Theory of cluster radioactive decay and of cluster formation in nuclei

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A new model is proposed for the mechanism of cluster formation and then penetration of the confining nuclear interaction barrier in radioactive nuclei. The cluster formation is treated as a quantum-mechanical fragmentation process and the WKB penetrability is found analytically. Applications of the model are made to ^{14}C decay of $^{222-224}$Ra and ^{24}Ne decay of ^{232}U. The branching ratio for ^{14}C decay of ^{232}U is also calculated and is found to be incredibly small as compared to that for its ^{24}Ne decay.

I. INTRODUCTION

Spontaneous decay of radioactive nuclei via emission of clusters heavier than the α particle is now well established experimentally.[1] Emission of ^{14}C from ^{222,223,224,226}Ra and ^{24}Ne from ^{230}Th, ^{231}Pa, and ^{232}U are observed and their branching ratios with respect to α particle are measured.[2-10] Knowing that radioactive nuclei also fission spontaneously, this new phenomenon can either be simply a case of strongly asymmetric fission or an exotic process of cluster formation and escape from the parent nucleus by making many assaults on the confining barrier, similar to α decay. Thus, all the theoretical attempts made so far stem from either Gamow theory of nuclear α decay or the nuclear fission.

In Gamow theory applied to heavy cluster decay, though some very recent calculations[11] do consider the complete two-step mechanism of first formation and then tunneling, the early calculations were made simply for the one-dimensional WKB penetrability through a Coulomb-potential-plus-square-well nuclear potential of width $R=r_0(A_1^{1/3}+A_2^{1/3})$. Here A_1 and A_2 are, respectively, the mass numbers of the daughter nucleus and of the emitted cluster. Many such calculations have been made[2,5,7] to fit empirically the Gamow penetrability factor to the measured data. The resulting empirical values of the effective nuclear radii, however, are not incommensurate with our present day knowledge of nuclear sizes. Another WKB penetrability calculation due to Shi and Swiatecki[12] is based on the potential barrier determined in a two-spheres approximation as a sum of Coulomb and nuclear proximity potential for $R \geq R_t$, where R_t defines the touching configuration of two spheres. For the overlap of two spheres ($R < R_t$), down to the radius R_0 of the parent nucleus, these authors use the simple power-law interpolation and calculate the WKB penetrability factors for the turning points corresponding to potential energies equal to Q value of the reaction. Inclusion of the proximity potential is found to make the penetrability ratios several orders of magnitude smaller than for a pure Coulomb barrier. Notice that, as in Gamow factor calculations, these calculations also assume the preformation factor to be unity for both the heavy clusters and α-particle emissions. In this sense, these works treat the heavy cluster emission as a fission process.

The first quantitative calculations of the decay constants (λ) for this new nuclear process were made by Săndulescu *et al.*[13] and Poenaru *et al.*[14-17] In their improved version, these authors[15] use an analytical superasymmetric fission model (ASAFM) that is fitted to the extensive data on α-decay half-lives ($T_{1/2}$) as well as to the ^{14}C decay of ^{223}Ra. The ASAFM gives an analytical expression for half-life, calculated as the WKB penetration probability through a barrier $V(R)$ approximated by a second-order polynomial in the separation distance R for the overlap of two spheres from the parent nucleus radius R_0 up to the touching point R_t, and by a Coulomb (V_C) plus centrifugal (V_l) potential for the touching configuration $R=R_t$. The half-life for a metastable system ($Q>0$) is defined as

$$T_{1/2}=\frac{\ln 2}{\lambda}=\frac{\ln 2}{\nu P}\ , \tag{1}$$

where the assault frequency (escape attempts) ν in ASAFM is given in terms of *empirical* zero-point vibration energy $E_{\rm vib}$, as

$$\nu=\frac{\omega}{2\pi}=\frac{2E_{\rm vib}}{h} \tag{2}$$

with

$$E_{\rm vib}=Q\left[0.056+0.039\exp\left[\frac{4-A_2}{2.5}\right]\right] \quad \text{for } A_2\geq 4\ , \tag{3}$$

and P is the WKB penetrability, calculated at the turning points corresponding to the potential energies equal to $Q'=Q+E_{\rm vib}$. From a systematic study,[16] these authors find that for the trans-lead nuclei the decay rates are largest ($T_{1/2}$ smallest) for clusters leaving the daughter nuclei to be a magic (^{208}Pb) or almost magic nucleus. In other words, the shell effects are found to play the most significant role in selecting out the new decay modes.

Very recently, a couple of theoretical papers have also included the clustering formation factor in their work. Landowne and Dasso[18] studied the effects of residual cou-

pling interactions to the low-lying excited states of the daughter nucleus (^{208}Pb) and estimated the preformation factor for ^{14}C relative to that for the α particle. For decay into excited states of the daughter nucleus, another model, called excitation model, is developed by Greiner and Scheid.[19] Both these works show an order of magnitude enhancement in the ground-state transmission probability, similar to what Poenaru *et al.*[15] obtained by including the emission from excited states by simply redefining $Q'=Q+E_{\rm vib}+E^*$; here E^* is the fraction of excitation energy concentrated in this collective mode leading to separation. Landowne and Dasso[18] have also investigated a new mechanism of ^{12}C cluster transforming into ^{14}C by picking up two neutrons while tunneling out. Their estimate gives the preformation factor of over 100 for ^{12}C relative to ^{14}C. This result, however, is not surprising in view of a systematic calculation by Iriondo *et al.*[11] who have shown that the formation amplitudes diminish with increase of cluster size. Within the Gamow theory, these authors[11] have for the first time studied both the steps of first formation and then penetration through the Coulomb barrier. Based on their experience with microscopic calculations for α-cluster formation inside the nucleus (by using large shell model configuration spaces with neutron-neutron and proton-proton pairing interactions included), they defined the heavy-cluster formation probability relative to α particle simply as ratio of penetrability of the Coulomb barrier at $r_0=0.98$ to that at $r_0=1.2$ fm. A more direct method of calculating the preformation factor by using the shell model wave functions for ^{14}C and Pb nuclei, and allowing for pairing correlations in the wave functions for the Ra nuclei, is given by Blendowske *et al.*[20] Roughly speaking, the preformation probability (also called the spectroscopic factors by these authors) is proportional to the squared product of the overlaps between the nucleon states in the emitted fragment (the daughter nucleus) and the last 4 or 14 states in the ground-state wave function of the parent nucleus.

In this paper, we propose a new mechanism for the clustering formation in nuclei, as the quantum mechanical fragmentation process, and give an analytical method for calculating the WKB penetrability of the confining nuclear interaction barrier. Without any fitting parameter, our calculated branching ratios and half lives compare reasonably well with experiments and our calculations of the relative cluster preformation probabilities agree within an order of magnitude with other available calculations and estimates. Our model of decay process, decoupled in two steps of clustering formation and tunneling of the interaction barrier, is described in Sec. II. The results of our calculations and a summary of our results are given, respectively, in Secs. III and IV.

II. THE MODEL

We consider (i) the formation of two fragments (the cluster and the daughter nucleus) in their ground states with probability P_0 and (ii) the tunneling of the confining nuclear interaction barrier $V(\eta,\eta_Z,R)$ with probability P by impinging on it with frequency ν. The step (i) is described by introducing the dynamical collective coordinates of mass and charge asymmetries of the two fragments, defined[21–25] as

$$\eta=\frac{A_1-A_2}{A} \quad \text{and} \quad \eta_Z=\frac{Z_1-Z_2}{Z} \ . \tag{4}$$

Step (ii) refers to the motion in the R coordinate. In principle, these two steps, describing the η (and η_Z) and R motions, are coupled. However, it has been shown by Gupta and co-workers[26,27] that the coupling effects of relative motion to asymmetry coordinates in the potential are very small, at least for fission charge distributions and α-particle transfer resonances. The potentials are calculated by using the two-center shell model (TCSM) or the experimental binding energies. Also, it is known[21–23] that the cranking coupling masses $B_{R\eta}$ and $B_{R\eta_Z}$ are very small such that $B_{R\eta}\ll(B_{RR}B_{\eta\eta})^{1/2}$ and $B_{R\eta_Z}\ll(B_{RR}B_{\eta_Z\eta_Z})^{1/2}$ hold good. In view of these results, we treat the two motions in a decoupled approximation and define the decay constant

$$\lambda=P_0\nu P \ . \tag{5}$$

Knowing λ, we can calculate the decay width Γ ($=\hbar\lambda$) or the half-life time $T_{1/2}$ ($=\ln 2/\lambda$). In the following subsections we describe the methods used to calculate the three quantities of Eq. (5).

A. The clustering formation probability P_0

We define the preformation probability as a quantum-mechanical probability of finding the fragments A_1 and A_2 (with fixed charges Z_1 and Z_2, respectively) at a point R of the relative motion. For this purpose we solve the stationary Schrödinger equation in η at fixed η_Z and R:

$$\left[-\frac{\hbar^2}{2\sqrt{B_{\eta\eta}}}\frac{\partial}{\partial\eta}\frac{1}{\sqrt{B_{\eta\eta}}}\frac{\partial}{\partial\eta}+V(\eta,\eta_Z,R)\right]\psi^{(\omega)}_{R\eta_Z}(\eta) = E^{(\omega)}_R\psi^{(\omega)}_{R\eta_Z}(\eta) \ . \tag{6}$$

Then, the probability of finding the mass fragmentation η at the position R and with fixed η_Z on the decay path is proportional to $|\psi^{(\omega)}_{R\eta_Z}(\eta)|^2$. This probability, when scaled to a fractional mass yield at a mass number, say, A_1 of one fragment ($d\eta=2/A$), gives for the ground state ($\omega=0$), the formation probability

$$P_0(A_1)=|\psi^{(0)}_{R\eta_Z}(A_1)|^2\sqrt{B_{\eta\eta}(A_1)}\frac{2}{A} \ . \tag{7}$$

The quantum number $\omega=0,1,2,\ldots$ counts the vibrational states $\psi^{(\omega)}_{R\eta_Z}$ in the potential V. In the present model, we have considered the fragments to be formed in the ground state. This means that the clustering preformation process is assumed to be an elastic process. However, it may be noted that excited state ($\omega\neq 0$) are also possible which modify the internal structure of the fragments (and hence the formation probability) due to the Pauli principle. It has not been feasible to include such effects even in a microscopic calculation by Blendowske *et al.*[20]

In our collective model description, for fission and some heavy-ion collision studies[21,22,25] we have allowed the possible consequences of such excitations by assuming a Boltzmann-type occupation of excited states, but for the cluster-decay process this introduces other unmanageable complications (e.g., see Sec. II B).

The R and η_Z dependences in (6) enter through both the potential and mass parameters. We fix η_Z by minimizing the potential in this coordinate. In other words, we assume the decay process to take its path through the bottom of the valley of the potential in η_Z (such potential energy surfaces are always[22–25] of nearly the harmonic oscillator shapes). Then, the fragmentation potential $V(\eta,R)$ in (6), is calculated by using the two-spheres approximation. This approximation simplifies the calculations and is justified in view of our earlier calculations of $V(\eta)$ at different R values, using TCSM in the Strutinsky method for the overlap regions and the experimental binding energies for asymptotic R (see, e.g., Fig. 5 in Ref. 28, Fig. 1 in Ref. 29, or Fig. 1 in Ref. 30). It is shown clearly that the general shape of the potential $V(\eta)$, including the positions of all the potential energy minima, is independent of the choice of R value. (See also Sec. III A for a further discussion). Thus, for two touching (or overlapping) spheres, we define $V(\eta,R)$ as a sum of the experimental binding energies, Coulomb interaction and the proximity potential,

$$V(\eta,R)=-\sum_{i=1}^{2} B_i(A_i,Z_i)+\frac{Z_1Z_2e^2}{R}+V_P \ . \tag{8}$$

As stated above, the charges Z_i are fixed here by minimizing the sum of the two binding energies and Coulomb potential in charge asymmetry coordinate η_Z. The proximity potential V_P is given by Blocki *et al.*[31]

$$V_P=4\pi\gamma b\frac{C_1C_2}{C_1+C_2}\phi(C-C_1-C_2) \tag{9}$$

with nuclear surface tension coefficient

$$\gamma=0.9517[1-1.7826(N-Z)^2/A^2]\ \text{MeV fm}^{-2} \ . \tag{10}$$

The width (diffuseness) of nuclear surface is $b\approx 1$ and the Süssman central radii C_i of fragments, related to the effective sharp radii R_i is

$$C_i=R_i-\frac{b^2}{R_i} \tag{11}$$

For R_i, we use the semiempirical formula

$$R_i=1.28A_i^{1/3}-0.76+0.8A_i^{-1/3} \ . \tag{12}$$

ϕ is the universal function of proximity potential,

$$\phi(\xi)=\begin{cases}-\frac{1}{2}(\xi-2.54)^2-0.0852(\xi-2.54)^3 & \text{for } \xi\le 1.2511 \ , \\ -3.437\exp(-\xi/0.75) & \text{for } \xi\ge 1.2511 \ , \end{cases} \tag{13}$$

where $\xi=(C-C_1-C_2)/b$. For touching configuration of two nuclei $\phi(0)=-1.7817$ (from Table I of Blocki *et al.*[31]).

The angular momentum dependent term V_l $[=\hbar^2l(l+1)/2\mu R^2$, with $\mu=m(A_1A_2/A)$ as the reduced mass, m being the nucleon mass] is not included in Eq. (8) since l values involved here are small ($\sim 5\hbar$) and its contribution to lifetime estimates are also shown to be small.[15,32]

For the mass parameters $B_{\eta\eta}(\eta)$ in (6), we use the classical model of Kröger and Scheid.[33] This model gives a simple analytical expression whose predictions are shown to compare nicely with microscopic calculations.[34] The model gives

$$B_{\eta\eta}=\frac{AmR^2}{4}\left[\frac{v_t(1+\beta)}{v_c(1+\delta)^2}-1\right]$$

with

$$\beta=\frac{R_c}{2R}\left[\frac{1}{1+\cos\theta_1}\left[1-\frac{R_c}{R_1}\right]+\frac{1}{1+\cos\theta_2}\left[1-\frac{R_c}{R_2}\right]\right] \ ,$$

$$\delta=\frac{1}{2R}[(1-\cos\theta_1)(R_1-R_c)+(1-\cos\theta_2)(R_2-R_c)] \ ,$$

$$v_c=\pi R_c^2R \ , \tag{14}$$

and $v_t=v_1+v_2$ the total conserved volume. For angles θ_1 and θ_2, we refer to the inset in Fig. 3. Apparently $\theta_1=\theta_2=0°$ and $\delta=0$ for two touching spheres. R_c ($\neq 0$) is the radius of a cylinder of length R, having a homogeneous flow in it, whose existence is assumed for the mass transfer between the two spherical fragments. The flow is assumed to be radial. The authors use a special ansatz for R_c:

$$R_c=a\ \min(R_1,R_2)f(R/R_t) \tag{15}$$

with $f(x)=1$ for $x\le 1$ and $a=0.4$–0.8 for best fit to the microscopic calculations.

B. The assault frequency ν

For the assault or escape frequency ν, we use the simple relation

$$\nu=\frac{v}{R_0}=(2E_2/\mu)^{1/2}/R_0 \ , \tag{16}$$

where R_0 is the radius of the parent nucleus and E_2 is the kinetic energy of the emitted cluster. Since we have assumed both the emitted cluster and the daughter nucleus to be produced in the ground state, the entire positive Q value is the total kinetic energy ($Q=E_1+E_2$) available for the decay process. This is shared between the two fragments, such that for the emitted cluster

$$E_2=\frac{A_1}{A}Q\ , \tag{16a}$$

and $E_1=Q-E_2$ is the recoil energy of the daughter nucleus. For the observed ^{14}C decay of Ra nuclei and ^{24}Ne decay of ^{232}U, the assault frequency is very large, $\sim 10^{21}$ $\sec^{-1}$.

C. The tunneling probability P

We use the WKB approximation and calculate the tunneling probability analytically. The nuclear interaction potential $V(R)$, from Eq. (8), for fixed η and η_Z, on normalizing to the sum of the binding energies, becomes

$$V(R)=\frac{Z_1Z_2e^2}{R}+V_P \quad \text{for } R\geq R_t\ . \tag{17}$$

This expression is similar to that used by Shi and Swiatecki[12] and is illustrated in Fig. 1 (solid lines) for $^{222}Ra\rightarrow{}^{14}C+{}^{208}Pb$. For $R<R_t$, we simply join smoothly the potential calculated at $R=R_t$ from (17) to the Q value at the parent nucleus radius, $R=R_0$. For the region $R_0<R<R_t$, we are not interested in the actual shape of the potential, since our calculations in Sec. III suggest to use $R=R_t$ for the evaluation of the preformation factor. In other words, the first (inner) turning point in the WKB penetrability integral is chosen at $R=R_t$. The outer (second) turning point, $R=R_b$, is taken to give the Q value of the reaction, i.e., $V(R_b)=Q$.

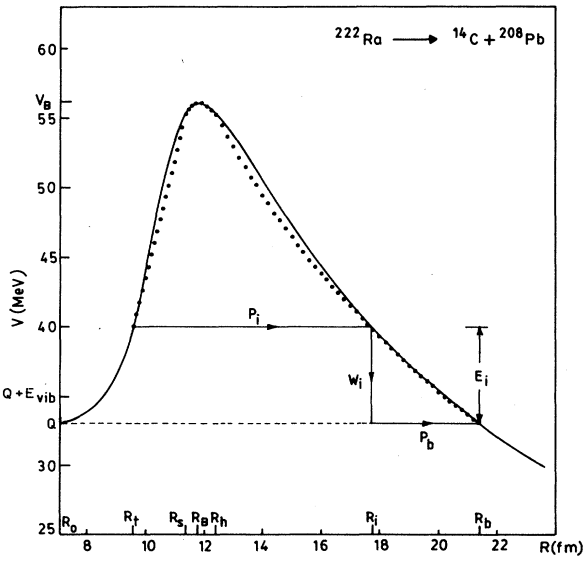

FIG. 1. The nuclear interaction potential for $^{222}Ra\rightarrow{}^{14}C+{}^{208}Pb$, calculated as a sum of Coulomb and proximity potentials (solid lines). The path of tunneling is also shown. The dotted lines show the parametrized potential [Eq. (24)], used to calculate the penetrability analytically.

We notice in Fig. 1 that for the inner and outer turning points, respectively, at $R=R_t$ and R_b, the transmission probability P consists of three contributions: the penetrability P_i from R_t to R_i, the deexcitation probability W_i at R_i and then the penetrability P_b from R_i to R_b. Thus,

$$P=P_iW_iP_b\ . \tag{18}$$

Notice that shifting the first turning point from $R=R_t$ to $R=R_0$ leads to model of Shi and Swiatecki[12] for penetrability calculations.

Following Greiner and Scheid[19] (see also Refs. 18 and 32), the choice of starting the tunneling process at an energy $V(R_t)$, i.e., above the Q value, is a decay into the excited states of the daughter nucleus (or the cluster or both). These authors suggest to scale the deexcitation probability W_i, exponentially with the excitation energy E_i:

$$W_i=\exp(-bE_i)\ . \tag{19}$$

This apparently means that the deexcitation process is restricted to only first-order transitions. If the parameter b is allowed to depend on R_i, it would then become a process of multiple deexcitation into excited states and proceed as a step like process. This means that second- and higher-order transitions have to be calculated, which make the model complicated, though calculable. The ansatz (19) is applied to α decay of $^{222,224}Ra$ and ^{232}U and it is shown[19] that for R_i values of interest, the parameters b must be very small. Therefore, for heavy cluster emission, the authors assumed $b=0$, which means

$$W_i=1\ . \tag{20}$$

Then, Eq. (18), defining the transmission probability reduces to

$$P=P_iP_b\ , \tag{21}$$

where in WKB theory, the penetrabilities P_i,P_b are defined as

$$P_i=\exp\left[-\frac{2}{\hbar}\int_{R_t}^{R_i}\{2\mu[V(R)-V(R_i)]\}^{1/2}dR\right]\ , \tag{22}$$

$$P_b=\exp\left[-\frac{2}{\hbar}\int_{R_i}^{R_b}\{2\mu[V(R)-Q]\}^{1/2}dR\right]\ , \tag{23}$$

We solve the integrals in Eqs. (22) and (23) analytically. For this purpose we parametrize the potential $V(R)$, calculated from Eq. (17) and illustrated in Fig. 1, as follows:

$$V(R)=\begin{cases} V(R_t)+s(R-R_t), & R_t\leq R\leq R_s\ , \\ V_B-\frac{1}{2}k(R-R_B)^2, & R_s\leq R\leq R_h\ , \\ V(R_h)-C_1\dfrac{R-R_h}{R}, & R_h\leq R\leq R_i\ , \\ V(R_i)-C_2\dfrac{R-R_i}{R}, & R_i\leq R\leq R_b\ , \end{cases} \tag{24}$$

Equation (24) shows that the first part of potential from R_t to R_s is a straight line of slope s, the top part between R_s and R_h being an inverted harmonic oscillator and the rest from R_h to R_i and R_i to R_b are a Coulomb potential of the type $1/R$. V_B and R_B in Eq. (24) give the height and position of the barrier. Then, using the fact that

$$\int_{R_t}^{R_i} f(R)dR = \int_{R_t}^{R_s} f(R)dR + \int_{R_s}^{R_h} f(R)dR + \int_{R_h}^{R_i} f(R)dR \ , \tag{25}$$

we get

$$P_i = \exp\left[-\frac{2}{\hbar}\sqrt{2\mu}\{\tfrac{2}{3}[V(R_s)-V(R_t)]^{1/2}(R_s-R_t) - \frac{1}{\sqrt{2k}}[V_B-V(R_i)][\vartheta_2-\tfrac{1}{2}\sin 2\vartheta_2-\vartheta_1+\tfrac{1}{2}\sin 2\vartheta_1]+\sqrt{C_1R_hR_i}[\vartheta_3-\tfrac{1}{2}\sin 2\vartheta_3]\}\right] \tag{26}$$

with

$$\vartheta_1=\cos^{-1}\frac{R_s-R_B}{\sqrt{\alpha_2}}, \quad \vartheta_2=\cos^{-1}\frac{R_h-R_B}{\sqrt{\alpha_2}}, \quad \vartheta_3=\tan^{-1}\left[\frac{R_i-R_h}{R_h}\right]^{1/2},$$

$$\alpha_2=\frac{2}{k}[V_B-V(R_i)], \quad k=\frac{2\{[V_B-V(R_s)]^{1/2}+[V_B-V(R_h)]^{1/2}\}^2}{(R_s-R_h)^2}, \quad C_1=\frac{R_i[V(R_h)-V(R_i)]}{R_i-R_h}$$

and

$$P_b=\exp\left[-\frac{2}{\hbar}\sqrt{2}\mu\sqrt{C_2R_iR_b}[\vartheta_4-\tfrac{1}{2}\sin 2\vartheta_4]\right] \tag{27}$$

with

$$\vartheta_4=\tan^{-1}\left[\frac{R_b-R_i}{R_i}\right]^{1/2}, \quad C_2=\frac{R_b[V(R_i)-V(R_b)]}{R_b-R_i} \ .$$

Substituting (26) and (27) in (21) we get the analytical expression for the tunneling probability P, which in turn substituted in (5), along with (7) and (16), gives the decay constant λ.

III. CALCULATIONS AND RESULTS

A. The relative clustering formation probability

In this section, we have first calculated the fragmentation potentials $V(\eta)$ at various relative positions R, starting from touching configuration ($R=R_t$) to a large overlap of two spheres. This is illustrated in Fig. 2 for ^{222}Ra. We notice that the positions and depths of all the potential energy minima are almost independent of the R value. As already discussed in Sec. II A, the situation would remain exactly the same if, instead of two spheres approximation, the TCSM were used for calculating the potential energy surfaces $V(\eta)$. It may, however, be mentioned that at present this statement is true only for $A_2 \gtrsim 12$, since the TCSM is not yet tested for very light clusters like the α particle. Following this result, the fragmentation potentials were also calculated earlier[35] at the touching configurations for a number of cases ($^{222-224}$Ra, ^{224,226}Th, and ^{232}U). In each case, deep potential energy minima are found to occur at the usual fission fragments (e.g., ^{88}Kr and its complementary fragment ^{134}Te in ^{222}Ra) and ^{4}He, ^{10}Be, ^{14}C, and $^{46-50}$Ca clusters. For ^{232}U, a deep minimum also appears at ^{24}Ne. In this work[35] it is also shown that inclusion of proximity potential does not bring any alteration in these minima, except that the minima at large mass asymmetry become slightly deeper. This evidently means that the potential energy minima, determining the possible clustering formation

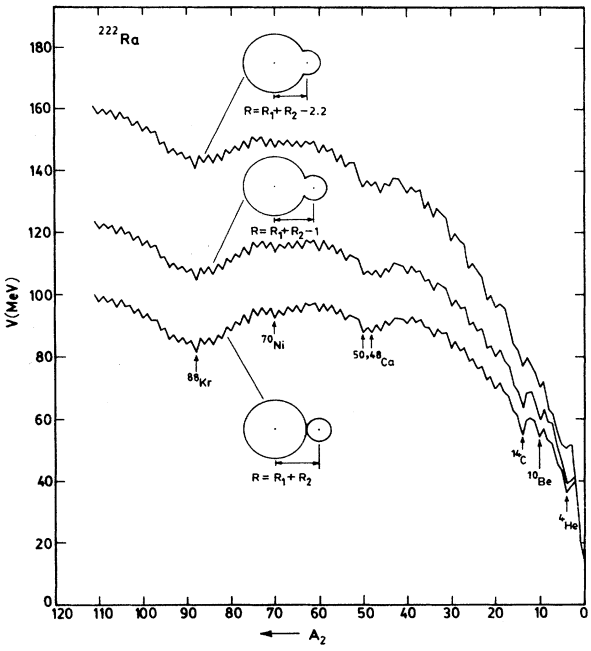

FIG. 2. The fragmentation potential $V(\eta)$ for ^{222}Ra, calculated at various relative positions R of the two fragments, using Eq. (8) with $Vp=0$. The nuclear shape configurations shown here refer approximately to ^{14}C+^{208}Pb. Calculations are also made for $R=R_1+R_2-0.5$ (not shown here), presenting exactly the same results.

TABLE I. Cluster preformation probabilities, P_0 (cluster), relative to one, and their ratios for ^{222}Ra, at various relative separations.

R (fm)	$P_0(^{14}C)$	$P_0(\alpha)$	$P_0(^{14}C)/P_0(\alpha)$
R_1+R_2	1.40×10^{-14}	9.93×10^{-8}	1.41×10^{-7}
$R_1+R_2-0.5$	4.12×10^{-15}	1.12×10^{-7}	0.37×10^{-7}
R_1+R_2-1	2.33×10^{-6}	9.33×10^{-1}	0.25×10^{-7}
$R_1+R_2-2.2$	0	3.93×10^{-10}	

(and decay) channels are only due to shell effects.[35]

Table I gives our calculated preformation probabilities P_0 for ^{14}C and α particle in ^{222}Ra at different R values (using the potentials of Fig. 2 with V_P added and classical mass parameters of Fig. 3). We notice that P_0 depends strongly on position R of the two fragments (cluster and daughter nucleus). At $R=R_1+R_2-2.2$, i.e., when the overlap of the cluster with the daughter nucleus is large, the formation yield for ^{14}C is zero and very small ($\sim10^{-10}$) for the α particle. However, it is interesting to observe that as the overlap of fragments decreases (the relative separation R increases), the relative cluster preformation yield with respect to α particle, i.e., the ratio $P_0(^{14}C)/P_0(\alpha)$, remains almost constant. In the following, we have interpreted this result in terms of the zero point vibration energy E_{vib}, introduced by Poenaru *et al.*[15] empirically to fit the cluster decay half-life times.

Using Eq. (3) for E_{vib}, we find that the configuration of larger overlap ($R=R_1+R_2-2.2$) refers to an excitation energy below E_{vib} but the ones with smaller overlaps ($R=R_1+R_2-1$ to R_1+R_2) above it. This suggests that the clustering formation begins at relative position corresponding to $V(R)\approx Q+E_{vib}$, and then the cluster formation probability relative to α particle remains almost the same up to the touching configuration. This result allows us to choose the touching configuration ($R=R_t$) as our starting point for the tunneling process.

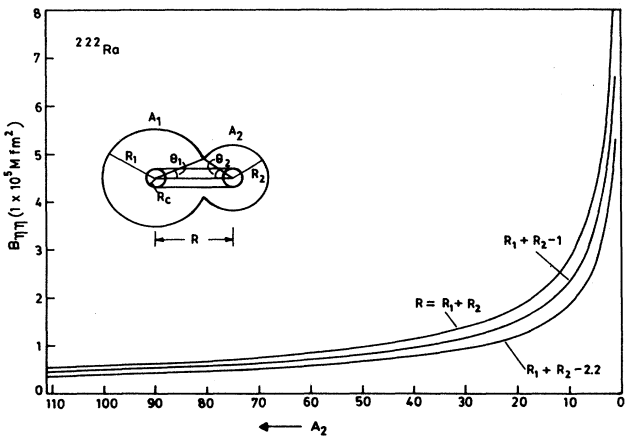

FIG. 3. The mass parameters $B_{\eta\eta}(\eta)$, calculated for different R values, using the classical model of Kröger and Scheid (Ref. 33) with $a=0.4$ in Eq. (15). Calculations are also made for $R=R_1+R_2-0.5$ but are not shown here.

In addition to simplifying the calculations of relative cluster preformation yields, this choice of R has the added advantage of not including the undetermined part of the scattering potential from R_0 to R_t in our analytical calculations of the tunneling probabilities.

Table II gives our calculated relative cluster preformation yields with respect to α particle (at $R=R_t=R_1+R_2$) for all the nuclei. ^{222}Ra is also included in this table for completeness. The results of the calculations of other authors are also given here for comparisons. We notice that our calculations agree within an order of magnitude with all the authors, except with Blendowske *et al.*[20] These authors obtain $P_0(^{14}C)/P_0(\alpha)\sim10^{-10}$ for radium nuclei. However, they argue that the formation probability should perhaps scale with the mass of cluster. Knowing the typical value of $P_0(\alpha)\sim10^{-2}$ for even nuclei in lead region, such an estimate yields $P_0(^{14}C)/P_0(\alpha)\sim10^{-5}$. Hence, Blendowske *et al.*[20] seem to underestimate the cluster preformation factor. Furthermore, in agreement with Iriondo *et al.*,[11] our calculations also show, at least qualitatively, a decrease in preformation probability with increase of cluster size.

We have also included in Table II, our calculations for ^{14}C decay of ^{232}U. We notice that the relative preformation probability for ^{14}C is about 100 times smaller than for ^{24}Ne. This explains why ^{24}Ne emission, rather than ^{14}C, is observed in the decay of ^{232}U. We shall see in the following (Table III) that the calculated branching ratio for the ^{14}C decay of ^{232}U, relative to α particle, is also so small that it is beyond the reach of present day experiments.

B. The tunneling probability

We first calculate the elastic scattering potential $V(R)$ by using Eq. (17) and fitting it to the parametrized potential of Eq. (24). This is illustrated in Fig. 1, showing a reasonable comparison between the calculated and fitted results. Here the position R_s is determined for a best fit to the straight line and inverted harmonic oscillator parts of the potential. Then, R_h is fixed by using an additional condition $V(R_s)=V(R_h)$. Knowing that by our definition, the positions R_i and R_b are given by the conditions $V(R_i)=V(R_t)$ and $V(R_b)=Q$, respectively, the tunneling probability P is calculated by using the analytical expression (26) and (27) in Eq. (21).

TABLE II. Relative clustering formation probabilities with respect to α-particle $P_0(\text{cluster})/P_0(\alpha)$.

Nucleus	Emitted cluster	Present work	Ref. 2	Ref. 18	Ref. 11	Ref. 20
^{222}Ra	^{14}C	1.41×10^{-7}		$\sim10^{-7}$	1.09×10^{-5}	3.05×10^{-10}
^{223}Ra	^{14}C	5.46×10^{-8}	7×10^{-5}–4×10^{-7}		9.61×10^{-6}	2.78×10^{-10}
^{224}Ra	^{14}C	1.69×10^{-8}			7.29×10^{-6}	1.94×10^{-10}
^{232}U	^{24}Ne	1.92×10^{-11}			4.0×10^{-10}	
	^{14}C	1.80×10^{-13}				

C. The branching ratios and the half-life times

Using Eq. (5), we have calculated the branching ratios $\lambda(\text{cluster})/\lambda(\alpha)$ in two different ways: (i) using $\lambda(\alpha)$ deduced from the measured α-decay half-life times[36] and (ii) using $\lambda(\alpha)$ calculated within our model. Results of both of these calculations are given in Table III, marked I and II, respectively. For comparisons we have also given in Table III, the experimental data as well as the calculations of Blendowske *et al.*[20] that contain the preformation factor and that of Poenaru *et al.*[16] and Shi and Swiatecki[12] that are without the preformation factor. We notice that our calculation I for ^{14}C decay of Ra nuclei compare with experiments better than the other calculations. For ^{24}Ne decay of ^{232}U, however, our model predictions are off by an order of magnitude. In this connection, it may be relevant to note that for this decay the measured spontaneous fission branching ratio[37] $\lambda_{SF}(^{24}\text{Ne})/\lambda(\alpha)=1.2\times10^{-12}$ is comparable to the recently measured[6] cluster decay branching ratio $\lambda(^{24}\text{Ne})/\lambda(\alpha)$ given in Table III. This points out that the predominant phenomenon in ^{24}Ne decay of ^{232}U may be fission rather than the cluster decay. Considering the fission of ^{232}U as a dynamical fragmentation process,[21] we have estimated the yields for ^{24}Ne fragment relative to α particle at the touching configuration using classical mass parameters. This comes out to be $\sim10^{-11}$, which compares rather well with other fission calculations[12,16] and lie within a factor of 10 of the experimental data. This point certainly needs a further study.

Our calculation II in Table III compares poorly with experiments, which means that the decay constants for α-particle emission in our model are, in general, underestimated. However, we do not expect our collective picture to give a proper description of the α decay. For that matter, for the heavy cluster decays, we have compared in Table IV our calculated half-lives with other calculations and the experimental data. Once again a better comparison with experimental data is obtained in case of the calculations based on two-step mechanism of clustering formation and barrier penetration (the present work and that of Ref. 20) for ^{14}C decay of Ra nuclei and that of fission calculations (Ref. 16) for ^{24}Ne decay of ^{232}U.

IV. SUMMARY

We have shown that the cluster radioactive decay can be very nicely considered as a two-step, decoupled mechanism of clustering formation and tunneling of the confining nuclear interaction barrier, at least for the

TABLE III. Experimental and calculated branching ratios (ratio of decay constants or decay widths) for heavy cluster emission relative to α-particle emission.

			Calculated branching ratios				
		Experiments	With preformation factor included			Without preformation factor	
			Present work				
Decay	Ref.	Branching ratio	Calc. I	Calc. II	Ref. 20	Ref. 16	Ref. 12[a]
$^{222}\text{Ra}\rightarrow{}^{14}\text{C}+{}^{208}\text{Pb}$	5	$(3.7\pm0.6)\times10^{-10}$	1.92×10^{-10}	5.94×10^{-7}	3.7×10^{-10}	1.0×10^{-11}	1.7×10^{-9}
	8	$(3.1\pm1.0)\times10^{-10}$					
$^{223}\text{Ra}\rightarrow{}^{14}\text{C}+{}^{209}\text{Pb}$	2	$(8.5\pm2.5)\times10^{-10}$					
	3	$(7.6\pm3.0)\times10^{-10}$					
	4	$(5.5\pm2.0)\times10^{-10}$	7.25×10^{-9}	5.95×10^{-7}	6.0×10^{-10}	3.2×10^{-9}	6.9×10^{-9}
	5	$(6.1\pm1.0)\times10^{-10}$					
	7	$(4.7\pm1.3)\times10^{-10}$					
$^{224}\text{Ra}\rightarrow{}^{14}\text{C}+{}^{210}\text{Pb}$	5	$(4.3\pm1.2)\times10^{-10}$	3.50×10^{-10}	1.10×10^{-8}	4.3×10^{-11}	1.6×10^{-12}	6.2×10^{-11}
$^{232}\text{U}\rightarrow{}^{24}\text{Ne}+{}^{208}\text{Pb}$	6	$(2.0\pm0.5)\times10^{-12}$	7.67×10^{-8}	2.04×10^{-8}		1.3×10^{-11}	4.9×10^{-11}
$^{232}\text{U}\rightarrow{}^{14}\text{C}+{}^{218}\text{Rn}$			1.50×10^{-21}	4.01×10^{-22}			

[a]Only the ratio of the penetration probabilities, $P(\text{cluster})/P(\alpha)$, are calculated.

TABLE IV. Calculated and experimental cluster-decay half-life times.

Decay	Experiments Ref.	Experiments $\log_{10}(T_{1/2})$	Calculated values of $\log_{10}(T_{1/2})$ Present work	Ref. 20	Ref. 16[a]	Ref. 32
$^{222}Ra \rightarrow {}^{14}C + {}^{208}Pb$	5,8	10.9–11.1	11.2	11.0	12.6	12.4
$^{223}Ra \rightarrow {}^{14}C + {}^{209}Pb$	2–5,7	14.9–15.5	14.1	15.2	14.8	14.5
$^{224}Ra \rightarrow {}^{14}C + {}^{210}Pb$	5	15.8–16.0	15.0	15.9	17.4	17.1
$^{232}U \rightarrow {}^{24}Ne + {}^{208}Pb$	6	21.3–21.5	16.5		20.4	

[a]Calculations without odd-even effects.

lighter clusters like ^{14}C. For the heavier clusters, perhaps the fission process is more predominant.

The clustering formation in our model is treated as the quantum-mechanical mass- and charge-fragmentation process. The static fragmentation potential $V(\eta,R)$ calculations clearly show that, independent of R value, the possible cluster formation (and decay) channels are fixed by shell effects in it. The dynamical clustering formation is shown to begin at the relative separation corresponding to $V(\eta,R)\approx Q+E_{\text{vib}}$ and its probability with respect to α-particle formation is found to remain nearly constant up to a separation distance corresponding to touching configuration. Our calculations support the estimates of Poenaru *et al.*[15] for the zero-point vibration energy E_{vib}. The role of dynamics is shown to be important by predicting the ^{14}C formation in ^{232}U to be at least 100 times less probable than ^{24}Ne formation, relative to the α particle.

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