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FORMATION OF NUCLEAR MOLECULES IN CLUSTER RADIOACTIVITY
On Interpretation of the Cluster Radioactivity Mechanism

V. V. Volkov, E. A. Cherepanov [1]

Joint Institute for Nuclear Research, Dubna

The basis for cluster radioactivity is the property of nuclei of light isotopes of elements heavier than lead to spontaneously form clusters — nuclei of light elements — from valence nucleons, which gives rise to asymmetric nuclear molecules. The cluster formation proceeds through successive excitation-free transfer of valence nucleons to the α particle and to subsequent light nuclei. Nuclear molecule formation is accompanied by a considerable amount of released energy, which allows quantum-mechanical penetration of the cluster through the exit Coulomb barrier.

В основе кластерной радиоактивности лежит свойство ядер легких изотопов элементов тяжелее свинца спонтанно формировать из валентных нуклонов кластеры — ядра легких элементов, в результате чего возникают асимметричные ядерные молекулы. Процесс формирования кластера протекает путем последовательной, без возбуждения, передачи валентных нуклонов α-частице и последующим легким ядрам. Формирование ядерной молекулы сопровождается выделением значительной энергии, которая делает возможным квантово-механическое проникновение кластера через выходной кулоновский барьер.

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INTRODUCTION

Cluster radioactivity is underbarrier emission of nuclei of light elements from ground-state nuclei of radium and actinide elements. This phenomenon is intermediate between spontaneous fission and emission of particles. A possibility of this nuclear process was theoretically predicted by Sandulescu, Poenaru, and Greiner in 1980 [1]. Four years later, cluster radioactivity was first observed experimentally by Rose and Jones [2], and by Aleksandrov et al. [3]. Both groups observed emission of ^{14}C from the ^{223}Ra nucleus.

The discovery of cluster radioactivity aroused lively interest of both experimenters and theorists. Though observation of cluster radioactivity is quite a difficult experimental problem, a lot of cluster emitters have been found in the past years, and emission of light nuclei ranging from ^{14}C to ^{34}Si was detected. The experimental data are reviewed in [4–6].

Two theoretical approaches to interpretation of cluster radioactivity have been established. One of them treats this nuclear process as superasymmetric fission [1, 5, 7] and the other treats it as a process similar to α-particle emission [6, 8–10]. Both approaches are based on

[1] E-mail: cher@jinr.ru

two well-studied nuclear processes, spontaneous fission and particle emission. A comparative analysis of the two approaches was performed in [11].

Investigation of interaction between two nuclei in low-energy collisions resulted in discovering two new objects in the nuclear microworld: nuclear molecules [12] and double nuclear systems [13, 14]. We believe that these discoveries make it possible to formulate a more realistic concept of cluster radioactivity.

1. SPONTANEOUS FORMATION OF NUCLEAR MOLECULES IN CLUSTER RADIOACTIVITY

Cluster radioactivity is observed in light isotopes of radium and actinide elements. A cluster is formed from valence nucleons that are in states higher than states of nucleons in the doubly magic ^{208}Pb nucleus playing the role of the core. The cluster comprises practically all valence nucleons of the parent nucleus. For example, in the ^{222,223,224}Ra nuclei the cluster is the ^{14}C, in ^{232,233}U it is the ^{24}Ne nucleus, in ^{236}Pu it is the ^{28}Mg nucleus, and in ^{242}Cm it is the ^{34}Si nucleus.

The nuclear system consisting of the ^{208}Pb nucleus and a light cluster nucleus is actually an asymmetric nuclear molecule. Both nuclei are in the ground state and interact with each other through the nucleus–nucleus potential. Condensation of valence nucleons of the parent nucleus to a cluster is accompanied by release of energy Q amounting to tens of MeV. This energy is converted to the oscillatory motion of the light nucleus-cluster. However, Q is always much smaller than the exit Coulomb barrier, and cluster emission is due to quantum-mechanical penetration of the cluster through the potential barrier. Cluster radioactivity proceeds in two stages. At the first stage an asymmetric nuclear molecule is formed and at the second stage it decays due to underbarrier cluster emission. The theoretical approaches to the description of cluster radioactivity include consideration of both stages. The main difference between them is the description of the cluster formation mechanism. In the adiabatic approach, the nuclear molecule results from superasymmetric deformation of the initial heavy nucleus. Figure 1 taken from [7] demonstrates an example of how this stage proceeds. In the nonadiabatic approach, the nuclear molecule results from quantum fluctuation or quantum fragmentation in the initial state of the heavy nucleus.

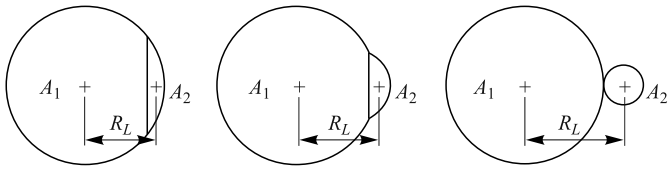

Fig. 1. Evolution of the system over the collective variable R_L from [7]

2. INTERPRETATION OF THE CLUSTER FORMATION MECHANISM WITHIN THE NEW APPROACH

It is the shell structure of the nucleus that underlies cluster radioactivity. Due to the shell structure, a nuclear system of a closely packed doubly magic ^{208}Pb nucleus and loosely bound valence nucleons moving on the periphery of the nucleus is formed in the heavy parent

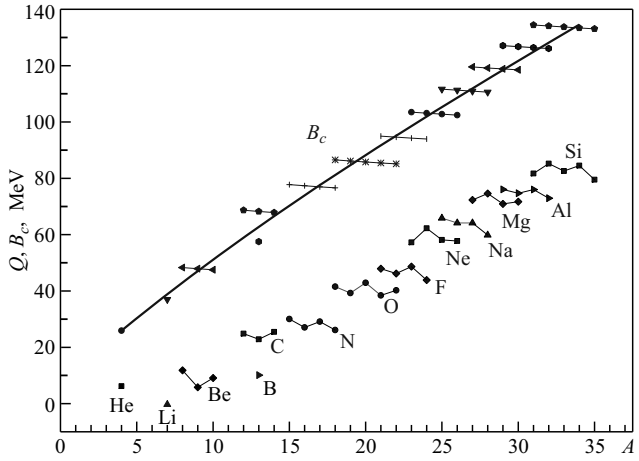

Fig. 2. Energy release Q in the ^{232}U nucleus clustering process as a function of the light nucleus mass. For comparison, the Coulomb barrier B_c between the cluster and residue nucleus conjugate to it is shown

nucleus. Yet, to our mind, condensation of valence nucleons to a light element nucleus, cluster, does not directly follow from the shell model. The discovery of cluster radioactivity revealed a new fundamental property of heavy atomic nuclei — an ability to spontaneously form asymmetric nuclear molecules.

We believe that cluster formation proceeds as follows. In the surface of heavy nuclei, an α particle is formed with a high probability because it is energetically quite favorable to unite two protons and two neutrons into a ^{4}He nucleus. As is evident from Fig. 2, it is energetically favorable to transfer valence nucleons to the particle and then to the light nucleus increasing in mass.

Nucleon transfer goes on until practically all valence nucleons of the heavy nucleus are transferred to the light cluster nucleus. This is the end of the nucleon transfer process because transfer of tightly bound nucleon of the ^{208}Pb nucleus to loosely bound states of the neutron-rich light cluster nucleus is energetically unfavorable. Thus, cluster formation is not an instantaneous quantum fluctuation in the ground state of the initial nucleus but a process in which valence nucleons of the heavy nucleus are gradually condensed to a light-element nucleus cluster.

Valence nucleons of the heavy parent nucleus and nucleons of the light cluster nucleus are in very different states. Valence nucleons have considerable angular momenta, and trajectories of their motion are concentrated in the surface of the heavy nucleus. On the contrary, angular momenta of light cluster nuclei are small and their motion is concentrated in an appreciably smaller spatial domain. For this reason, condensation of valence nucleons to a cluster with the formation of an asymmetric nuclear molecule is quite deep nucleon structure transformation in the initial nucleus. The probability for this restructuring is low. It can be characterized by a factor S that is similar to the reduced clustering width in the nonadiabatic approach. The decay constant, which governs the cluster radioactivity half-life, will involve, as in the nonadiabatic approach, two terms, the cluster formation probability S and the exit Coulomb barrier penetrability for the cluster:

$$\lambda = S \cdot P. \tag{1}$$

Formula (1) has the same structure as in the nonadiabatic approach, but the physical nature of the factor S is radically different. Since cluster formation in our approach is not a single event but a nuclear process that takes time to occur, our approach to interpretation of the cluster radioactivity mechanism can no longer be referred to as nonadiabatic.

Concluding the section, we would like to draw the reader's attention to the singularity of the cluster formation process. Experiment shows that the sum of the kinetic energies of the cluster and the heavy nucleus conjugate to it coincides with the energy Q released in clustering. It means that both nuclei that make up a nuclear molecule are in the ground state. This situation can arise only if the cluster formation proceeds without excitation of the participating nuclei. The clustering process is a chain of quantum ground–ground transitions with nucleon transfer ending in deep restructuring of the initial heavy nucleus. Clustering radically differs from evolution of a double nuclear system, which is of statistic nature. To our mind, the clustering process should be given a special name. It could be referred to as, for example, spontaneous restructuring of a nucleus. It is readily seen that a process of this type can be directly associated with the spontaneous fission of heavy nuclei, at least with its initial stage.

3. CLUSTER RADIOACTIVITY IN NUCLEI OF TRANSCURIUM ELEMENTS

Unfortunately, the authors cannot yet propose a model for condensation of valence nucleons of a heavy nucleus to a cluster. This model is needed for calculating cluster radioactivity half-lives. Yet, the half-life does not allow the most realistic model of this nuclear process to be identified. The published data show that fairly good agreement between the calculated and experimental results for cluster radioactivity half-lives can be obtained within various models.

We think that to evaluate realism of the approach, cluster radioactivity of nuclei of transcurium elements should be considered. In nuclei of radium and light isotopes of the first actinide elements, condensation of valence nucleons to a cluster terminates when all valence nucleons are collected into a cluster. However, if the initial nucleus is a quite heavy nucleus of a transuranium element, e.g., ^{251,252}Cf, condensation of nucleons to a cluster becomes a qualitatively different process.

Condensation of all valence nucleons of the ^{252}Cf nucleus results in formation of the ^{44}S nucleus as a cluster. Near to it there is the doubly magic nucleus ^{48}Ca, and it is energetically favorable to form the cluster that is exactly the ^{48}Ca nucleus. In this case, some of the tightly bound nucleons of the ^{208}Pb nucleus are transferred to the cluster. The neutron and proton binding energies in the ^{208}Pb nucleus are $B_n = 7.4$ MeV and $B_p = 8.0$ MeV, while in the ^{48}Ca nucleus they are substantially higher, $B_n = 10$ MeV and $B_p = 15.8$ MeV.

Calculations of the half-lives for the cluster radioactivity of ^{251}Cf and ^{252}Cf with emission of ^{48}Ca as a cluster, which were carried out within the adiabatic approach, yielded values well accessible to experimental measurements: $\lg T(^{251}\text{Cf}) = 21.9$ and $\lg T(^{252}\text{Cf}) = 21.6$ [5].

In the nonadiabatic approach, the cluster formation probability sharply decreases with increasing cluster mass and charge. The probability for formation of ^{48}Ca in the ^{251,252}Cf nuclei turns out to be vanishingly small. Even for ^{34}Si emission from the ^{242}Cm nucleus, the quantity $T_{1/2}$ calculated within the nonadiabiatic approach [9] turned out to be 500 times larger than the experimentally measured value [15]. In [15, 16] it was assumed that for such massive clusters as ^{34}Si and heavier, only the adiabatic approach holds.

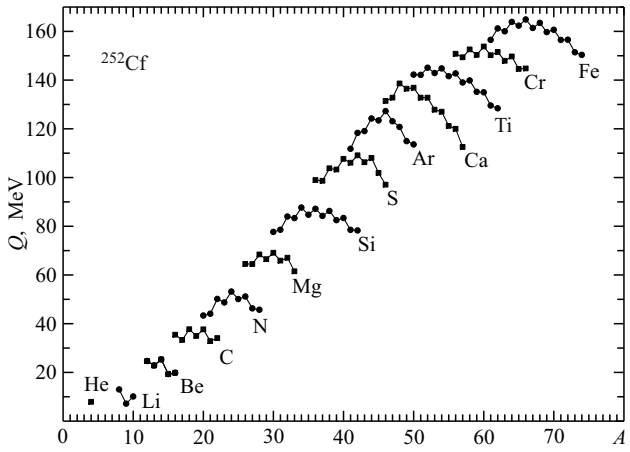

Fig. 3. Released energy Q in clustering in the ^{252}Cf nucleus as a function of the light nucleus mass (for convenience, only Z-even nuclei are chosen)

An important consequence follows from our proposed interpretation of the cluster formation process: cluster radioactivity cannot occur in nuclei of transcurium elements because it inevitably turns to spontaneous fission of the initial nucleus. The point is that the condensation of the valence nucleons of the ^{251,252}Cf nuclei to a cluster does not stop when the ^{48}Ca is formed as a cluster because behind this nucleus there are nuclei with maximum closely packed nucleons. For example, in the ^{64}Ni nucleus $B_n = 9.7$ MeV and $B_p = 12.5$ MeV. As is evident from Fig. 3, the energy Q released in the clustering continues increasing after the formation of the ^{48}Ca cluster as well. Transfer of valence nucleons from the heavy nucleus to the light nucleus stops only when the nucleon binding energies in both nuclei become equal.

When neutron binding energies in both nuclei are equal, the nuclear system of two interacting nuclei undergoes a radical change in character. In the nuclear molecule, nucleons of the light cluster nucleus and of the heavy nucleus conjugate to it are localized in their nuclei, which interact with each other through the nucleus–nucleus potential. When a massive cluster is formed, the neutron binding energies in both nuclei become equal and the motion of the loosely bound neutrons embraces both nuclei, which gives rise to a covalent bond (see Fig. 4). As a result, the nuclear molecule changes into a double nuclear system (DNS).

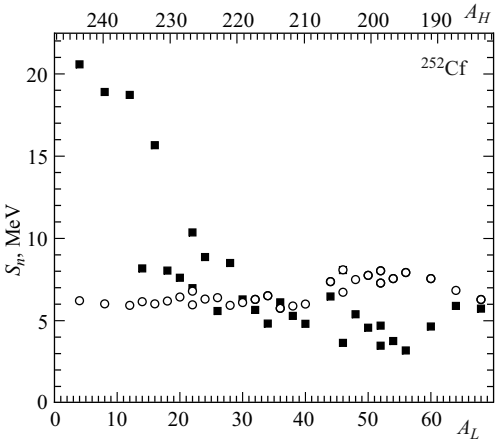

Fig. 4. Neutron binding energy in the light nucleus (squares) and the heavy nucleus (circles) versus the mass of the light fragment (bottom scale) and heavy fragment (top scale). For convenience, only Z-even nuclei are chosen

A nuclear molecule typically changes its state by decaying into two nuclei due to quantum-mechanical penetration of the light nucleus through the Coulomb barrier. A typical way of changing for the DNS is its evolu-

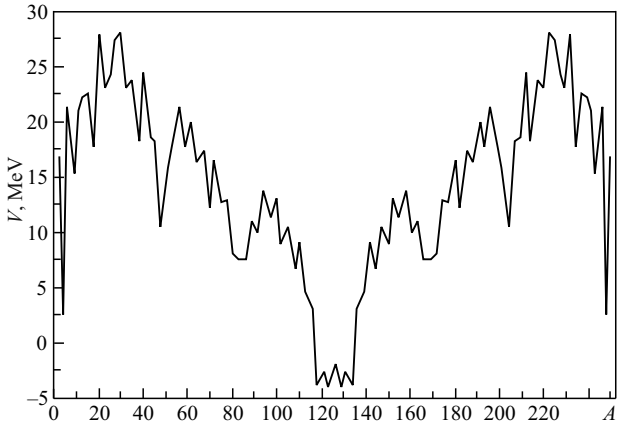

Fig. 5. Potential energy of the DNS for the ^{252}Cf parent nucleus versus the atomic number of one of the DNS nuclei

tion that goes on through nucleon transfer from one nucleus of the system to the other and is governed by the potential energy of the system. It is evident from the shape of the potential energy in Fig. 5 that the evolution of the DNS which resulted from the ^{252}Cf nucleus will be towards symmetrization of its form. Coulomb repulsion between the nuclei will increase, and the DNS will decay into two fragment nuclei; i.e., spontaneous fission of the initial nucleus will occur.

Thus, from the interpretation of the cluster radioactivity process proposed by us it follows that cluster radioactivity cannot occur in nuclei of californium and heavier transuranium elements.

CONCLUSIONS

The central problem of cluster radioactivity is the cluster formation mechanism. Two established theoretical approaches interpret it in fundamentally different ways. In the adiabatic approach, the cluster configuration results from syperasymmetric deformation of the nucleus. In the nonadiabatic approach, it arises from quantum fluctuation in the initial state of the nucleus. Both theoretical approaches are based on the well-known nuclear processes, namely, spontaneous fission of heavy nuclei and emission of α particles.

The discovery of nuclear molecules, double nuclear systems, and deep inelastic transfer reactions allow a new interpretation to be proposed for the cluster formation mechanism. It is assumed that nuclei of elements heavier than lead are capable of spontaneously condensing valence nucleons to nuclei of light elements — clusters. This process results in formation of an asymmetric nuclear molecule, in which both nuclei are in the ground state and interact with each other through the nucleus–nucleus potential. The cluster is formed by successive transfer of valence nucleons to the α particle, which is formed with a high probability in the surface of the initial nucleus, and further on to light nuclei increasing in mass. Cluster formation is an exoergic process. However, the energy release Q in this process is below the exit Coulomb barrier and cluster emission (decay of the nuclear molecule) proceeds as a quantum-mechanical process of penetration through the potential barrier.

Realism of the proposed concept of the cluster formation mechanism can be evaluated by considering cluster radioactivity of quite heavy nuclei like ^{251,252}Cf. Within the adiabatic approach the cluster to be emitted by these nuclei can be the ^{48}Ca nucleus with the experimentally measurable half-life.

Within the proposed approach, emission of the ^{48}Ca cluster is impossible because the process of nucleon transfer from the heavy nucleus to the light nucleus will continue after the formation of ^{48}Ca, ending in spontaneous fission of the initial nucleus.

More than twenty years have passed since the adiabatic calculations of the ^{251,252}Cf half-lives in the decay with emission of the ^{48}Ca cluster were published, and so far nobody in the world has observed cluster radioactivity in the ^{251,252}Cf nuclei.

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REFERENCES

1. *Sandulescu A., Poenaru D. N., Greiner W.* // Part. Nucl. 1980. V. 11, No. 6. P. 1334.
2. *Rose H. J., Jones G. A.* // Nature. 1984. V. 307. P. 245–247.
3. *Aleksandrov D. V. et al.* // Pis'ma Zh. Eksp. Teor. Fiz. 1984. V. 40. P. 152–154.
4. *Price P. B.* // Nucl. Phys. A. 1989. V. 502. P. 41c–58c.
5. *Greiner W. et al.* // Treatise on Heavy-Ion Science / Ed. D. A. Bromley. N. Y.; London, 1989. V. 8. P. 640–722.
6. *Zamyatin Yu. S. et al.* // Part. Nucl. 1990. V. 21, No. 2. P. 537–594.
7. *Pik-Pichak G. A.* // Yad. Fiz. 1986. V. 44. P. 1421–1429.
8. *Kadmenskii S. G. et al.* // Izv. Akad. Nauk SSSR. Ser. Fiz. 1986. V. 50. P. 1786–1795.
9. *Blendowske R., Fliessbach T., Walliser H.* // Nucl. Phys. A. 1987. V. 464. P. 75–86.
10. *Malik S. S., Gupta R. K.* // Phys. Rev. C. 1989. V. 39. P. 1992–2000.
11. *Tchuvil'sky Yu. M.* Cluster Radioactivity. M.: Mosk. Gos. Univ., 1997 (in Russian).
12. *Almgvist E., Bromley D. A., Kuehner J. A.* // Phys. Rev. Lett. 1960. V. 4. P. 515.
13. *Volkov V. V.* // Phys. Rep. 1978. V. 44. P. 93–157.
14. *Schroder W. V., Huizenga J. R.* // Treatise on Heavy-Ion Science / Ed. D. A. Bromley. N. Y.; London, 1984. V. 2. P. 115.
15. *Ogloblin A. A. et al.* // Intern. Conf. «Nuclear Shells — 50 Years», Dubna, 1999 / Ed. Yu. Ts. Oganessian and R. Kalpakchieva. Singapore: World Sci., 2000. P. 124–133.
16. *Sagaidak R. N. et al.* // Phys. Rev. C. 2007. V. 76. P. 034605-1.

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