Calculation of two-proton radioactivity and application to ^{9}Be, ^{6,7}Li, ^{3,6}He, and ^{2,3}H emissions

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The $2p$ radioactivity half-lives have been calculated within a tunneling process through the potential barrier determined from the original version of the generalized liquid drop model and quasimolecular shapes. The sparse experimental data on the $2p$ radioactivity half-lives of the ^{12}O, ^{16}Ne, ^{19}Mg, ^{45}Fe, ^{48}Ni, ^{54}Zn, and ^{67}Kr nuclei are roughly reproduced when taking into account the Q_{2p}^{exp} value. An analytic formula is provided to obtain rapidly these different half-lives. Using the Q_{2p} values extracted from the recent Nubase2020 table, extrapolations to other perhaps possible $2p$ emitters are proposed. Within the same approach, formulas to determine the ^{9}Be, ^{6,7}Li, ^{3,6}He, and ^{2,3}H decay half-lives at low excitation energy are also provided.

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I. INTRODUCTION

The simultaneous emission of two protons by nuclei near or beyond the proton drip line was first predicted by Zel'dovich [1] and Goldansky [2,3] in the 1960s. The idea was that the two-proton radioactivity may occur due to the proton pairing in a nucleus, owing to which it is energetically easier to eject from the nucleus a pair of protons at once than to break them apart. This should be observed mainly for neutron-deficient isotopes of even-Z light elements while for odd-Z nuclei the one proton radioactivity remains the main decay mode.

This $2p$ radioactivity ($Q_{2p} > 0$ and $Q_p < 0$) was observed at GANIL [4] and GSI [5] in 2002 in ^{45}Fe nucleus. The $2p$ radioactivity observation of the ^{54}Zn [6], ^{48}Ni [7], and ^{67}Kr [8] nuclei has followed. The decay mechanism of the ^{67}Kr nucleus is actually discussed [9–11]. The two-proton emission of the very short-lived nuclei ^{12}O [12], ^{16}Ne [13], ^{19}Mg [14] has been also reported. The experimentally measured Q_{2p} and $T_{1/2}$ are given in [15,16]. The whole landscape of two-proton radioactivity has been studied and some other possible $2p$ emitters have been proposed [17].

Theoretically, several models have been used to study the $2p$ radioactivity: the direct decay model [17], the simultaneous versus sequential decay model [18], the diproton model [19], the three-body potential [20], the effective liquid drop model [15], a version of the generalized liquid drop model including a spectroscopic factor and an assault frequency depending on the kinetic energy and the mass of the emitted $2p$ pair [16], a Gamow-like model [21], and the Coulomb and proximity potential model for deformed nuclei [22]. A four parameter empirical formula [23] and the two parameter new Geiger-Nutall law [24] have been also proposed to reproduce the experimental data.

In spite of its complexity, it has been shown very recently [25] that the two-proton emission process obeys similar rules as for binary emission processes like proton, α, and heavy cluster decays. The purpose of this paper is to verify this assumption in determining the half-lives of the $2p$ radioactivity with the first version of the generalized liquid drop model (GLDM) [26] without changing anything and, also, in using a simple three parameter formula looking like the α decay half-life formula proposed in [27]. The experimental Q values have been used when they are known. The half-lives being highly sensitive to the Q values, the new values extracted from the new Nubase2020 tables [28], which are different from the ones given in Ref. [29], have been used for predictions of other possibly $2p$ emitters. Finally, formulas of the same type is also proposed to calculate the partial half-lives of other light nucleus emissions.

II. GENERALIZED LIQUID DROP MODEL AND DECAY THROUGH THE POTENTIAL BARRIER

The generalized liquid drop model (GLDM) has been developed first to study heavy-ion reactions and the fusion and fission processes [26,30,31] within two- or one-body compact and creviced quasimolecular shapes [32] and, later, to α and clusters emissions [27,33]. The GLDM energy is given by [26]

$$E = E_V + E_S + E_C + E_{\text{prox}}, \tag{1}$$

where the different terms are, respectively, the volume, surface, Coulomb, and nuclear proximity energies.

For one-body shapes, the first three contributions are expressed as

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

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

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I is the relative neutron excess and S is the surface of the deformed nucleus,

$$E_C = 0.6e^2(Z^2/R_0)B_C. \quad (4)$$

The Coulomb shape dependent function B_C is defined as

$$B_C = \frac{15}{16\pi^2 R_0^5}\int d\tau \int \frac{d\tau'}{|r-r'|}. \quad (5)$$

It has been determined using the axial symmetry of the system and complete elliptic integrals

$$B_C = 0.5\int (V(\theta)/V_0)(R(\theta)/R_0)^3 \sin\theta d\theta, \quad (6)$$

where $V(\theta)$ is the electrostatic potential at the surface and V_0 the surface potential of the sphere. The radius R_0 of the spherical nucleus is given by

$$R_0 = (1.28A^{1/3} - 0.76 + 0.8A^{-1/3})\ \text{fm}. \quad (7)$$

The elliptic lemniscatoid family has been defined and used to describe the one-body shapes [32].

For two-body shapes the volume, surface, and Coulomb energies of the two nuclei have been added as well as the interaction Coulomb energy. There is no frozen density approximation or parabolic approximation.

All along the deformation path the proximity energy term E_{prox} takes into account the nuclear attractive forces between nucleons in the neck, in the case of a deformed one-body shape, or across the gap, in the case of two separated fragments. In the quasimolecular shape valley where the necks are narrow and well developed this correction to the surface energy plays a main role on a large part of the deformation path and specially around the touching point. When the proximity energy is taken into account, the potential barrier is smooth and the maximum corresponds to two separated nuclei maintained in unstable equilibrium by the balance between the repulsive Coulomb forces and the attractive nuclear proximity forces. The proximity energy is defined as

$$E_{\text{prox}}(r) = 2\gamma\int \Phi[D(r,h)/b]2\pi h dh, \quad (8)$$

where r is the distance between the mass centers. Φ is the proximity function. h is the transverse distance varying from zero, for separated fragments or the neck radius for one-body shapes, to the height of the neck border. b the surface width fixed at the standard value of 0.99 fm. D is the distance between the opposite surfaces on a line parallel to the fission axis. Finally the surface parameter γ is given by a geometric mean between the surface parameters of the two fragments:

$$\gamma = 0.9517\big(1-k_sI_1^2\big)^{1/2}\big(1-k_sI_2^2\big)^{1/2}\ \text{MeV fm}^{-2}. \quad (9)$$

This model has helped to explain and reproduce some of the fusion [26,34], fission [30,35], cluster [33,36], and α radioactivity [27] data. Its advantage is its relative simplicity (as the mass formula), its weakness here is its adjustment to reproduce the Q value.

The α [27] and cluster [33] emissions have been viewed as an adiabatic tunneling process through a potential barrier like a very asymmetric fission process. The same approach has been applied here to the $2p$ radioactivity. Then, the decay constant is simply given by

$$\lambda = \nu_0 P, \quad (10)$$

the assault frequency ν_0 being taken as

$$\nu_0 = 10^{20}\ \text{s}^{-1}. \quad (11)$$

The barrier penetrability P is determined within the action integral

$$P = \exp\left[-\frac{2}{\hbar}\int \sqrt{2B(r)(E(r)-E(\text{sphere}))}dr\right]. \quad (12)$$

$(E(r) - E(\text{sphere}))$ is the difference between the energy of the decaying deformed nucleus at distance r and the energy of the initial spherical nucleus within the GLDM. The inertia $B(r)$ defined in [33] has been retained but the reduced mass plays the essential role since the main part of the potential barrier corresponds to two separated nuclei.

The partial half-life is related to the decay constant λ by

$$T_{1/2} = \frac{\ln 2}{\lambda}. \quad (13)$$

TABLE I. Comparison between the experimental decimal logarithm of the half-lives (in s) for $2p$ emission from ${}^{12}_{8}$O, ${}^{16}_{10}$Ne, ${}^{19}_{12}$Mg, ${}^{45}_{26}$Fe, ${}^{48}_{28}$Ni, ${}^{54}_{30}$Zn, ${}^{67}_{36}$Kr, and the theoretical values calculated from the GLDM and formula (16).

Nucleus	Q_{2p}^{exp} (MeV)	$\log_{10}(T_{1/2})$ Exp	Ref.	$\log_{10}(T_{1/2}^{th})$ GLDM	$\log_{10}(T_{1/2}^{th})$ Formula (16)
${}^{12}_{8}$O	1.820(120)	$-20.94^{+0.43}_{-0.21}$	[13]	−19.62	−21.75
${}^{12}_{8}$O	1.790(40)	$-21.10^{+0.18}_{-0.13}$	[12]	−19.58	−21.67
${}^{12}_{8}$O	1.800(400)	$-21.12^{+0.78}_{-0.26}$	[38]	−19.60	−21.70
${}^{16}_{10}$Ne	1.330(80)	$-20.64^{+0.30}_{-0.18}$	[13]	−17.69	−18.77
${}^{16}_{10}$Ne	1.400(20)	$-20.38^{+0.03}_{-0.03}$	[39]	−17.88	−19.10
${}^{19}_{12}$Mg	0.750(50)	$-11.40^{+0.14}_{-0.20}$	[14]	−13.1	−11.98
${}^{45}_{26}$Fe	1.100(100)	$-2.40^{+0.26}_{-0.26}$	[5]	−3.16	−1.66
${}^{45}_{26}$Fe	1.140(50)	$-2.07^{+0.24}_{-0.21}$	[4]	−3.65	−2.32
${}^{45}_{26}$Fe	1.154(16)	$-2.55^{+0.13}_{-0.12}$	[7]	−3.80	−2.54
${}^{45}_{26}$Fe	1.210(50)	$-2.42^{+0.03}_{-0.03}$	[40]	−4.43	−3.40
${}^{48}_{28}$Ni	1.350(20)	$-2.08^{+0.40}_{-0.78}$	[41]	−4.13	−3.43
${}^{48}_{28}$Ni	1.290(40)	$-2.52^{+0.24}_{-0.22}$	[7]	−3.52	−2.60
${}^{48}_{28}$Ni	1.310(40)	$-2.52^{+0.24}_{-0.22}$	[42]	−3.72	−2.88
${}^{54}_{30}$Zn	1.480(20)	$-2.43^{+0.20}_{-0.14}$	[43]	−3.94	−3.45
${}^{54}_{30}$Zn	1.280(210)	$-2.76^{+0.15}_{-0.14}$	[6]	−1.86	−0.66
${}^{67}_{36}$Kr	1.690(17)	$-1.70^{+0.02}_{-0.02}$	[8]	−1.47	−1.12

III. EXPERIMENTAL $2p$ RADIOACTIVITY Q_{2p} VALUES AND HALF-LIVES

The experimental $2p$ radioactivity Q_{2p} values and half-lives given in [15,16,22,24] are recalled in Table I. The uncertainty on the experimental Q_{2p} and $T_{1/2}$ is relatively large. The values of the $\log_{10}(T_{1/2})$ obtained from the original version of the GLDM are indicated in the fifth column of this table. No

TABLE II. Comparison between the logarithm of the half-lives (in s) for $2p$ emission from various nuclei calculated from the GLDM and formula (16) and Q^{th}_{2p} and Q^{th}_{1p} values extracted from Nubase2020.

Nucleus	Q^{th}_{2p} (MeV) Nubase2020	$\log_{10}(T_{1/2})$ GLDM	$\log_{10}(T_{1/2})$ Formula (16)	Q^{th}_{1p} (MeV) Nubase2020
		True $2p$ radioactivity		
$^{22}_{14}Si$	1.584	−15.93	−17.0	−0.739
$^{39}_{22}Ti$	1.058	−6.43	−5.26	−0.539
$^{42}_{24}Cr$	1.472	−8.44	−8.75	−0.539
$^{49}_{28}Ni$	1.082	−1.08	0.752	−0.489
$^{59}_{32}Ge$	1.602	−3.59	−3.30	−0.119
		Not true $2p$ radioactivity		
$^{26}_{16}S$	2.357	−16.98	−19.02	0.201
$^{30}_{18}Ar$	3.422	−18.04	−20.97	0.761
$^{34}_{20}Ca$	2.512	−15.45	−17.52	0.061
$^{36}_{21}Sc$	2.792	−15.68	−18.02	3.671
$^{38}_{22}Ti$	3.242	−16.29	−18.97	0.301
$^{40}_{23}V$	2.142	−12.72	−14.13	2.681
$^{47}_{27}Co$	1.022	−1.19	0.823	2.12
$^{56}_{31}Ga$	2.822	−11.06	−13.14	3.141
$^{58}_{32}Ge$	3.232	−11.97	−14.49	0.541
$^{61}_{33}As$	1.982	−5.82	−6.43	3.041
$^{63}_{34}Se$	2.362	−7.48	−8.77	0.281

variable assault frequency and no spectroscopic factor have been added since the decay is considered as a fission like process. The values are slightly lower than the experimental data but there is a correct agreement at 20 orders of magnitude. The root mean square deviation σ between the experimental and theoretical (GLDM) decimal logarithms of the half-lives is 1.6, which compares favorably with the values obtained by different models and recalled in [22].

To describe analytically all the α-decay half-lives several formulas have been proposed to generalize the Geiger and Nutall law [37]. One of these formulas has the following form:

$$\log_{10}[T_{1/2}(s)] = a + bA^{\frac{1}{6}}\sqrt{Z} + \frac{cZ}{\sqrt{Q}}, \quad (14)$$

where A, Z, and Q are the mass and charge numbers of the emitter and Q_α value; a, b, and c being adjustable parameters.

For example, in Ref. [27], the following formula was proposed to determine the α-decay half-lives. It was fitted on 373 data and leads to a root mean square (rms) deviation of 0.42 between the experimental data and theoretical values of $\log_{10}(T_{1/2})$:

$$\log_{10}[T_{1/2}(s)] = -26.06 - 1.114A^{\frac{1}{6}}\sqrt{Z} + \frac{1.5837Z}{\sqrt{Q}}. \quad (15)$$

The same procedure has been applied for the $2p$ radioactivity on the 16 known experimental data. It leads to the following coefficients and formula:

$$\log_{10}[T_{1/2}(s)] = -24.054 - 1.541A^{\frac{1}{6}}\sqrt{Z} + \frac{1.501Z}{\sqrt{Q}}, \quad (16)$$

and a rms deviation σ of 1.00. The values are displayed in the sixth column of Table I. Once more, the terms of the formula (14) are very efficient. The adjustment is done from only 16 data which strongly limits the efficiency of extrapolation.

IV. PREDICTED HALF-LIVES OF OTHER ENERGETICALLY POSSIBLE 2p EMITTERS

The half-lives of the main energetically possible and still unknown $2p$ emitters have been determined within the GLDM and formula (16). The values are displayed in Table II. The unique input data is Q_{2p}. They have been extracted from the Nubase2020 [28]. Q_{2p} is the difference between the mass excess of the parent nucleus and the sum of the mass excess of the $2p$ system and the daughter nucleus. The mass excess of the $2p$ system is twice the proton mass excess since the $2p$ system is unbound. The Q_{1p} values are also given. Comparisons with other provided values [15,16,22,24] are difficult since they have been often calculated with the Nubase2016 data [29]. As examples, Table III indicates a change of some Q values between the Nubase2016 and Nubase2020 for several nuclei.

The mass excess of these nuclei are still only extrapolations with large uncertainty error bars for the mass of the parent and daughter nuclei.

The fact that a two-body approach and a quantum tunneling allow to reproduce the half-lives seems to indicate that the effect of the repulsion between the daughter nucleus and the two proton system play the main role at the beginning

TABLE III. Comparison between the Q_{2p}^{th} and Q_{1p}^{th} values given in Nubase2016 [29] and Nubase2020 [28].

Nucleus	Q_{2p}^{th} (MeV) (2016)	Q_{2p}^{th} (MeV) (2020)	Q_{1p}^{th} (MeV) (2016)	Q_{1p}^{th} (MeV) (2020)
${}^{26}_{16}$S	1.76	2.36	0.05	0.20
${}^{34}_{20}$Ca	1.47	2.51	−0.48	0.061
${}^{38}_{22}$Ti	2.74	3.24	0.06	0.30
${}^{42}_{24}$Cr	1.00	1.47	−0.88	−0.54
${}^{59}_{32}$Ge	2.1	1.60	0.38	−0.12

of the decay and during the tunneling of the potential barrier, the weaker repulsion between the two protons and the three-body process playing finally a main role for the angular distribution of the two separated protons at the end of the decay.

Naturally for *o-o*, *e-o*, *o-e* nuclei the angular momentum plays a role. Here, for $2p$ emission one knows, for only around 15 emitters, the experimental Q_{2p} and Q_{1p} values and with a large uncertainty. Furthermore, the Q_{2p} and Q_{1p} values calculated for other possible emitters are determined from a roughly extrapolated mass of the parent and daughter nuclei. Consequently, it is very difficult to adjust new terms in the formulas to take into account the angular momentum dependence. Furthermore, at least for the α emission, the improvement by the introduction of new terms depending on the angular momentum is relatively weak.

V. Be, Li, He, AND H DECAY HALF-LIVES AT LOW EXCITATION ENERGY

As for the α and $2p$ emissions, formulas given $\log_{10}(T_{1/2})$ have been researched for the ^{9}Be, ^{6,7}Li, ^{3,6}He, and ^{2,3}H emissions viewed as a very asymmetric fission without adjustable preformation factor. For these decays the Q value is negative and it is necessary to extend the formula (14) to take into account a low excitation energy E^*, lower than the decay barrier height to remain within the quantum tunneling approach. The experimental data do not allow to realize an adjustment and the following formulas have been obtained in adjusting on numerous nuclei and calculated with the GLDM for each cluster [44].

As an example, for the ^{209}Pb nucleus, the decay barrier height is 39.9 MeV for the emission of ^{9}Be, 36.8 for ^{7}Li, 37.6 for ^{6}Li, 29.2 for ^{6}He, 32.0 for ^{3}He, 20.2 for ^{3}H, and 19.7 for the ^{2}H.

For ${}^{9}_{4}$Be with $\sigma = 0.48$,

$$\log_{10}[T_{1/2}(s)] = \left(-31.69 - 2.238A^{\frac{1}{6}}\sqrt{Z} + \frac{4.47183\,Z}{\sqrt{Q+E^*}}\right)(1 + 2.384 \times 10^{-3}\,E^* - 9.556 \times 10^{-5}\,E^{*2}). \quad (17)$$

For ${}^{7}_{3}$Li with $\sigma = 0.38$,

$$\log_{10}[T_{1/2}(s)] = \left(-27.55 - 1.796A^{\frac{1}{6}}\sqrt{Z} + \frac{3.0016\,Z}{\sqrt{Q+E^*}}\right)(1 + 2.665 \times 10^{-3}\,E^* - 1.109 \times 10^{-4}\,E^{*2}). \quad (18)$$

For ${}^{6}_{3}$Li with $\sigma = 0.36$,

$$\log_{10}[T_{1/2}(s)] = \left(-27.45 - 1.647A^{\frac{1}{6}}\sqrt{Z} + \frac{2.8051Z}{\sqrt{Q+E^*}}\right)(1 + 3.107 \times 10^{-3}\,E^* - 1.213 \times 10^{-4}\,E^{*2}). \quad (19)$$

For ${}^{6}_{2}$He with $\sigma = 0.35$:

$$\log_{10}[T_{1/2}(s)] = \left(-24.85 - 1.424A^{\frac{1}{6}}\sqrt{Z} + \frac{1.8773Z}{\sqrt{Q+E^*}}\right)(1 + 2.873 \times 10^{-3}\,E^* - 1.288 \times 10^{-4}\,E^{*2}). \quad (20)$$

For ${}^{3}_{2}$He with $\sigma = 0.24$,

$$\log_{10}[T_{1/2}(s)] = \left(-23.60 - 1.003A^{\frac{1}{6}}\sqrt{Z} + \frac{1.3665Z}{\sqrt{Q+E^*}}\right)(1 + 3.896 \times 10^{-3}\,E^* - 1.662 \times 10^{-4}\,E^{*2}). \quad (21)$$

For ${}^{3}_{1}$H with $\sigma = 0.14$,

$$\log_{10}[T_{1/2}(s)] = \left(-22.65 - 0.7187A^{\frac{1}{6}}\sqrt{Z} + \frac{0.6775Z}{\sqrt{Q+E^*}}\right)(1 + 3.079 \times 10^{-3}\,E^* - 1.795 \times 10^{-4}\,E^{*2}). \quad (22)$$

For $^{2}_{1}$H with $\sigma = 0.16$,

$$\log_{10}[T_{1/2}(s)] = \left(-22.02 - 0.6039A^{\frac{1}{6}}\sqrt{Z} + \frac{0.5626Z}{\sqrt{Q+E^*}}\right)(1 + 3.060\times10^{-3}E^* - 1.871\times10^{-4}E^{*2}). \quad (23)$$

The one proton radioactivity has been soon studied with the GLDM [45].

VI. CONCLUSION

The $2p$ radioactivity can be viewed at the beginning of the process as a tunneling process through a potential barrier determined from the original GLDM and quasimolecular shapes. The sparse experimental data on the $2p$ radioactivity of the ^{12}O, ^{16}Ne, ^{19}Mg, ^{45}Fe, ^{48}Ni, ^{54}Zn, and ^{67}Kr nuclei are roughly reproduced using the Q_{2p}^{exp} value. A new analytic formula dependent on three parameters allows to determine rapidly these different half-lives, as for the α decay, but adjusted only on 16 experimental data. Extrapolations to other perhaps possible $2p$ emitters are proposed from the GLDM and the new formula. They are based on the Q_{2p} value extracted from the most recent but still extrapolated data provided in the Nubase2020 table. Formulas to calculate the ^{9}Be, ^{6,7}Li, ^{3,6}He, and ^{2,3}H decay half-lives at low excitation energy are also given.

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