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## Novel aspects of the carbon-decay mode of radium

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The effects of residual coupling interactions on the newly discovered decay $^{222}Ra \rightarrow {}^{14}C + {}^{208}Pb$ are investigated. The couplings to the low-lying collective excitations of $^{208}Pb$ produce an order of magnitude enhancement in the tunneling probability. A new possible mechanism for this decay is discussed whereby a $^{12}C$ fragment is transformed into $^{14}C$ by picking up two neutrons as it tunnels out.

Gamow's explanation of nuclear $\alpha$ decay in terms of tunneling through a potential barrier was one of the early, and enduring, triumphs of quantum mechanics.[1] Renewed attention has been focused on nuclear decay processes by the recent discovery of rare spontaneous carbon emission from radium isotopes.[2–4] A number of calculations have been reported[2,4–6] which attempt to describe these exotic decays. The conceptual basis for these calculations stems either from an application of Gamow's approach[2–4] or from the theory of nuclear fission[5,6] (see Ref. 5 for references to earlier works). In each case an essential step consists of calculating penetration probabilities for tunneling through a one-dimensional barrier.

During the last few years there has been, on the other hand, considerable interest in the tunneling phenomena in connection with studies of low-energy heavy-ion fusion reactions.[7] These fusion rates can be orders of magnitude larger than the results of conventional one-dimensional barrier penetration calculations. This has been explained by the fact that coupling interactions between the composite systems during the tunneling process generally enhance the penetration probability.[8] Such effects have also been considered previously in alpha-decay work. In particular, studies of the role played by the deformation of the daughter nuclei have a long history (see, e.g., Ref. 9 and references therein). In contrast, the new carbon-decay mode involves a spherical daughter and, as will be seen, allows for much stronger and more varied effects than have been discussed in connection with alpha decay.

The developments in the description of low-energy fusion reactions can be most easily applied to the problem of nuclear decay if one adapts the traditional picture which envisions the decay products as being preformed in their ground states and impinging on a confining barrier at a given rate. In this model the decay rate is given by

$$\lambda = \lambda_0 P_0 T \ ,$$

where $\lambda_0$ is the frequency of escape attempts, $P_0$ is the ground state preformation factor, and $T$ is the tunneling probability. In a conventional calculation $T$ would be computed using the WKB expression

$$T^{(0)}(E) = \exp\left[-2\int_a^b dr\,\sqrt{2\mu[U(r)-E]/\hbar^2}\right] \ ,$$

where $U(r)$ is the potential energy for the decay fragments, $\mu$ is their reduced mass, and $E=Q$ is the difference between the rest energies of the initial and final systems. The turning points $a,b$ occur where $U(r)=E$.

A more general expression would also account for the effect of the coupling interactions between the decay products during the tunneling process. These couplings affect the transmission coefficient in the ground state and also allow for transitions to final excited configurations. The tunneling probability for a given channel $\beta$ can be written as[10]

$$T_\beta = |C_\beta|^2 T^{(0)}(E-\epsilon_\beta) \ ,$$

where $\epsilon_\beta$ is the excitation energy of the state and $C_\beta$ is the amplitude of the channel wave function traveling past the barrier. For fusion reactions the waves are ingoing and one normally sums over all final states. In the decay problem the waves are outgoing and there is the possibility of observing these states separately.

It is interesting to note that the couplings generally induce transfer reactions as well as inelastic excitations. In particular, $\epsilon_\beta$ is negative for exothermic transfer processes. We shall return to this point below.

In this work we focus on the specific decay[4]

$$^{222}_{88}Ra \rightarrow {}^{208}_{82}Pb + {}^{14}_{6}C \ ,$$

which has a $Q$ value of 33 MeV. In general, one should consider excitations of both fragments as well as transfer modes. Here we first assess the effects of couplings to the low-lying collective states of $^{208}Pb$. This can be done con-

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veniently by treating these degrees of freedom as independent harmonic vibrations. Consider a set of such modes with frequencies $\omega_1, \ldots, \omega_N$. An approximate expression for the transmission probability leading to the final state with phonon occupation numbers $n_1, \ldots, n_N$ can be obtained by following the discussion in Ref. 10, starting from the inside of the barrier and working outwards. The result is

$$T_{n_1,\ldots,n_N} = e^{\Delta_0}\left[\prod_{i=1}^{N} \frac{(I_i^2)^{n_i}}{n_i!}\right] T^{(0)}\left[E - \sum_{i=1}^{N} n_i \hbar\omega_i\right] ,$$

where

$$\Delta_0 = \sum_{i=1}^{N} (2/\hbar^2) \int_0^\tau dt\, V_i[r(t)]e^{-\omega_i t} \int_0^t dt'\, V_i[r(t')]e^{\omega_i t'} ,$$

$$I_i = (1/\hbar) \int_0^\tau dt\, V_i[r(t)]e^{\omega_i t}$$

(for related work, see also Ref. 11). Here $V_i$ specifies the interaction which couples to the $i$th mode and $r(t)$ is the trajectory in the inverted potential which leads from $r(0) = a$ to $r(\tau) = b$, that is,

$$r(t) = a + \int_0^t dt \sqrt{2(U-E)/\mu} .$$

For the case of tunneling in the ground state the transmission probability leading to a decay rate $\lambda_G$ reduces to

$$T_G = e^{\Delta_0} T^{(0)}(E) .$$

The exponential gives an enhancement with respect to the conventional barrier penetration. It is also interesting to note that the rate $\lambda_1$ leading to a one-step channel excitation is such that

$$\lambda_1/\lambda_G = (I)^2 T^{(0)}(E - \hbar\omega)/T^{(0)}(E) \simeq (I)^2 e^{-2\omega\tau} .$$

This provides a simple basis for predicting relative decay rates.

For the $^{14}$C + $^{208}$Pb calculations we have scaled a nuclear potential $U_N$ used to describe $^{12}$C + $^{208}$Pb elastic scattering data.[12] The parameters of this Woods-Saxon potential are $V_0 = 40$ MeV, $r_0 = 1.26$ fm, and $a_0 = 0.56$ fm. The couplings are specified using the macroscopic model expression (see, e.g., Ref. 13)

$$V(r) = \frac{\beta R}{\sqrt{4\pi}}\left[-\frac{\partial U_N}{\partial r} + \frac{3Z_1Z_2e^2}{2L+1}\frac{R^{L-1}}{r^{L+1}}\right] ,$$

where $R$ is the nuclear radius, $\beta$ is the deformation parameter of the mode, and $L$ is its multipolarity. We included the low-lying $2^+$, $3^-$, $4^+$, and $5^-$ states of $^{208}$Pb. The corresponding excitation energies are 4.1, 2.6, 4.3, and 3.2 MeV while the deformation parameters are 0.06, 0.12, 0.07, and 0.07, respectively.[14]

With these parameters we obtain an enhancement factor

$$e^{\Delta_0} = 16$$

and a ground state transmission probability

$$T_G = 4.6 \times 10^{-25} .$$

The transmission probability to the other excited states is negligible. For instance, the decay to the octupole state is hindered by a factor of about $10^{-5}$ with respect to the ground state. The penetration factors are fairly stable with respect to variations of the nuclear potential in the interior side of the barrier because most of the tunneling occurs under the long-ranged Coulomb interaction.

Using the simple estimate

$$\lambda_0 = (\sqrt{2E/\mu})/R_0 ,$$

where $R_0$ is the radius of the parent nucleus and the measured decay rate (half-life $\tau_{1/2} = \ln 2/\lambda = 10^{11}$ s)[4] we obtain a preformation factor of

$$P_0(^{14}\mathrm{C}) \sim 0.5 \times 10^{-8} .$$

Similar calculations for the $\alpha$ decay of $^{212}$Po to $^{208}$Pb ($Q = 8.9$ MeV, $\tau_{1/2} = 0.3\ \mu$s) obtained using the $\alpha + {}^{208}$Pb elastic scattering potential of Ref. 15 ($V_0 = 96.4$ MeV, $r_0 = 1.07$ fm, $a_0 = 0.625$ fm) produce enhancements of only $\sim 1.2$ and consequently values of $P_0(\alpha) \sim 0.02$. The smaller enhancement in this case is due to somewhat weaker coupling strengths, combined with the smaller value of the reduced mass $\mu$. Likewise, we estimate $P_0(\alpha) \sim 0.04$ for the $\alpha$ decay of $^{222}$Ra to $^{218}$Rn ($Q = 6.7$ MeV, $\tau_{1/2} = 38$ s). In this way we obtain a ratio

$$P_0(^{14}\mathrm{C})/P_0(\alpha) \sim 10^{-7}$$

for the preformation of $^{14}$C in $^{222}$Ra relative to that for $\alpha$ particles.

Up to this point our analysis presumes that the observed $^{14}$C originated from a preformed $^{14}$C nucleus in the interior of $^{222}$Ra. From a nuclear structure point of view it may be argued that it is easier to cluster $^{12}$C. The fact that $^{14}$C is observed rather than $^{12}$C is currently attributed to the more favorable $Q$ value (by 4 MeV) for $^{14}$C decay.[2,5] However, the $^{12}$C-decay mode can gain back the difference in $Q$ value for the price of picking up two neutrons as it tunnels out. The first-order integral for this process is given by

$$I_T = (1/\hbar) \int_0^\tau dt\, V_T[r(t)]e^{-Q_T t/\hbar} ,$$

where $V_T$ now stands for the transfer coupling interaction and $Q_T$ is the transfer reaction $Q$ value. Using the macroscopic model for $V_T$ proposed in Ref. 16 we estimate

$$(I_T)^2 \sim 0.01$$

for the $^{210}$Pb ($^{12}$C,$^{14}$C) $^{208}$Pb reaction. With this value, the $^{12}$C-decay mode of $^{222}$Ra gives a ratio

$$\lambda_T/\lambda_G \sim 10^6 .$$

Thus if $^{12}$C is formed in $^{222}$Ra it will almost inevitably escape by transforming into $^{14}$C as it tunnels out. This may be the actual reason why $^{12}$C decay has not yet been identified. The fraction of observed $^{14}$C activity due to this mechanism depends mainly on the ratio of $P_0(^{14}\mathrm{C})$ to the product $P_0(^{12}\mathrm{C})\ I_T^2(^{12}\mathrm{C} \rightarrow {}^{14}\mathrm{C})$. For instance, based on the above estimate it would take a preformation factor $P_0(^{12}\mathrm{C}) \gtrsim 100 P_0(^{14}\mathrm{C})$ to make the formation of $^{12}$C followed by two-neutron transfer the dominant mechanism in the observed carbon-decay mode of radium.

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