1/2}$ and the corresponding Q values:

$$\log_{10} T_{1/2} = b + aQ^{-1/2} . \quad (1)$$

In particular, this relation was found [18,19] to hold for the ground state to ground-state decays of even-even nuclei having the same Z and varying neutron number N. In this respect we found an indication that cluster radioactivity obeys the same type of correlation. A similar indication is given in Ref. [17]. We have considered three Ra isotopes (^{222}Ra, ^{224}Ra, and ^{226}Ra) which are even-even nuclei with the the same Z and $N > 126$. All three nuclei decay with the emission of ^{14}C. In Fig. 1 we have plotted the $\log_{10} T_{1/2}$ with respect to $Q^{-1/2}$. The correlation has a correlation factor of 1.000.

There are three main conclusions that can be reached from Tables I–IV and Fig. 1. From Fig. 1 we can see that there is a similarity to α decay with respect to the relation between $\log_{10} T_{1/2}$ and the Q values. Also, the actual emitted clusters have the highest Q values. For the four cases in which carbon is emitted, the isotope ^{14}C, which is the actual one emitted, has the highest Q value compared to all other carbon isotopes. The same conclusion is valid for the Ne and Mg isotopes. The isotope ^{24}Ne, which is the actual emitted particle among all the Ne isotopes, has the highest Q value of all the Ne isotopes for the three emitters (^{230}Th, ^{231}Pa, and ^{232}U). For the Mg isotope, ^{28}Mg has the highest Q value of all Mg isotopes. This result has been obtained for the two Mg emitters, ^{234}U and ^{236}Pu. The third conclusion is that for all the observed cases with definite identification of the emitted clusters, the daughter nuclei are "magic" with respect to the number of neutrons 126 or the number of protons 82. Among the nine cases with positive identification of the daughter nuclei, four are ^{208}Pb, which is "double magic," with respect to the neutrons and protons.

TABLE III. The Q values in MeV for different Ne isotopes.

Parent Isotope	Q value for isotope (MeV)				
	^{22}Ne	^{23}Ne	^{24}Ne	^{25}Ne	^{26}Ne
^{230}Th	. . .	. . .	*57.77*	55.32	55.13
^{231}Pa	55.11	55.35	*60.42*	57.85	56.82
^{232}U	57.36	57.38	*62.31*	59.21	57.95

TABLE IV. The Q values in MeV for different Mg isotopes.

Parent isotope	Q value for isotope (MeV)				
	^{26}Mg	^{27}Mg	^{28}Mg	^{29}Mg	^{30}Mg
^{234}U	. . .	. . .	*74.13*	71.18	71.96
^{236}Pu	71.30	72.56	*77.13*	73.54	73.24

These observed results indicate the importance that shell effects play in cluster radioactivity. Cluster emission rate is proportional to the frequency factor and the penetrability given by [20].

$$P = \exp - \left[\frac{2}{\hbar} \int_{R_a}^{R_b} \sqrt{2M(r)[V(r) - Q]}\, dr \right] . \quad (2)$$

The integration is between two turning points. The two fragments of the decay have atomic numbers Z_1 and Z_2 and their radii are R_1 and R_2. The energy barrier is equal to the Coulomb potential $Z_1 Z_2 e^2/r$ for $r > R_1 + R_2$.

Thus, if we are considering the possible cluster decay of isotopes of the same element, then Z_1 and Z_2 are the same, and the Coulomb potential will be about the same. So from Eq. (1) we can see the important role that the Q value has on P. Thus, it is not surprising that, in all the cluster emissions, those actually observed have the highest Q values.

These high Q values certainly raise an interesting question: Why do these particular combinations of clusters and daughter products have the highest Q values? Or, in other words, why are these particular clusters and their associated daughter nuclei observed? Part of the answer to this question has already been suggested by Sandulescu, Poenaru and Greiner [21]. They predict that "the penetrabilities of heavy clusters in some two-body channels are comparable to or even greater than the penetrability in the α-particle decay channel. This occurs because of shell effects in the value of Q in the corresponding channels."

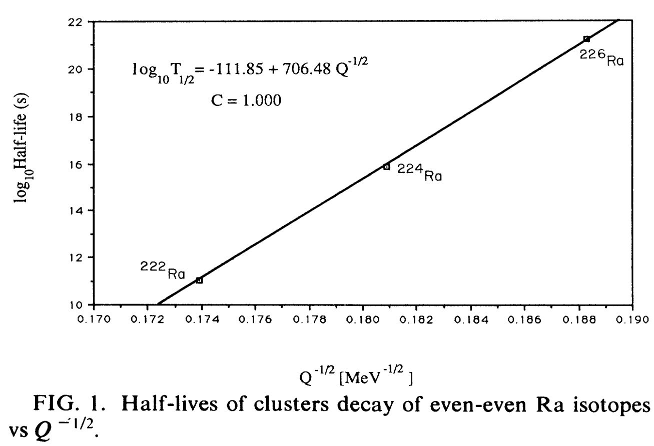

FIG. 1. Half-lives of clusters decay of even-even Ra isotopes vs $Q^{-1/2}$.

Indeed, the search for cluster emissions, as well as their actual discovery, began with the assumption that in all the cases of cluster emissions the decay daughter products must be nuclei close to double magic nucleus ^{208}Pb.

The experimental results of cluster emissions support the fact that shell effects are involved and, indeed, all the daughter products are close to the double magic ^{208}Pb. In all the cases in which the daughter products were identified (see Table I), these daughter products are either magic with the number of neutrons (126) or with the number of protons (82) or both ($^{208}_{82}$Pb). So the "rule of thumb," suggested by Wang *et al.* [16] might be slightly changed to read "the most favorable parent for emission of a particular fragment is one leading to a heavy *magic* daughter close to the double magic ^{208}Pb."

However, there are some problems in this rule of thumb and with the shell effects explanations. If shell effects were the only effects associated with cluster emission, we would have expected that the daughter nuclei would actually be ^{208}Pb rather than only close to it. Why do ^{223}Ra and ^{224}Ra emit ^{14}C, and not ^{15}C and ^{16}C which will lead to ^{208}Pb as the daughter product? Furthermore, in the case of ^{226}Ra the daughter product is ^{212}Pb which is not so close to ^{208}Pb. For Si emission from ^{238}Pu [16,22] the experiment was not able to resolve whether ^{32}Si or ^{34}Si clusters were emitted. If the Q value is the dominant factor, we should expect ^{34}Si to be emitted, since its Q value is 92.89 MeV compared to 91.21 MeV for ^{32}Si emission. On the other hand, calculations [16] of rate, using Gamow penetration factor, predict that ^{32}Si is much more likely to be emitted than ^{34}Si. However, if the cluster emitted in the ^{238}Pu case is actually ^{34}Si, then the daughter product is ^{204}Hg which is not magic.

So, there are some problems with an explanation based solely on shell effects. We must conclude then that the cluster itself plays a role. The fact that four isotopes emit ^{14}C, three or even up to five isotopes emit ^{24}Ne, and two or three isotopes emit ^{28}Mg, indicates that there is something special about these emitted clusters. Thus, it seems that the clusters themselves play a role in these decays and not only the daughter nuclei. Furthermore, the similarity of the cluster decay to the α decay also indicates that the emitted cluster is important. This poses an interesting question: What is so important about ^{14}C as a cluster? If clusters are important in these decays, intuitively we should expect ^{12}C to be that cluster, as Rose and Jones [3] had expected in their first experiment on cluster decay. The isotope ^{12}C is the most stable isotope of all carbon isotopes; it can be considered as a combination of three α decays. In the case of ^{222}Ra, the daughter would have been ^{210}Pb, which is the actual daughter obtained for ^{224}Ra decay. However, ^{14}C, and not ^{12}C, was observed.

So the immediate question arises: What is so special about the observed clusters ^{14}C, ^{24}Ne and ^{28}Mg?

It should be noted that in the experiments only long-lived parent nuclei are feasible. The N/Z ratios of these parent nuclei are relatively high. The lower N/Z ratio was obtained for the parent nucleus ^{236}Pu with $N/Z = 1.5106$. The highest N/Z ratio is for the parent nucleus ^{226}Ra with $N/Z = 1.5682$. On the other hand, the heavy daughter with double shell closure at $Z = 82$, $N = 126$ has a N/Z ratio of 1.5366. This ratio is lower than the majority of its neighbors in the same mass region of the valley of stability. The N/Z ratios of the daughter nuclei ranges from 1.5366 of ^{208}Pb to 1.5854 of ^{212}Pb. So, in order to maintain a relatively low N/Z ratio of the daughter nucleus we should expect rather high N/Z ratios for the emitted clusters. In this respect the observed clusters ^{14}C, ^{24}Ne, ^{28}Mg, and ^{34}Si have N/Z ratios of 1.333, 1.4, 1.333, and 1.4286, respectively. However, the difference for instance between the stable ^{12}C and the emitted cluster ^{14}C is only two neutrons which do not have too strong an effect on the N/Z ratios of the daughter nuclei.

In a recent paper [23], we have suggested that most of the nuclei are composed of two cluster blocks of deuterons and tritons. The number of the deuteron clusters (Nd) is $2Z - N$ (Z is the number of protons and N is the number of neutrons) and the number of triton clusters Nt is then $N - Z$. The total number of clusters in a given isotope is Z. Thus, all isotopes of the same element have the same number of cluster blocks.

We have calculated the BE (binding energy) per cluster for all the C, Ne, Mg, and Si isotopes. The BE per cluster is calculated as

$$\mathrm{BE} = (Nd \times md + Nt \times mt - m)/(Nd + Nt) , \qquad (3)$$

where m, md, and mt are the masses (in MeV) of the isotope in question and of the deuteron and the triton, respectively. The results of the calculations are given in Tables V–VII.

From these tables we can see that the emitted clusters ^{14}C, ^{24}Ne, and ^{28}Mg each has the highest BE per cluster.

As a result of this observation we would like to suggest the following "rule" for cluster emission: The most favorable parents for cluster emissions are those that emit clusters which have the highest BE per cluster, and in which the daughter nuclei is preferably magic, close to the double magic ^{208}Pb.

TABLE V. The binding energy per cluster for carbon isotopes.

BE per cluster (MeV)				
^{12}C	^{13}C	^{14}C	^{15}C	^{16}C
13.04	12.84	*13.17*	12.35	12.03

TABLE VI. The binding energy per cluster for neon isotopes.

BE per cluster (MeV)						
^{20}Ne	^{21}Ne	^{22}Ne	^{23}Ne	^{24}Ne	^{25}Ne	^{26}Ne
13.74	13.81	14.23	14.13	*14.40*	14.21	14.14

TABLE VII. The binding energy per cluster for Mg isotopes.

BE per cluster (MeV)							
^{24}Mg	^{25}Mg	^{26}Mg	^{27}Mg	^{28}Mg	^{29}Mg	^{30}Mg	^{31}Mg
14.20	14.30	14.71	14.73	*14.93*	14.73	14.75	14.47

The cluster BE for the elements Si, Ar, and Ca was also calculated. From these calculations we obtained that among these elements the nuclei which have the highest BE per cluster are ^{34}Si, ^{44}Ar, and ^{48}Ca.

So, according to our suggestion, we should expect ^{34}Si emission from ^{238}Pu; this suggestion suits the fact that the highest Q value is obtained for the emission of ^{34}Si.

For the emission of ^{44}Ar, the best candidate according to our suggestion, with a relatively long half-life (13.08 yr), is ^{250}Cf:

$$^{250}_{98}\mathrm{Cf} \rightarrow {}^{44}_{18}\mathrm{Ar} + {}^{206}_{80}\mathrm{Hg}\,.$$

With respect to Ar emission, the reaction with emission of $^{44}_{18}$Ar has the highest Q value. The daughter nuclei $^{206}_{80}$Hg has a magic number of neutrons, and ^{206}Hg is close to ^{208}Pb. Furthermore, the relatively long half-life for spontaneous fission (1.7×10^4 yr) also makes ^{250}Cf a favorable candidate from the experimental point of view.

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