Developments in radioactive decay during the last Century

R. J. Liotta
Department of Physics, Royal Institute of Technology, SE-10691 Stockholm, Sweden
E-mail: liotta@kth.se

Abstract. In this talk a review of the developments in radioactive decay processes that have taken place during the last Century, and the great outgrowths of these developments in Modern Physics, is presented.

1. Historical background
At the beginning of last Century a revolutionary transition in Physics was taking place. Already at the end of the 1800’s the forms of radiation that nowadays are called alpha and beta were discovered. In 1896 Becquerel found that charged particles were emitted by uranium, and soon afterwards Marie and Pierre Curie confirmed these observations. In 1899 Rutherford discovered that these radiations could be divided in two separated strings according to the penetration power they had. He called this two forms of radiation alpha and beta. Alpha radiation could penetrate only thin foils of aluminium while beta rays could go through many millimeters of the same material. Only in 1900 the third form of radiation was found, which had even more penetration power than beta rays, and these were called gamma rays. It was soon found that the alpha ray was actually an alpha particle, that the beta ray was an electron and that the gamma ray was a photon.

The discovery that alpha radiation was actually the emission of $^4$He nuclei was the basis of the Chemistry Nobel prize that Rutherford got in 1908. But Rutherford continued working at a very high level even after receiving the Nobel prize. In fact, he produced his most revolutionary work after receiving the Nobel prize, and that was the discovery of the nucleus. Due to this he was also known as the father of nuclear physics. But Rutherford, who introduced the concept of ”half lives” in relation to decaying systems, was not only a great experimentalist. He was also a great teacher. He had many brilliant students, some of whom became themselves Nobel laurates, like James Chadwick. John Cockcroft, Ernest Walton and Edward Appleton. Others were strongly influenced by him, like Niels Bohr.

Among these brilliant students a number were from Germany, who preferred to visit Rutherford at the University of Manchester rather to be unemployed in the saturated market for physicists in German Universities at that time. Among those students there were H. Geiger and E. Mardsen, who together created the Geiger counter in 1909. At this time Rutherford proposed Geiger and J. M. Nuttall, an English student, to analyze the relation between the half lives of alpha emitting nuclei and their kinetic energies. They thus succeeded in 1911 to find the...
famous Geiger-Nuttall law of alpha radioactive decay [1], i. e.,

$$\log T_{1/2} = \frac{a}{\sqrt{Q_\alpha}} + b$$  \hspace{1cm} (1)

where $a$ and $b$ are $Z$-dependent constants and $Q_\alpha$ is the kinetic energy of the outgoing $\alpha$-particle.

There has been an intense investigation on different aspects of the simple-looking Geiger-Nuttall law lasting for many years after it was formulated. For instance, much work was dedicated to find a generalization of the law valid for all $Z$. Thus, 55 years after Geiger-Nuttall, Viola and Seaborg found that, with a great precision, the dependence of the constants upon $Z$ can be written as [2] $a(Z) = 2.11329Z - 48.9879$, $b(Z) = -0.390040Z - 16.9543$.

But the most important outcome from the many attempts performed in order to explain the origin of the Geiger-Nuttall law was the one due to Gamow, which profoundly changed the formulation and interpretation of physical laws. This we will now analyze.

2. Gamow treatment

Gamow considered the alpha-particle as a compact object which existed in the nucleus. He did not consider that the alpha particle actually consists of other more "elementary" particles, like neutrons and protons. He could not do that simply because the neutron was not discovered at that time yet, i. e. at the end of the 1920’s. Gamow, as well as Rutherford and all other relevant physicist at that time, thought that the alpha particle was a conglomerate of four protons and two electrons. Gamow assumed that these six particles were strongly bound together and that this bunch of tightly bound particles existed inside the nucleus as a whole before decaying. He wrote the Schrödinger equation corresponding to the $\alpha$-daughter system, assuming spherical symmetry and relative angular momentum $l=0$. He thus evaluated the penetrability of the $\alpha$-particle through the Coulomb barrier, ignoring any other interaction [3]. Assuming that the local wave number does not change strongly inside the barrier, the corresponding penetrability is,

$$P = \exp(-2\gamma)$$  \hspace{1cm} (2)

and

$$\gamma = \frac{1}{\hbar} \int_{r_i}^{r_e} \sqrt{2m(Z_1Z_2e^2/4\pi\epsilon_0r - Q)}dr$$  \hspace{1cm} (3)

where the notation is standard. In particular $r_i$ and $r_e$ are the internal and external turning points.

Gamow interpreted the penetrability as the probability that the alpha particle went through the Coulomb barrier. This can not happen classically, since under the barrier the kinetic energy is negative, and therefore the velocity is imaginary. This was a great revolution in physics and confirmed the probabilistic interpretation of Quantum Mechanics, a feature that a number of distinguished physicists found to be weird. This is debated even today [4].

In order to get the right dimensions, Gamow used the very simple argument that the alpha particle is inside the nucleus without interacting with the protons and electrons that he assumed to be the nuclear constituents. That is, he considered the alpha particle as a little ball moving inside the nucleus, hitting the walls induced by the Coulomb barrier when arriving to the nuclear surface, going forward and back again. Within this assumption, the probability decay per second (i. e. the inverse of the mean life) is the probability of getting through the barrier ($P$) times the number of attempts to hit the wall of the nucleus. This number is $v/R$, where $v$ is the velocity of the alpha particle and $R$ is the nuclear radius. Therefore the mean life $T$ is,

$$1/T = \frac{v}{R}P$$  \hspace{1cm} (4)
This expression for the mean life has had a tremendous success in explaining decay processes. One reason for this success is that it is extremely simple and can readily be applied to all forms of radioactive decay. Yet, it is an effective formula, without a firm theoretical background. One important quantity which is not included in Eq. (4) is the probability that the alpha particle is indeed present on the nuclear surface of the decaying nucleus. This probability is called “preformation factor” and is usually denoted by $P_0$. The value of $P_0$ was included in later analysis of the decay and its effective value was obtained through fittings of experimental data. This task was performed through many years of investigation, and the result has been of great help to the experimental interpretation of the decay data. We will see below how the decay can be analyzed within a complete theoretical framework. However, in strong contrast to the effective approach, thorough theoretical treatments are usually very complicated and difficult to apply. Therefore most radioactive decay studies are performed within effective approaches. In these approaches the flux $1/T = \lambda$ is usually called ”decay constant”, and the frequency of escape attempts is denoted by $v/R = \nu_0$. One thus gets,

$$\lambda = \nu_0 P_0 P$$  \hspace{1cm} (5)

A recent study of determinations of $\nu_0$ and $P_0$, obtained through the analysis of many decay processes, can be found in [5].

Gamow also realized that one can describe the decay process within an stationary formalism by assuming that the decaying state carries a complex energy. Thus, if the energy is $E = E_r - i\Gamma/2$, the wave function becomes

$$\varphi(r, t) = \varphi(r, 0) e^{-iEt/\hbar} = \varphi(r, 0) e^{-iE_t/\hbar} e^{-\Gamma t/2\hbar}$$  \hspace{1cm} (6)

and the probability density that the alpha particle (or any cluster) decays is

$$P(r, t) = P(r, 0) e^{-\Gamma t/\pi}$$  \hspace{1cm} (7)

Which provides for the mean life the relation $TT = \hbar$, which sometimes is called, wrongly, the time-energy uncertainty relation.

It is convenient to point out that Eq. (7) would be valid only if all the imaginary parts are small, since only then one can interpret $\text{Re}(\varphi^2(r)) \approx |\varphi^2(r)|$ as a probability. A detailed discussion on this, and on the study of resonances by an extension of the formalism to the complex energy plane, can be found in [6].

So far we have considered effective theories that follow Gamow original formalism. There has also been a fully microscopical development of the theory. This started from the derivation of the width $\Gamma$ as the residues of the R-matrix. In a rather difficult paper Thomas performed the corresponding calculation for the case of alpha decay [7]. But one can also get the width in a simple fashion by following scattering theory arguments [8]. To show this we consider a decaying cluster. At large distances only the centrifugal and Coulomb interactions are felt. Therefore the corresponding wave function is,

$$r^2 \varphi_{ij}^{\text{out}}(r) = N_{ij} [G_{ij}(r) + iF_{ij}(r)]$$  \hspace{1cm} (8)

where the notation is standard. The probability rate per second that the particle goes through a surface element $dS = r^2 \sin \theta d\theta d\phi$ is,

$$P_{ij} = |\varphi_{ij}^{\text{out}}(r)|^2 v dS$$  \hspace{1cm} (9)

where $v$ is the velocity of the cluster at large distances, i. e. $v = \hbar k/m$. Since at large distances $|G_{ij}(r) + iF_{ij}(r)|^2=1$, one obtains, by integrating over the angles, the decay
probability per second as $1/T = |N_{ij}|^2v$. Matching the out and the inner solution at $R$ one gets $R\varphi_{ij}(R) = N_{ij}[G_{ij}(R) + iF_{ij}(R)]$ and

$$\Gamma_{ij} = \frac{\hbar}{T} = \frac{\hbar k}{m} \frac{R^2|\varphi_{ij}(R)|^2}{F_{ij}^2(R) + G_{ij}^2(R)}$$

This is the famous Thomas expression for the decay width, which he obtained as the residues of the R-matrix. $\varphi_{ij}(R)$ is the cluster formation amplitude and $kR/(F_{ij}^2(R) + G_{ij}^2(R))$ is the penetrability through the centrifugal and Coulomb barriers. It is important to notice that the width should not depend upon $R$ if the calculation of the formation amplitude is properly performed. For the cluster (C) decay process $B \rightarrow A + C$ the formation amplitude is

$$\varphi(R) = \int d\xi_A d\xi_C \varphi_B(\xi_B) \varphi_A(\xi_A) \varphi_C(\xi_C)$$

This expression is the basis of all microscopical calculations of cluster decay. We will briefly review below the large amount of work performed through many years in this subject.

3. Shell Model, BCS and Rotational treatments

During the 1950’s a revolution occurred in nuclear physics with the near simultaneous appearance of the Shell Model, the Rotational Model and the BCS formalism. For alpha decay an equally important ingredient was the Thomas formulation leading to Eq. (11). This equation was applied to alpha decay from spherical nuclei by Rassmusen [9], Mang [10] and Sandulescu [19], and in deformed nuclei by Bohr and Mottelson, as described by Froman [20]. The main point of these calculations was the description of the mother nucleus $B$ in terms of the daughter nuclei plus two neutrons and two protons, as described in detail in [21]. For the case in which the daughter nucleus $A$ is a magic core the mother wave function can be written as,

$$\varphi_B(\xi_B) = \sum_{\pi\nu} X(\pi\nu; B)[\phi_{A+2}^\pi(\xi_\pi)\phi_{A+2}^\nu(\xi_\nu)]_B \phi_A(\xi_A)$$

For superconducting nuclei this equation is also valid but the amplitudes are,

$$\phi_{A+2}(\xi) = \sum_{\Omega_1 < \Omega_2} \sum_{l_1 + l_2 = A} u^A_{\Omega_1} u^B_{\Omega_2} [\varphi_{\Omega_1}(r_1) \varphi_{\Omega_2}(r_2)]$$

For spherical nuclei this expression is relatively easy to compute, but for a deformed nucleus one has to take into consideration the different directions that the decay process may choose. The corresponding formalism is very involved, as can be seen in Ref. [20]. Moreover, the approximations involved in the derivations of the corresponding equations in this case implies that the results are not reliable for large deformations, as seen in Fig. 1.

With the computing facilities of today it is more convenient to expand the wave function in spherical waves. The l-spherical component of the deformed wave function, $g_l$, satisfies the equation,

$$\left\{ -\frac{\hbar^2}{2m_\alpha} \frac{d}{dr^2} + \frac{\hbar^2 l(l+1)}{2m_\alpha r^2} \right\} g_l(r) + \sum_\nu V_{\nu l}(r) g_\nu(r) = E_\alpha g_l(r)$$

At large distances the l-component of the outgoing wave function has the form,

$$S^{(k)}_l(R) \rightarrow \delta_{kl}[G_l(R) + iF_l(R)]$$

and the deformed wave function at $R$ is

$$g_l(R) = \sum_k S^{(k)}_l(R) C_k \rightarrow C_l[G_l(R) + iF_l(R)]$$
Figure 1. Ratio between the theoretical and experimental decay widths corresponding to the transition $^{152}\text{Dy} \rightarrow ^{148}\text{Gd} + \alpha$ as a function of the quadrupole deformation parameter. The width $\Gamma_{WKB}$ corresponds to the expression given in Ref. [20], while $\Gamma_{cc}$ is the one provided by the coupled channel equations (14). (Taken from reference [23]).

The fitting of the internal wave function $g_l(R)$ and the external one, $G_l(R) + iF_l(R)$, allows one to evaluate $C_l$ and the corresponding decay width, as in the derivation of Eq. (10).

A detailed description of these equations, and some applications, can be found in Ref. [22].

The first microscopical calculations of alpha decay were wrong by some six orders of magnitude [9, 10]. This pioneers in the microscopic treatments of alpha decay realized that the reason of this huge difference was that, due to computing limitations, only one configuration was included in the calculations. It was found much later that that was indeed the case, and that the inclusion of high lying configurations increased the value of the decay width by five orders of magnitude [11]. At about the same time it was realized that the reason of this increase was due to the clustering of the two neutrons and two protons, which eventually constitute the alpha particle, induced by the pairing force [12]. Yet the decay width was still too small. It was thought that this was due to the neutron-proton interaction, which was not taken into account in that work. However, it was shown that even including the neutron-proton interaction the decay width was too small by about one order of magnitude. This indicates that there is an intrinsic component of preformed alpha particle in the wave function, as indeed was shown in Ref. [13]. This feature was recently exploited to describe the decay pattern of $^{212}\text{Po}$, where electromagnetic transitions compete with alpha decay in some cases [14].

There have been attempts to generalize the Geiger-Nuttall law including all types of radioactive decays, i.e. alpha, proton and cluster emissions as well as all possible angular momenta. This has not been achieved yet. However important steps in this direction have been taken. In particular, generalizations of proton decay laws [15] as well as of alpha and cluster decays [16, 17, 18] have been presented recently.

4. Cluster decay
In the 1970s a breakthrough in radioactive decay was achieved by the theoretical prediction of cluster decay. This was possible through a formalism developed by A. Sandulescu for radioactive decay as an extension of the one corresponding to nuclear fission [24]. This is not surprising, since decay and fission are similar processes which are determined by the Coulomb and centrifugal barriers. It was through the analysis of the penetrability shown in Fig. 2 that the decay of $^{224}\text{Ra}$ by emission of a $^{14}\text{C}$ cluster as well as the emission of $^{24}\text{Ne}$ clusters from $^{230}\text{Th}$ was predicted. This was because, as seen in the figure, the penetrability corresponding to the decay of these
clusters is large.

Figure 2. Logarithm of the penetrability as a function of the cluster mass corresponding to the systems $^{224}Ra + ^{14}C$ and $^{230}Th + ^{24}Ne$. (Taken from Ref. [25].)

This prediction was confirmed experimentally several years afterward by Rose and Jones [26]. Naturally, it also induced many works performed to analyze this rare form of decay as well as the features in the nuclear structure that make the decay possible. It is beyond the scope of this talk to go through these developments in detail. For a review see Ref. [27]. However, it is worthwhile to mention that one of these investigations showed that the formation amplitudes of clusters (including protons) decreases exponentially with the mass of the cluster. This amplitude is of the order unity for protons, $10^{-2}$ for alpha particles, $10^{-4}$ for $^8Be$, $10^{-5}$ for $^{14}C$ and $10^{-19}$ for $^{46}Ar$ [28].

One remarkable property of the penetrability shown in Fig. 2 is that it is extremely large, at least as large as for alpha decay, for clusters with mass number $A=80$. It is therefore not surprising the recent prediction that heavy clusters should be emitted with higher probability than alpha particles from superheavy nuclei [29]. This is an important property, since it may help to identify superheavy nuclei just by detecting the emitted heavy cluster.

Another important development in cluster radioactivity was the formulation of a model where the cluster (including alpha-particle) was assumed to move around the daughter nucleus (the core) under the influence of a central field. This potential was assumed to be very deep and therefore a harmonic oscillator approximation was adopted. Thus the cluster wave function contains a number of nodes given by the equivalent of the harmonic oscillator quantum number $n$. The principal quantum number is called $G$ (global) and its value is $G=2n+l$. For decays into the lead region it is assumed $G \approx 20$. There have been many publications related to this very successful model, which has been used even to study molecular structures in nuclei, as can be seen, e. g., in Refs. [30, 31] and references therein.

5. Summary
The study of radioactive decay processes have been fundamental in physics. To name but a few of its achievements one can mention,

It established the fundamental principle of the probabilistic interpretation of quantum mechanics.

It extended the study of physical processes to complex energies, This includes the introduction of propagators and S-matrix theories through which the treatment of resonances was possible.
For the case of many-body resonances one can mention the relatively recent shell model in the complex energy plane.

It has been a powerful tool to investigate the structure of nuclei and the correlations leading to nuclear clustering.

It has allowed one to determine the quantum numbers of nuclear levels and it seems that, in the near future, will also be a powerful tool to identify exotic, very heavy nuclei.

References
[1] Geiger H and Nuttall J M 1911 Philos. Mag. 22 613
[2] Viola V E and T. Seaborg G T 1966 J. Inorg. Nucl. Chem. 28 741
[3] Gamow G 1928 Z. Phys. 51 204
[4] Hooft G, 2002 in Conf. Proceedings "Quo Vadis Quantum Mechanics", Philadelphia arXiv : quant – ph/0212095
[5] Zhang H F, Roger G and Li J Q 2011 Phys. Rev. C 84 027303
[6] Betan R Id, Liotta R J, Sandulescu N, Vertse T and Wyss R 2005 Phys. Rev. C 72 054322
[7] Thomas R G 1954 Prog. Theor. Phys. 12 253
[8] Maglione E, Ferreira L S and Liotta R J 1998 Phys. Rev.mLett. 81 538
[9] Rassmusen J O 1965 Alpha-, Beta- and Gamma-Ray Spectroscopy vol. 1 (North-Holland, Amsterdam)
[10] Maglione E, Ferreira L S and Liotta R J 1998 Phys. Rev. Lett. 103 073501