Branching ratios of α-decay to excited states of even-even nuclei

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Y. Z. Wang[1], H. F. Zhang[1],[*] J. M. Dong[1], and G. Royer[2]

[1] *School of Nuclear Science and Technology, Lanzhou University Lanzhou 730000, People's Republic of China*
[2] *Laboratoire Subatech, UMR: IN2P3/CNRS-Université-Ecole des Mines, Nantes 44, France*
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Branching ratios of α-decay to members of the ground state rotational band and excited 0^+ states of even-even nuclei are calculated in the framework of the generalized liquid drop model (GLDM) by taking into account the angular momentum of the α-particle and the excitation probability of the daughter nucleus. The calculation covers isotopic chains from Hg to Fm in the mass regions $180 < A < 202$ and A$\geq$ 224. The calculated branching ratios of the α-transitions are in good agreement with the experimental data and some useful predictions are provided for future experiments.

I. INTRODUCTION

The α decay process was first explained by Gamow [1] and by Condon and Gurney [2] in 1928 as a quantum tunneling effect and it is one of the first examples proving the need to use the quantum mechanics to describe nuclear phenomena and its correctness. On the basis of the Gamow's theory, the experimental α-decay half-lives of nuclei can be well explained by both phenomenological and microscopic models [3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13]. Such α-decay calculations are mainly concentrated on the favored cases, e.g. the ground state α-transitions of even-even nuclei ($\Delta l = 0$) [14, 15]. Besides the favored α-transitions, the ground state of the parent nucleus can also decay to the excited states of the daughter nucleus ($\Delta l \neq 0$) [15]. Recently, there is increasing interest in two kinds of α transitions of even-even nuclei from both experimental and theoretical sides, i.e. the α-decay to excited 0^+ states and to members of ground state rotational band [16, 17, 18, 19, 20, 21, 22, 23]. These α transitions belong to the unfavored case, which are strongly hindered as compared with the ground state ones. Theoretically, the hindered α-transition is an effective tool to study the properties of α-emitters because it is closely related to the internal structure of nuclei [16, 17, 18, 19, 20, 21, 22, 23]. However, it is difficult to describe quantitatively the unfavored α-transitions due to the influence of both non-zero angular momentum and excitation of nucleons, especially for α-emitters in the neighbourhood of the shell closures. Although the favored α-decay model can be straightforwardly applied to the unfavored α-transition, the calculated branching ratios usually deviate significantly from the experimental data. Thus it is necessary to improve the favored α-decay model to describe the unfavored hindered α-decay. Experimentally it is also very helpful to make theoretical predictions on unfavored hindered α-transitions for future studies.

The generalized liquid drop model (GLDM) has been successfully used to calculate the half-lives of the favored α-decays from the nuclear ground states of even-even nuclei [10, 11, 12, 13]. As far as we know, the unfavored hindered α-transitions have not been investigated within the GLDM. In this paper, the GLDM has been improved by taking into account the influence of the angular momentum of the α-particle and the excitation probability of the daughter nucleus, investigating the hindered α-transitions of even-even nuclei with mass numbers $180 < A < 202$ and $A \geq 224$. The calculated branching ratios of α-decays are compared with the experimental data and the good agreement allows to provide predictions for future experiments. This paper is organized as follows. In section 2 the theoretical framework is introduced. The numerical results and corresponding discussions are given in Section 3. In the last section, some conclusions are drawn.

II. METHODS

In the favored α-decays, such as the ground state to ground state α-transitions of even-even nuclei, the angular momentum l carried by the α-particle is 0. Thus in the former framework of the GLDM, the centrifugal potential energy $V_{\text{cen}}(r)$ is not included [10, 11, 12, 13]. In the case of the ground state of a parent nucleus to the ground state of its deformed daughter nucleus and to the excited state I^+, the angular momentum l carried by the α-particle is not 0. Thus the centrifugal potential energy $V_{\text{cen}}(r)$ which can no more be neglected has been introduced into the GLDM as

$$V_{\text{cen}}(r) = \frac{\hbar^2}{2\mu}\frac{l(l+1)}{r^2} \tag{1}$$

where r and μ are the distance between the two fragments and the reduced mass of the α-daughter system, respectively.

The macroscopic GLDM energy becomes

$$E = E_V + E_S + E_C + E_{\text{Prox}} + V_{\text{cen}}(r). \tag{2}$$

When the nuclei are separated

$$E_V = -15.494\left[(1 - 1.8I_1^2)A_1 + (1 - 1.8I_2^2)A_2\right] \text{ MeV,} \tag{3}$$

$$E_S = 17.9439 \left[(1 - 2.6I_1^2)A_1^{2/3} + (1 - 2.6I_2^2)A_2^{2/3} \right] \text{ MeV}, \tag{4}$$

$$E_C = 0.6e^2 Z_1^2/R_1 + 0.6e^2 Z_2^2/R_2 + e^2 Z_1 Z_2/r, \tag{5}$$

where A_i, Z_i, R_i and I_i are the mass numbers, charge numbers, radii and relative neutron excesses of the two nuclei. r is the distance between the mass centers. The radii R_i are given by

$$R_i = (1.28A_i^{1/3} - 0.76 + 0.8A_i^{-1/3}) \text{ fm.} \tag{6}$$

For one-body shapes, the volume, surface and Coulomb energies are defined as

$$E_V = -15.494(1 - 1.8I^2)A \text{ MeV}, \tag{7}$$

$$E_S = 17.9439(1 - 2.6I^2)A^{2/3}(S/4\pi R_0^2) \text{ MeV}, \tag{8}$$

$$E_C = 0.6e^2(Z^2/R_0) \times 0.5 \int (V(\theta)/V_0)(R(\theta)/R_0)^3 \sin\theta d\theta. \tag{9}$$

S is the surface of the one-body deformed nucleus. $V(\theta)$ is the electrostatic potential at the surface and V_0 the surface potential of the sphere.

The surface energy results from the effects of the surface tension forces in a half space. When there are nucleons in regard in a neck or a gap between separated fragments an additional term called proximity energy must be added to take into account the effects of the nuclear forces between the close surfaces. This term is essential to describe smoothly the one-body to two-body transition and to obtain reasonable fusion barrier heights. It moves the barrier top to an external position and strongly decreases the pure Coulomb barrier.

$$E_{\text{Prox}}(r) = 2\gamma \int_{h_{\min}}^{h_{\max}} \Phi \left[D(r,h)/b \right] 2\pi h \, dh, \tag{10}$$

where h is the distance varying from the neck radius or zero to the height of the neck border. D is the distance between the surfaces in regard and $b = 0.99$ fm the surface width. Φ is the proximity function of Feldmeier. The surface parameter γ is the geometric mean between the surface parameters of the two nuclei or fragments. The combination of the GLDM and of a quasi-molecular shape sequence has allowed to reproduce the fusion barrier heights and radii, the fission and the α and cluster radioactivity data.

In the unfavored α-transitions, the parent nucleus decays to the excited states of the daughter nucleus. Thus the excitation energy E_I^* has influence on the penetration probability of α-particle through the Coulomb barrier. The α transitions are assumed to occur from the ground state 0^+ of an even-even parent nucleus to the rotational band $(0^+, 2^+, ..., I^+, ...)$ of the ground state of a daughter nucleus. The α decay of $0^+ \longrightarrow I^+$ transition requires

that the α-particle carries an angular momentum of $l = I$ to satisfy angular momentum conservation and parity conservation. The α-decay energy from the ground state of a parent nucleus is related to the excitation energy of the I^+ states in the daughter nucleus. It is the subtraction between the decay energy of the ground state and the excitation energy of the I^+ state

$$Q_{0^+ \longrightarrow I^+} = Q_{0^+ \longrightarrow 0^+} - E_I^* \tag{11}$$

Partial half-life of the ground state of a parent nucleus to each state of a rotational band of its daughter nucleus can be obtained with different orbital angular momentum l and α decay energy $Q_{0^+ \longrightarrow I^+}$.

The half-life of a parent nucleus decaying via α-emission is calculated using the WKB barrier penetration probability. The barrier penetrability $P(Q_\alpha, E_I^*, l)$ is calculated within the action integral

$$P(Q_\alpha, E_I^*, l) = \exp \left[-\frac{2}{\hbar} \int_{R_{\text{in}}}^{R_{\text{out}}} \sqrt{2B(r)(E(r) - E_{sph})} dr \right] \tag{12}$$

The deformation energy (relative to the sphere) is small until the rupture point between the fragments and the two following approximations may be used: $R_{\text{in}} = R_{\text{d}} + R_\alpha$ and $B(r) = \mu$, where μ is the reduced mass. R_{out} is simply $e^2 Z_{\text{d}} Z_\alpha / Q_\alpha$.

The residual daughter nucleus after disintegration has the most probability to stay in its ground state, and the probability to stay in its excited state is relatively much smaller. Therefore it is a reasonable assumption that the probability of the residual daughter nucleus to stay in its excited states $(I^+ = 2^+, 4^+, 6^+, ...)$ obeys the Boltzmann distribution [24]

$$\omega_I(E_I^*) = \exp\left[-cE_I^*\right] \tag{13}$$

where E_I^* is the excitation energy of state I^+ and c is a free parameter. This excitation probability function has been added to the model with a value of the parameter c fixed to 3.0. This means that only a single parameter is introduced in the whole calculation. It is stressed that the inclusion of the excitation probability is reasonable in physics and it can lead to good agreement between experiment and theory. Here I_{I^+} is defined as the product of the penetration factor and the excitation probability

$$I_{I^+} = \omega_I(E_I^*)P(Q_\alpha, E_I^*, l). \tag{14}$$

It is the probability of α-transition from the ground state of the parent nucleus to the excited states I^+ of the daughter nucleus. It is very convenient to estimate the influences of these factors on the hindered α-transitions from I_{I^+}. With the help of I_{I^+}, the branching ratios of α-decay to each state of the rotational band of the daughter nucleus can be written as

$$b_{\text{g.s.}}^{0^+}\% = I_{0^+}/(I_{0^+} + I_{2^+} + I_{4^+} + I_{6^+} + ...) \times 100\%$$

$$b_{\text{g.s.}}^{2^+}\% = I_{2^+}/(I_{0^+} + I_{2^+} + I_{4^+} + I_{6^+} + ...) \times 100\%$$

$$b_{\text{g.s.}}^{4^+}\% = I_{4^+}/(I_{0^+} + I_{2^+} + I_{4^+} + I_{6^+} + ...) \times 100\%$$
$$... \quad (15)$$

Similarly, the branching ratio of the α-decay to the excited 0^+ state of the daughter nucleus is given by

$$b_{\text{e.s.}}^{0^+} = b_{\text{g.s.}}^{0^+} \times \frac{\omega_0(E_0^*)}{\omega_0(0)} \frac{P_\alpha(Q_\alpha, E_0^*, 0)}{P_\alpha(Q_\alpha, 0, 0)} \quad (16)$$

where $b_{\text{g.s.}}^{0^+}\%$ is the branching ratio of α-transition between the ground states. The α-transition to the excited 0^+ state of the daughter nucleus does not involve the variation of the angular momentum l, which is an ideal case for theoretical studies of hindered α-transitions.

FIG. 2: The α-decay branching ratios to the rotational band of the ground state of ^{238}Pu. The α-decay energy Q_α and excitation energy E_I^* are measured in MeV.

FIG. 1: The α-decay branching ratios to the rotational band of the ground state of ^{234}U. The α-decay energy Q_α and excitation energy E_I^* are measured in MeV.

III. RESULTS AND DISCUSSIONS

The systematic calculation on α-decay branching ratios to the rotational band is rare because some data of the excited states have been obtained recently [15]. Experimentally it is known that the ground state of the even-even actinides mainly decays to the 0^+ and 2^+ states of their daughter nucleus [15]. The sum of branching ratios to the 0^+ and 2^+ states is as large as 99% in many cases. The α-transitions to other members of the rotational band ($I^+ = 4^+, 6^+, 8^+,...$) are strongly hindered. This is different from the α-transition to the ground or excited 0^+ state where the angular momentum carried by the α-particle is zero ($\Delta l = 0$). Here the influence of the non-zero angular momentum should be included for $\Delta l \neq 0$ transitions. In Figs. 1-3, three typical

figures for the α-decay fine structure for ^{238}Pu, ^{242}Cm and ^{246}Cf are shown. The α-decay branching ratios of ^{238}Pu and ^{242}Cm have been measured up to 8^+ state of the rotational band, and the branching ratio of ^{246}Cf has been up to 6^+ state experimentally. It is shown in these figures that the calculated values agree with the experimental ones for both the low-lying states ($0^+, 2^+$) and the high-lying ones ($6^+, 8^+$), however, the calculated branching ratio to the 4^+ state is slightly larger than the experimental one. The branching ratios to the ground state rotational band for other even-mass α-emitters, such as $^{224-230}$Th, $^{228-238}$U, $^{236-244}$Th and so on have also been calculated. The discrepancy in describing the 4^+ state also exists for these nuclei in the calculations. There should be unknown physics reason behind it. The abnormity of the 4^+ state does not allow to explain the small discrepancy. It is very interesting to pursue this by performing more microscopic and reasonable calculations in the future. Nevertheless, the overall agreement of branching ratios to the rotational band of these nuclei is acceptable.

Besides the calculations on the α-decay branching ratios to the rotational band, a systematic calculation on the unfavored α-decays to the excited 0^+ states of even-even α-emitters in the actinide region has been done. The table 1 gives the experimental and calculated branching ratios of the α-transition to the excited 0^+ states for even-mass isotopes of Th, U, Pu and Cm (in table 1, the symbol ^{a} represents the cases where the experimental branching ratio is unknown). The experimental ground state branching ratio ($b_{\text{g.s.}}^{0^+}\%$) is used and its variation range mainly from 67.4% to 77.9% for different nuclei in this region [15]. However, the variation of the experimental branching ratio to the excited 0^+ state is relatively much larger and its amplitude is as

TABLE I: Experimental and calculated branching ratios of α-decay to the excited 0^+ states of the daughter nucleus. Q_α is the ground state α-decay energy and $b_{\text{g.s.}}^{0^+}\%$ is the branching ratio of the α decay to the ground state of the daughter nucleus. $E_{0^+}^*$ is the excitation energy of the excited 0^+ state. $b_{\text{e.s.}}^{0^+}\%$ is the corresponding experimental or theoretical α decay branching ratio.

Nuclei	Q_α (MeV)	$b_{\text{g.s.}}^{0^+}\%$ (Expt.)	$E_{0^+}^*$ (MeV)	$b_{\text{e.s.}}^{0^+}\%$ (Expt.)	$b_{\text{e.s.}}^{0^+}\%$ (Calc.)
^{226}Th	6.452	75.5%	0.914	$3.4\times10^{-4}\%$	$1.46\times10^{-4}\%$
^{228}Th	5.520	71.1%	0.916	$1.8\times10^{-5}\%$	$5.89\times10^{-6}\%$
^{230}Th	4.770	76.3%	0.825	$3.4\times10^{-6}\%$	$1.33\times10^{-6}\%$
^{232}Th	4.083	77.9%	0.721	a	$3.29\times10^{-7}\%$
^{230}U	5.993	67.4%	0.805	$3.0\times10^{-4}\%$	$1.75\times10^{-4}\%$
^{232}U	5.414	68.0%	0.832	$2.2\times10^{-5}\%$	$1.47\times10^{-5}\%$
^{234}U	4.859	71.4%	0.635	$2.6\times10^{-5}\%$	$1.25\times10^{-4}\%$
^{236}U	4.572	73.8%	0.730	a	$3.45\times10^{-6}\%$
^{236}Pu	5.867	69.3%	0.691	$6.0\times10^{-3}\%$	$7.66\times10^{-4}\%$
^{238}Pu[1]	5.593	70.9%	0.810	$5.0\times10^{-5}\%$	$3.65\times10^{-5}\%$
^{238}Pu[2]	5.593	70.9%	1.045	$1.2\times10^{-6}\%$	$2.89\times10^{-7}\%$
^{240}Pu	5.256	72.8%	0.919	$6.3\times10^{-7}\%$	$8.66\times10^{-7}\%$
^{242}Pu	4.983	77.5%	0.926	a	$1.78\times10^{-7}\%$
^{242}Cm[1]	6.216	74.0%	0.942	$5.2\times10^{-5}\%$	$1.36\times10^{-5}\%$
^{242}Cm[2]	6.216	74.0%	1.229	$5.1\times10^{-7}\%$	$1.02\times10^{-7}\%$
^{244}Cm[1]	5.902	76.4%	0.861	$1.55\times10^{-4}\%$	$3.20\times10^{-4}\%$
^{244}Cm[2]	5.902	76.4%	1.089	a	$3.78\times10^{-7}\%$

TABLE II: The same as Table 1, but for even isotopes of Rn, Po, Pb and Hg. The experimental data of ^{180}Hg-^{202}Rn are taken from Ref.[17]. The experimental data of ^{190}Po are taken from Ref. [18].

Nuclei	Q_α (MeV)	$b_{\text{g.s.}}^{0^+}\%$ (Expt.)	$E_{0^+}^*$ (MeV)	$b_{\text{e.s.}}^{0^+}\%$ (Expt.)	$b_{\text{e.s.}}^{0^+}\%$ (Calc.)
^{180}Hg	6.257	33%	0.443	$2.6\times10^{-2}\%$	$1.1\times10^{-1}\%$
^{182}Hg	5.997	8.6%	0.422	$2.9\times10^{-2}\%$	$2.8\times10^{-2}\%$
^{184}Hg	5.658	1.25%	0.478	$2.0\times10^{-3}\%$	$1.1\times10^{-3}\%$
^{186}Pb	6.474	<100%	0.328	<0.20%	1.67% b
^{188}Pb	6.110	(3-10)%	0.375	$(2.9-9.5)\times10^{-2}\%$	$(1.9-6.4)\times10^{-2}\%$
^{190}Po[1]	7.695	96.4%	0.523	3.3%	0.35%
^{190}Po[2]	7.695	96.4%	0.650	0.3%	0.093%
^{194}Po	6.986	93%	0.658	0.22%	0.036%
^{196}Po	6.657	94%	0.769	$2.1\times10^{-2}\%$	$4.8\times10^{-3}\%$
^{198}Po	6.307	57%	0.931	$7.6\times10^{-4}\%$	$1.2\times10^{-4}\%$
^{202}Rn	6.775	(80-100)%	0.816	$(1.4-1.8)\times10^{-3}\%$	$(2.2-2.8)\times10^{-3}\%$

high as $0.006\%/(5.1\times10^{-7}\%) \approx 10^4$ times (see Table 1). Therefore it is a challenging task to obtain a quantitative agreement between experimental and theory. The last two columns of Table 1 allow to observe that the calculated results from the GLDM are in correct agreement with the experimental data. Besides the first excited 0^+ state, the α-transitions to the second excited 0^+ state have also been observed in experiment for some nuclei, such as ^{238}Pu and ^{242}Cm. Our calculated branching ratios also agree with the experimental ones in these cases. In Table 1, the experimental branching ratios to the first excited 0^+ state have not been measured yet for nuclei ^{232}Th, ^{236}U and ^{242}Pu [15]. The corresponding predicted values for these nuclei are listed in Table 1. Meanwhile, the branching ratio to the second excited 0^+ state in decay of ^{244}Cm is also given in Table 1. It will be very interesting to compare these theoretical predictions with future experimental observations.

The experimental and the calculated branching ratios to the excited 0^+ states for even-mass isotopes of Hg, Pb, Po and Rn are listed in Table 2. The hindered transitions ($\Delta l = 0$) of these nuclei involve complex particle-hole excitations above or below the closed shell Z=82 [25]. Although the situation becomes more complicated, it is seen from Table 2 that the experimental results are reasonably reproduced in the framework of the GLDM by taking into account the excited energies of the daughter nucleus. The α-decay energies, the ground state branching ratios and the excitation energies of nuclei in Table 2 are taken from the experimental values [16, 17]. It is found that the calculated branching ratios of ^{202}Rn, $^{190,194-198}$Po, ^{188}Pb and $^{180-184}$Hg are consistent with the experimental data. For the ^{186}Pb, the calculated value slightly deviates from the experimental one (in table 2, the symbol b denotes the cases where the calculated branching ratio deviates from the experimental data). The agreement may be fur-

FIG. 3: The α-decay branching ratios to the rotational band of the ground state of ^{242}Cm. The α-decay energy Q_α and excitation energy E^*_I are measured in MeV.

ther improved by taking into account more factors, such as the nuclear deformations [25] and so forth.

IV. CONCLUSIONS

In summary, the α-decay branching ratios of even-even nuclei with mass numbers 180< A <202 and A$\geq$ 224 have been calculated in the framework of the GLDM by taking into account the angular momentum of the α-particle and the excitation probability of the daughter nucleus. The calculated branching ratios to the rotational band of the ground state of even-even actinides are consistent with the experimental data except for the branching ratios to the 4^+ state. The calculated branching ratios to the first and second excited 0^+ states of the daughter nucleus are also in agreement with the available experimental data. Some predicted branching ratios are calculated for the cases where the experimental values are still unknown. These theoretical predictions are very helpful to the un-observed hindered α-transitions for future experiments.

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