## Universal Decay Law in Charged-Particle Emission and Exotic Cluster Radioactivity

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A linear universal decay formula is presented starting from the microscopic mechanism of the chargedparticle emission. It relates the half-lives of monopole radioactive decays with the Q values of the outgoing particles as well as the masses and charges of the nuclei involved in the decay. This relation is found to be a generalization of the Geiger-Nuttall law in  $\alpha$  radioactivity and explains well all known cluster decays. Predictions on the most likely emissions of various clusters are presented.

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The first striking correlation between the half-lives of radioactive decay processes and the Q values of the emitted particle was found in  $\alpha$ -decay systematics by Geiger and Nuttall [1] as,

$$\log T_{1/2} = a Q_{\alpha}^{-1/2} + b, \tag{1}$$

where *a* and *b* are constants. However, the Geiger-Nuttall law in the form of Eq. (1) has limited prediction power since the coefficients *a* and *b* change for the decays of each isotopic series [2]. It may also change within a single isotopic chain when magic numbers are crossed [3]. Intensive works have been done trying to generalize the Geiger-Nuttall law for a universal description of all detected  $\alpha$  decay events [4–9]. Here we present a truly universal formula valid for the radioactivity of all clusters, including  $\alpha$  particles. This will allow us to search for new cluster decay modes and to carry out a simple and modelindependent study of the decay properties of nuclei over the whole nuclear chart.

We thus observe that the Q value dependence in Eq. (1) is a manifestation of the quantum penetration of the  $\alpha$ cluster through the Coulomb barrier. But this equation ignores the probability that the  $\alpha$  particle is formed on the nuclear surface starting from its four constituent nucleons moving inside the mother nucleus. This is the cause of the limitations of the Geiger-Nuttall law mentioned above. In general the decay process, ranging from proton to heavier cluster radioactive decays, can be described by a two-step mechanism [10]. In the first step the formation of the particle and its motion on the daughter nuclear surface is established. In macroscopic models the clusterization process is described by effective quantities adjusted to reproduce as many measured half-lives as possible. This procedure has shown to be very fruitful, providing a guide to experimental searches. In the second step the cluster, with the formation amplitude and corresponding wave function thus determined, is assumed to penetrate the centrifugal and Coulomb barriers [3-7,11]. The second step has been well understood since the pioneering work of Gamow. It is in fact one of the pillars of the probability

interpretation of quantum mechanics [11]. Its great importance in radioactive decay studies lies in the fact that within a given cluster the penetrability process is overwhelmingly dominant. This explains the great success of macroscopic models in describing radioactive decay.

We intend here to include the cluster formation probability as well as the corresponding penetration through the Coulomb barrier. We start from the *R*-matrix description of the cluster decay process [10,12]. This is the basis of all microscopic calculations of cluster decay [13]. The corresponding decay half-life is,

$$T_{1/2} = \frac{\hbar \ln 2}{\Gamma_c} \approx \frac{\ln 2}{\nu} \left| \frac{H_l^+(\chi, \rho)}{RF_c(R)} \right|^2, \tag{2}$$

where  $\nu$  is the outgoing velocity of the emitted particle which carries an angular momentum l. R is a distance around the nuclear surface where the wave function describing the cluster in the mother nucleus is matched with the outgoing cluster-daughter wave function. For the distance R we will take the standard value, i.e., R = $R_0(A_d^{1/3} + A_c^{1/3})$  where  $A_d$  and  $A_c$  are the mass numbers of the daughter and cluster nuclei, respectively.  $H_1^+$  is the Coulomb-Hankel function and its arguments are standard, i.e.,  $\rho = \mu \nu R/\hbar$  and the Coulomb parameter is  $\chi =$  $2Z_c Z_d e^2/\hbar\nu$  with  $\mu$  being the reduced mass and  $Z_c$  and  $Z_d$  the charge numbers of the cluster and daughter nucleus, respectively. The quantity  $F_c(R)$  is the formation amplitude of the decaying cluster at distance R. The penetrability is proportional to  $|H_l^+(\chi, \rho)|^{-2}$ . Equation (2) is valid for all clusters and for spherical as well as deformed cases. The ratio  $N_I = RF_c(R)/H_I^+(R)$ , and therefore the half-life itself, is independent of the radius R [13].

In microscopic theories the formation amplitude is evaluated starting from the single-particle degrees of freedom of the neutrons and protons that eventually become the cluster. This is generally a formidable task which requires advanced computing facilities as well as suitable theoretical schemes to describe the clusterization process. It is therefore not surprising that the first calculations of abso-

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lute decay widths (which require a proper evaluation of the formation amplitude) were performed after the appearance of the shell model. These calculations had limited success due to the small shell model spaces that could be included at that time. Only later, with better computing facilities, the calculated half-lives started to approach the corresponding experimental values. We will not deal with microscopic theories here. For details and references on this subject, including a historical background, see Ref. [13].

Our aim is to find few quantities that determine the halflife. Expanding in these quantities we hope to be able to find, at the lowest order of perturbation, an expression of the half-life which is as simple as the Geiger-Nuttall law but valid in general, i.e., for all isotopic series as well as all type of clusters. With this in mind we notice that the Coulomb-Hankel function can be well approximated by an analytic formula, which for the l = 0 channel reads [14],

$$H_0^+(\chi,\rho) \approx (\cot\beta)^{1/2} \exp[\chi(\beta - \sin\beta\cos\beta)], \quad (3)$$

where the cluster Q value is  $Q_c = \mu \nu^2/2$  and

$$\cos^2\beta = \frac{Q_c R}{e^2 Z_c Z_d}.$$
(4)

One sees that  $\cos^2\beta$  would be a small quantity if  $Z_cZ_d$ is large, i.e., for heavy and superheavy systems. In this case one can expand the last term in a power series of  $\cos\beta$ . By defining the quantities  $\chi' = Z_cZ_d\sqrt{A/Q_c}$  and  $\rho' = \sqrt{AZ_cZ_d(A_d^{1/3} + A_c^{1/3})}$  where  $A = A_dA_c/(A_d + A_c)$ , one gets, after some simple algebra,

$$\log T_{1/2} = a\chi' + b\rho' + \log\left(\frac{\cot\beta \ln 2}{\nu R^2 |F_c(R)|^2}\right) + o(3), \quad (5)$$

where  $a = e^2 \pi \sqrt{2m}/(\hbar \ln 10)$  and  $b = -4e\sqrt{2mR_0}/(\hbar \ln 10)$  are constants (*m* is the nucleon mass). The first two terms dominate the Coulomb penetration and o(3) corresponds to the remaining small terms. But still the strong dependence of the half-life upon the formation probability in the third term of Eq. (5) has to be taken into account. It is very difficult to make a microscopic calculation of the formation amplitude  $F_c(R)$ . But we can extract it from the experimental half-lives data by using Eq. (2), i.e.,

$$\log|RF_c(R)| = \frac{1}{2} \log\left[\frac{\ln 2}{\nu}|H_0^+(\chi,\rho)|^2\right] - \frac{1}{2} \log T_{1/2}^{\text{Expt.}}.$$
(6)

Taking  $R_0 = 1.2$  fm we evaluated the function  $\log |RF_c(R)|$  corresponding to  $\alpha$  as well as heavier clusters. We thus found that the formation probabilities of  $\alpha$  decays are located in the range  $\log |RF_c(R)| = -1.5$  to

-0.75 fm<sup>-1/2</sup>. The stability of the  $\alpha$  decay formation amplitude explains the success of the Geiger-Nuttall and other empirical laws where formation mechanism is not explicitly embedded. However, for all observed cluster decays, ranging from  $\alpha$  to the heavier <sup>34</sup>Si, the formation amplitude changes as much as 8 orders of magnitude.

Yet we found that Eq. (5) can still be written as a simple linear formula which properly takes into account the strong dependence of the formation amplitude upon the cluster as well as the mother nuclear structure to a first order of approximation. This we have achieved by exploiting the property that for a given cluster  $N_0 \equiv RF_c(R)/H_0^+(\chi, \rho)$ does not depend upon *R*. Proceeding as above one readily obtains the relation,

$$\log|RF_{c}(R)| \approx \log|R'F_{c}(R')| + \frac{2e\sqrt{2m}}{\hbar\ln 10}(\sqrt{R'_{0}} - \sqrt{R_{0}})\rho',$$
(7)

where  $R' = R'_0(A_d^{1/3} + A_c^{1/3})$  is a value of the radius that differs from *R*. Since for a given cluster any nuclear structure would be carried by the terms  $RF_c(R)$  and  $R'F_c(R')$  in exactly the same fashion, Eq. (7) implies that the formation amplitude is indeed linearly dependent upon  $\rho'$ . Therefore one can write,

$$\log T_{1/2} = a\chi' + b\rho' + c.$$
 (8)

We emphasize here that the coefficient *b* in this relation is different from that of Eq. (5). That is, the terms  $b\rho' + c$ , which do not depend upon  $Q_c$ , have to include the effects that induce the clusterization in the mother nucleus. Moreover, we found that the term  $\log \cot \beta / \nu$  in Eq. (5) varies only slightly for all the cases investigated below, from a minimum of 0.94 to a maximum of 1.2. The effects induced by this variation, as well as the higher-order terms in Eq. (5), are to be taken into account by a proper choosing of the constants *a*, *b* and *c*.



FIG. 1 (color online). UDL plots for the  $\alpha$  decays of even-even nuclei with Z = 78 - 118. The straight line is given as  $a\chi' + c$ .

Equation (8) holds for all cluster radioactivities. We will call this relation the universal decay law (UDL). A straightforward conclusion from the UDL is that  $\log T_{1/2}$  depends linearly upon  $\chi'$  and  $\rho'$ . For this to be valid, it should include the Geiger-Nuttall law as a special case. One sees that this is indeed the case since  $\rho'$  remains constant for a given  $\alpha$ -decay chain and  $\chi' \propto Q_c^{-1/2}$ . Below we will probe these conclusions, and the approximations leading to them.

We will analyze g.s. to g.s. radioactive decays of eveneven nuclei. We select 139  $\alpha$  decay events from emitters with  $78 \le Z \le 108$  for which experimental data are available. We take the data from the latest compilations of Refs. [15,16] and the lists of Refs. [17,18]. For the decay of heavier clusters we have selected 11 measured events ranging from <sup>14</sup>C to <sup>34</sup>Si [19]. In order to perform the calculations one has first to determine the values of the constants a, b, and c. We carried out an extensive search of the best values for these free parameters. Using a fitting procedure for the case of  $\alpha$  decay we obtained a = 0.4065, b = -0.4311, and c = -20.7889. The quality of the adjustment thus obtained can be seen in Fig. 1, where the values of  $\log T_{1/2} - b\rho'$  as a function of  $\chi'$  is shown. The UDL reproduces the available experimental half-lives within a factor of about 2.2. This compares favorably with modern versions of the Geiger-Nuttall law [8].

A significant deviation of the UDL in the figure is the nucleus  ${}^{254}$ Rf, at  $\chi' = 132.16 \text{ MeV}^{-1/2}$ , for which only the lower limit of the half-life is available. This nucleus has the value  $T_{1/2} > 1.5$  ms experimentally [15]. The half-life given by the UDL is  $T_{1/2} = 42$  ms, corresponding to a branching ratio of b = 0.055%. A more precise measurement of this half-life would be a welcome additional test of the UDL.

We will now analyze cluster decay processes by comparing the predictions of the UDL with the experimental data corresponding to the decay of the even-even nuclei mentioned above [19]. Using the parametrization set II in Table I we plotted, as before, the quantity  $\log T_{1/2} - b\rho'$  as a function of  $\chi'$ . As seen in the left part of Fig. 2 the agreement between experiment and the UDL is excellent. The UDL reproduces the available experimental half-lives within a factor of about 4.1.

TABLE I. Coefficient sets of Eq. (8) determined by fitting to experimental data in  $\alpha$  decay (I), cluster decay (II) and both  $\alpha$  and cluster decays (III), respectively. The last column is given by the Coulomb barrier penetration term of Eq. (5) with  $R_0 = 1.2$  fm.

	$I(\alpha)$	II(cluster)	III( $\alpha$ + cluster)	IV
a	0.4065	0.3671	0.3949	0.4314
b	-0.4311	-0.3296	-0.3693	-0.5015
С	-20.7889	-26.2681	-23.7615	

Finally we consider all decays together, i.e.,  $\alpha$  as well as heavier clusters. Using the parameter set III of Table I we obtained the results shown in the right panel of Fig. 2. Again the agreement between the UDL and experiment is excellent.

Using the UDL it is straightforward to evaluate the halflives of all cluster emitters throughout the nuclear chart if reliable values of the binding energies are provided. This we obtain by using the latest compilation of nuclear masses [16]. With the Q values thus obtained we have evaluated the decay half-lives of all isotopes included in that compilation by applying the UDL. We thus found that in all cases the experimental values lie between the ones calculated by using the parameters of the sets I and III in Table I, confirming the prediction power of the UDL. We also found that nuclei favoring cluster decays are mostly located in the trans-lead region.

In Table I we also give the values of the coefficients a and b as provided by Eq. (5). It can be seen that these values are close to the corresponding fitted values, confirming that effects induced by  $\log \cot \beta / \nu$  and higher-order terms in Eq. (5) are small.

In summary, we have presented in this Letter a simple formula that provides with great precision the half-lives corresponding to cluster decay. The formula is valid for all kind of clusters and for all isotopic series, as expected since we derived it from the general description of the decay half-life. This formula is of a universal validity and therefore we call it universal decay law (UDL). There are a few exceptions to this feature, in particular, the alpha-decay of <sup>254</sup>Rf for which only the lower limit of the half-life is available. The UDL predicts that this half-life should be  $T_{1/2} = 42$  ms. A measurement of this number, as well as other cases presented in this Letter for heavy and superheavy nuclei which may be of interest in present experimental facilities, would be most welcome to probe the extension of validity of the UDL. This law may also help in the ongoing search of new cluster decay modes from superheavy nuclei.



FIG. 2 (color online). Same as Fig. 1 but for the heavier cluster decays (left panel) and both  $\alpha$  and cluster decays (right panel).

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