Nuclear Decay by Cluster Emission

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Theoretical predictions within a scientific collaboration based in Germany for the spontaneous emission of clusters from nuclei have now been confirmed experimentally at some 10 laboratories.

Following the discovery of radioactivity by H. Becquerel (the 100th anniversary of which is celebrated next year), three natural decay modes (α, β and λ) of nuclei had been identified by 1940, when spontaneous fission was observed for the first time. A rich variety of other nuclear decay modes have since been discovered and studied [1].

Cluster radioactivity — an intermediate phenomenon between fission and α-decay — was predicted in 1980 [2] and confirmed experimentally in 1984 with the discovery of the ^{14}C decay of ^{223}Ra [3]. Interest in this unusual process has grown continuously as details have been revealed. In cluster radioactivity, a parent nucleus ^{A}Z with an atomic number Z breaks apart into two fragments, namely the emitted cluster $^{A_e}Z_e$ and the daughter cluster $^{A_d}Z_d$. The spontaneously emitted light fragment is a small nucleus (such as ^{14}C, ^{24}Ne, *etc.*) which is heavier than an α-particle, but lighter than the lightest fission fragment.

As in both spontaneous fission and α-decay, our basic understanding relies on the theory of quantum mechanical tunnelling through a potential barrier which was initially developed by Gamow (and independently by Condon and Gurney) in 1928. Theoretically, any nucleus with $Z > 40$ for which the released energy Q (the difference between the sums of the initial and final masses) is a positive quantity can be a cluster emitter. In practice, observations are severely restricted owing to limitations imposed by currently available experimental techniques which require a sufficiently short half-life and a sufficiently large branching ratio for the decay process.

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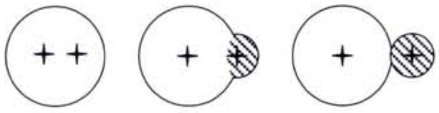

Fig. 1 — A sequence of intersected spherical shapes (from initial parent nucleus to two separated fragments in contact) for ^{24}Ne cluster radioactivity of ^{232}U as assumed by the superasymmetric model used to calculate the decay parameters. The process continues by increasing the separation distance between the centres.

Theoretical Aspects

Any theory of cluster radioactivity capable of making predictions should answer the following questions: Is the phenomenon allowed physically? Can it be detected? What are the most probable parent nuclei and emitted clusters? What is the order of magnitude of the emission rate? Four theoretical approaches answering at least some of these questions were reviewed in 1980 [2]. They were based on:

- Fragmentation theory by solving a Schrödinger equation with mass asymmetry as a variable to obtain the mass distribution of fission fragments;
- Penetrability calculations to determine the probability of quantum tunnelling through a fission barrier similar to those used in the traditional theory of α-decay;
- Superasymmetric fission models (Fig. 1), both numerical (NuSAF) and analytical (ASAF), describing a fission process with a very large mass asymmetry.

Other approaches, many of which had been undergoing development since as early as 1924, gave misleading conclusions.

It was a calculation of the fission-fragment mass distribution for ^{252}No using fragmentation theory and a two-centre shell model that first predicted a new superasymmetric peak owing to cluster radioactivity (the cluster was ^{38}S in this case). The two-centre shell model, developed by Mosel, Maruhn, Greiner *et al.* based in Frankfurt, allows one to estimate readily the wave function for a sequence of nuclei shapes generated during binary fission or fusion. Eight decay modes by cluster emission from 16 even-even parent nuclei (see insert) have since been predicted by calculating the penetrability.

Decay Modes

Penetrability theory predicts eight decay modes by the emission of clusters of:

^{14}C, ^{24}Ne, ^{28}Mg, ^{32,34}Si, ^{46}Ar, ^{48,50}Ca

from the following even-even parent nuclei:

^{222,224}Ra, ^{230,232}Th, ^{236,238}U, ^{244,246}Pu ^{248,250}Cm, ^{250,252}Cf, ^{252,254}Fm, ^{252,254}No

Three variants of the numerical superasymmetric fission model have been developed since 1979 by adding a phenomenological shell-correction term to the macroscopic deformation energy of a binary nuclear system with a different charge density for each of the nuclei, and by performing numerical calculations using the so-called Wentzel-Kramer-Brillouin (WKB) approximation. We have obtained good agreement between the predicted and experimentally determined half-lives for 58 even-even α-emitters over 24 orders of magnitude. Moreover, our semi-empirical relationship based on a fission theory of α-decay gave the best results on making comparisons with data for some 380 nuclei that decay by emitting α-particles.

The ASAF Model

A very large number of parent/emitted cluster combinations have to be considered in any systematic search for new decay modes. More precisely, one must examine on the order of 10^5 combinations in order to check the metastability, against the 200 isotopes of the elements with $Z_e = 2$-28, of the more than 2000 nuclides with measured masses tabulated by Wapstra and Audi. However, the numerical calculation of the three-fold integrals involved in the models mentioned above are very time-consuming. The large amount of computation can only be performed in a reasonable time by using an analytical relationship for the half-life. Since 1980, have we developed our ASAF model to fulfil this requirement. We started with the Myers-Swiatecki liquid drop model for the nucleus and, in the spirit of the Strutinsky method, adjusted it with phenomenological corrections accounting for the known overestimation of the barrier height and for shell and pairing effects.

The model was the first to be used to predict measurable quantities in cluster decay. It has been improved continuously and exploited intensively so that a total of more than 150 cluster decay modes have been predicted. Comprehensive tables of half-lives, branching ratios with respect to α-decay, and kinetic energies of emitted clusters published since 1984 have been used to guide experiments, while several groups of theorists have adopted the results as reference points. The main quantities which are determined experimentally in order to check the theoretical predictions are the partial half-life T and the kinetic energy of the emitted cluster $E_k = QA_d/A$, a quantity that is a direct consequence of the "cold" character of the cluster decay mode, where the total kinetic energy of the two separating fragments practically exhausts the released energy Q shared between the two fragments.

After taking proper account of the influence of even- and odd-numbered nuclei, there is excellent agreement between experimental data and our initial predictions made in 1984 for the half-lives for different

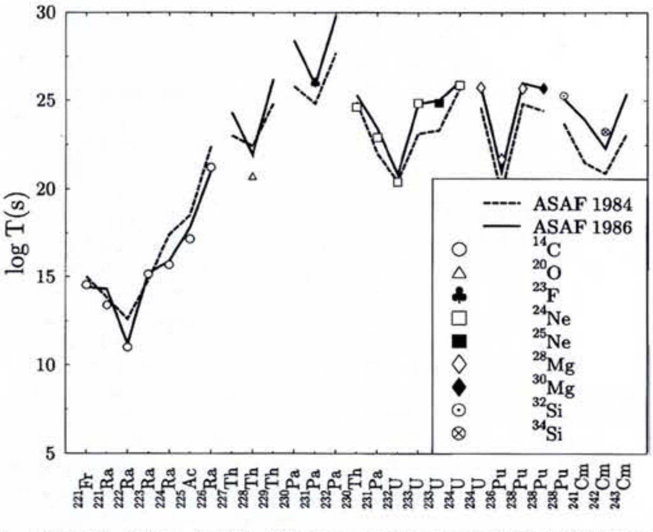

Fig. 2 — A comparison of half-lives T in seconds predicted by the analytical superasymmetric fission (ASAF) model with experimental data for various trans-francium parent nuclei (from ^{221}Fr to ^{242}Cm) against diferent cluster decay modes (from ^{14}C to ^{34}Si clusters). There is little difference between the initial ASAF predictions published in 1984 (dashed line) and the results of more refined calculations published in 1986 (solid line).

cluster decay modes obtained using the ASAF model as well as those reported in 1986 (see Fig. 2). The data are for an island of trans-francium parent nuclei, where the daughter nucleus is the doubly magic ^{208}Pb or one of its neighbours. We have also developed an elegant illustration of the ASAF model's unified approach by describing cold-fission cluster radioactivity and α-decay of ^{234}U, for which all these various decay modes have been observed experimentally. In addition, we have studied many other cold and bimodal fission processes in terms of cluster decay. The recent observation at the Joint Institute of Nuclear Research, Dubna, and at the GSI, Darmstadt, of the ^{12}C radioactivity of ^{114}Ba confirms the existence of an island of proton-rich cluster emitters with daughters around ^{100}Sn which we predicted in 1984. It is worthwhile to note that several groups have developed alternative approaches by extending either the fission theory to a larger asymmetry or the traditional many-body model of α-decay to heavier emitted clusters.

Fine Structure

The main experimental difficulty in observing cluster radioactivity comes from the need to identify a few rare events among an enormous number of background α-particles. Solid-state track detectors (which are insensitive to α-particles) and magnetic spectrometers (in which α-particles are deflected by a strong magnetic field) have been used to overcome this difficulty. The superconducting spectrometer SOLENO at the Institut de Physique Nucleaire, Orsay, has been employed since 1984 to detect and identify the ^{14}C clusters emitted spontaneously from ^{222,223,224,226}Ra parent nuclei. Moreover, its excellent energy resolution was exploited in 1989 to discover [4] a fine structure in the kinetic energy spectrum of ^{14}C emitted by ^{223}Ra. This type of cluster emission, which leads to excited states of the final fragments, was considered theoretically for the first time in 1986 by Greiner and Scheid.

Predicted Hindered Transitions

^{24}Ne decay of ^{233}U (5/2$^+$ ground state) to the 9/2$^+$ gs of ^{209}Pb	$H = 44$
^{23}F decay of ^{231}Pa (3/2$^-$ gs) to the 0$^+$ gs of ^{208}Pb	$H = 12$
^{14}C decay of ^{221}Fr (5/2$^-$ gs) to the 1/2$^+$ gs of ^{207}Tl	$H = 8.5$
^{14}C decay of ^{221}Ra to the 1/2$^-$ gs of ^{207}Pb	$H = 9$
^{24}Ne decay of ^{231}Pa (3/2$^-$ gs) to the 1/2$^+$ gs of ^{207}Tl	$H < 5$
^{14}C decay of ^{225}Ac (3/2$^-$ gs) to the 9/2$^-$ gs of ^{211}Bi	unhindered?

Following the discovery of the fine structure of the ^{14}C radioactivity of ^{223}Ra, it was shown that the transition towards the first excited state of the daughter nucleus was more pronounced than that towards the ground state. In other words, as in the spontaneous fission of odd-mass nuclei and in α-decays revealing a fine structure, one has hindered and favoured transitions, respectively. The physical explanation of this phenomenon relies on the single-particle spectra of neutrons or/and protons. The transition is favoured if the uncoupled nucleon is left in the same state in both the parent nuclei and the heavy fragment. If this is not the case, the difference in structure leads to a large hindrance H given by $H = T^{\exp}/T_{\text{e-e}}$, where $T^{\exp}$ is the measured partial half-life for a given transition and $T_{\text{e-e}}$ is the corresponding quantity for a hypothetical equivalent even-even decay, estimated either from systematics (a plot of log T versus $Q^{-1/2}$, for example) or from a model. A transition is favoured if $H \approx 1$, and it is hindered if $H > 5$.

There exists an unique possibility to study the transition from a deformed parent nucleus with complex mixing of pure shell-model wave-functions to a special nucleus with a pure shell-model wave function using the cluster radioactivity of odd-mass nuclides. This is not the case for α-decay since the initial and final states of the parent and daughter are not so very different from one another. One can in fact obtain direct spectroscopic information about spherical components of the deformed states. The most accurate experiment carried out so far was performed in 1994 by Hourany *et al.* using SOLENO together with high-quality ^{223}Ra sources implanted at CERN's ISOLDE detector. The interpretation, given by Sheline and Ragnarsson, was confirmed in that the main spherical component of the deformed parent wave function has an $i_{11/2}$ character (*i.e.*, the main component is spherical).

Work in unifying the theory of cold fission, cluster radioactivity and α-decay, as well as theoretical models and experimental results for cluster radioactivity will be reviewed in *Nuclear Decay Modes* (IOP Publishing, Bristol), to be published. See also: Price P.B., *Ann. Rev. Nucl. Part. Sci.* **39** (1989) 19; Ludu A. *et al.*, *Int. J. Mod. Phys. E* **1** (1992) 169.

The ASAF model can be used to simulate cluster decay in the even-even configuration. By adopting the allowed angular momenta determined from spin and parity conservation we obtained a large hindrance factor ($H = 44$) for ^{24}Ne decay of ^{233}U (5/2$^+$ ground state) to the 9/2$^+$ gs of ^{209}Pb. We have also predicted that other hindered transitions may exist (see insert). The ^{14}C transition from ^{225}Ac (3/2$^-$ gs) to the 9/2$^-$ gs of ^{211}Bi seems not to be hindered.

The decay constant λ (where $\lambda = \ln 2/T$) of the number of parent nuclei which characterises the well-known $\exp(-\lambda t)$ law describing the variation with time t of the rate of decay can be expressed as a product of three model-dependent quantities such that $\lambda = \nu SP$, where ν is the frequency of assaults on the tunnelling barrier, S is the probability that the cluster forms at the nuclear surface (the so-called preformation probability which depends strongly on the nuclear structure), and P is the quantum penetrability of the external part of the potential barrier. In models derived from a many-body description of α-decay, S is expressed as an overlap integral of the wave function of the three partners (parent and two fragments).

The most instructive method of plotting the experimental results uses a universal curve of log T *versus* log P. Instead of having different lines for various parent nuclei as in the Geiger-Nuttal plot of log T versus $Q^{-1/2}$, one obtains a single straight line with a slope equal to unity for a given even-even cluster decay mode. The theoretical model reproduces very accurately the even-even half-life measurements which have been reported to date (within a ratio of 3.86, equivalent to an root-mean square deviation of 0.587 orders of magnitude).

Our extensive study of one-, two- and three-dimensional fission dynamics over a wide range of mass asymmetry has in fact allowed us to identify nuclear shapes during the deformation process. One can establish that some misunderstanding and errors have arisen in the past. The most important have been to ignore the motion of the centre-of-mass in calculating nuclear inertia using the frequently invoked Werner-Wheeler approximation whereby for a hydrodynamic model of the nucleus, deformation is cylindrically symmetric with two independent velocities that obey specific scaling laws.

[1] Poenaru D.N. & Greiner W., in *Nuclear Decay Modes* (IOP Publishing, Bristol), to be published; Greiner W. *et al.*, in *Treatise on Heavy Ion Physics* **8** (Plenum, New York, NY) 1989; Poenaru D.N., Ivascu M. & Greiner W., in *Particle Emission from Nuclei* **3** (CRC, Boca Raton, FL) 1989.

[2] Sandulescu A., Poenaru D.N. & Greiner W., *Sov. J. Part. Nucl.* **11** (1980) 528.

[3] Rose H.J. & Jones G.A., *Nature* **307** (1984) 245.

[4] Brillard L. *et al.*, *Cont. Rend. Acad Sci.* **309** (1989) 1105.