Interpretation of the fine structure in the ¹⁴C radioactive decay of ²²³Ra

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The experimental hindrance factors determined from the fine structure in the ¹⁴C decay of ²²³Ra are strikingly similar to the hindrance factors observed in the alpha decay of odd-A reflection-asymmetric deformed nuclei in spite of the deformed to spherical shape which occurs in the ¹⁴C decay. Calculations of the overlap between the reflection-asymmetric ground state of ²²³Ra and the spherical shell-model orbitals of ²⁰⁹Pb involved in the ¹⁴C decay are consistent with the experimental hindrance factors from the ¹⁴C fine structure except that the $j_{15/2}$ orbital is more strongly populated experimentally than the calculations suggest.

Since the discovery¹ of the rare radioactive decay mode, $^{223}Ra \rightarrow ^{14}C + ^{209}Pb$, this decay has been verified² and rapid progress has been made in the experimental observation of other ^{14}C , ^{24}Ne , and ^{28}Mg radioactive decays.³ Very recently, the fine structure in the ^{14}C decay of ^{223}Ra has been observed by Brillard *et al.*,⁴ corresponding to the population of a few of the lowest-lying states in ^{209}Pb . The ^{223}Ra parent in this decay has a reflection-asymmetric deformed shape⁵ and the daughter, ^{209}Pb , with one neutron more than the double closed-shell nucleus, ^{208}Pb , is spherical with relatively pure singleparticle shell-model states. Our ability to explain the branching implied by the fine structure in this decay between such different nuclear structures is an interesting and important test of our knowledge of these structures.

In direct analogy with the pioneering work⁶ of Geiger and Nuttall and of Rosenblum on alpha decay, it has recently been recognized^{7,4} that the logarithm of the halflife in ¹⁴C decay from a sequence of even-even Ra isotopes, when plotted versus the inverse square root of the energy release in each radioactive decay ($Q^{-1/2}$), falls on a straight line. This plot for the ¹⁴C decay of ²²²Ra, ²²⁴Ra, and ²²⁶Ra (open circles) is shown as the solid straight line in Fig. 1.

Using the histogram of intensity versus energy observed in the fine-structure ¹⁴C decay of ²²³Ra by Brillard *et al.*,⁴ shown schematically in the decay scheme in Fig. 1, the solid points corresponding to this decay can be plotted. Figure 1 is similar to that of Brillard *et al.* except that we have plotted a third tentative solid point (shown in parentheses) corresponding to the weak third ¹⁴C group. Although the energy of this third group corresponds much better to the population of the $\frac{15}{2}^{-}$ 1423keV state in ²⁰⁹Pb, the limited resolution of the experiment cannot exclude the possibility that some of the intensity of this group results from the population of the $\frac{5}{2}^{+}$ 1567-keV state. Nonetheless, although we consider this point tentative, we have shown the point in Fig. 1 corresponding to the population of only the $\frac{15}{2}^{-}$ 1423keV state because of its better energy fit and our inability to divide the intensity between the two groups in any rational way.

As pointed out by Brillard *et al.*,⁴ since the solid points in Fig. 1 lie above the line, they represent hindrance factors (HFs) to ¹⁴C decay analogous to those observed in alpha decay to odd-*A* nuclei. If, as in alpha decay, the HFs of the ¹⁴C decay of the even-even Ra nuclei in Fig. 1 are assigned the value of 1, then the HFs of the ¹⁴C decays to the ²⁰⁹Pb ground state (g.s.), 779-keV, and 1427-keV states are 600 ± 120 , 3 ± 0.2 , and tentatively 3 ± 1.5 , respectively.⁴

Not only does the ¹⁴C decay of ²²³Ra represent a transition from a reflection-asymmetric shape to a spherical shape in ²⁰⁹Pb, but the spin changes are very large (involving transition from the $\frac{3}{2}^+$ ground state of ²²³Ra to the $\frac{9}{2}^+$, $\frac{11}{2}^+$, and $\frac{15}{2}^-$ states in ²⁰⁹Pb). In view of this, one might naively expect that the HFs observed would be large, whereas experimentally two of the three HFs are of the order of 3 implying favored ¹⁴C decay.

Since the HFs of the ¹⁴C decay of ²²³Ra are determined relative to the HFs of the ¹⁴C decay of ²²²Ra, ²²⁴Ra, and ²²⁶Ra defined as 1, the shape changes which also occur in the decay of the even-even Ra isotopes are taken out of consideration. This technique, first used in studying alpha decay, allows us to concentrate on the changes which occur in the configuration of the last odd neutron in the ¹⁴C decay of ²²³Ra to ²⁰⁹Pb.

The HF pattern observed in this 14 C decay, 600, 3, and 3 (high, low, and low), is precisely the type of HF pattern observed in the alpha decay of odd-*A* reflection-asymmetric deformed nuclei.⁸ For example, in the alpha decay of 227 Th to the ground state and low-lying bandheads with the the same parity mixed configuration in 223 Ra as in the parent, the HF patterns are 110, 5.6, and 14.⁵ This pattern results because the ground state of the parent is a parity mixed state very different from the ground state of the daughter but very similar to the parity mixed excited bandheads with the same configuration in the daughter. The high, low, and low HF pattern in the 14 C decay to shell-model states in 209 Pb implies that



FIG. 1. A plot of the logarithm of the partial half-life for ${}^{14}C$ decay against the inverse square root of the energy release in this decay for various Ra decaying parents. The ${}^{14}C$ decays of even-even Ra isotopes are shown with open circles and constitute the solid line which defines the hindrance factor as one. The hindrance factors of the ${}^{223}Ra$ decay are shown as solid points. Two of them fall on the dashed line corresponding to hindrance factor 3. The decays of ${}^{223}Ra$ to particular levels in ${}^{209}Pb$, indicated in the upper part of the figure, are connected with the appropriate hindrance factors by a line.

the shell-model parentage of the ²²³Ra ground state has low, high, and high components of the three lowest states in ²⁰⁹Pb. To check this idea we employ the parameters previously used^{5,9} to theoretically generate the band structure of ²²³Ra using the reflection-asymmetric rotator model. A reasonable description of the spectroscopic data is achieved at the deformations $\beta_2 = 0.129$, $\beta_3 = 0.10$, $\beta_4 = 0.075$, $\beta_5 = 0.01$, and $\beta_6 = 0.004$, although the $\Omega = \frac{1}{2}$ band immediately above the $\frac{3}{2}$ band requires $\beta_2 = 0.15$. The upper left-hand part of Fig. 2 illustrates how the single-neutron orbitals evolve in a continuous way from spherical shape to this strongly reflection-asymmetric deformation with $\beta_2 = 0.129$ and $\beta_3 = 0.10...$ Furthermore, in the upper right-hand part of Fig. 2, keeping all other deformations fixed at their "optimum values," we illustrate the dependence of the single-neutron orbitals on quadrupole deformation β_2 .

In the reflection-asymmetric rotor model, the ground state of ²²³Ra is mainly built from the $\frac{3}{2}^+$ state emerging from the $i_{11/2}$ orbital with small admixtures from other neighboring orbitals. Therefore, we illustrate in the lower parts of Fig. 2 the overlap between this $\frac{3}{2}^+$ orbital and the calculated orbitals for spherical shape. It is evident that, independent of quadrupole deformations, the main component is $i_{11/2}$ with small admixtures from $g_{7/2}$, $j_{15/2}$, and $g_{9/2}$. This is consistent with the favored decay to the $\frac{11}{2}^+$ state in ²⁰⁹Pb and also with the large hindrance for decay to the $\frac{9}{2}^+$ ground state. The favored decay to the $\frac{15}{2}^-$ state, however, is puzzling. Furthermore, the possibility that this decay is the $\frac{5}{2}^+$ state of ²⁰⁹Pb

would not help the interpretation, as the $\frac{3}{2}$ orbital for 135 neutrons in Fig. 2 has an even smaller $d_{5/2}$ than $j_{15/2}$ content.

In view of the uncertainty of the hindrance factor for ¹⁴C decay to the $\frac{15}{2}^{-}$ state of ²⁰⁹Pb, it seems questionable how serious the small calculated $j_{15/2}$ content of the $\frac{3}{2}^{+}$ ground state should be considered. There is, however, another factor which indicates that the negative-parity content of the orbital(s) used to build the ground state of ²²³Ra should be rather large. This is the splitting between the $\frac{3}{2}^{+}$ and $\frac{3}{2}^{-}$ bandheads of ²²³Ra which is experimentally observed as only 50 keV while the calculations of Ref. 5 give 160 keV.

The $\Omega = \frac{1}{2}$ orbital immediately above the $\Omega = \frac{3}{2}$ ground-state orbital has a 43% amplitude in the $j_{15/2}$ spherical band and also rather large amplitudes in the $i_{11/2}$, $d_{5/2}$, and $g_{7/2}$ orbitals but again a small amplitude (<2%) in the $g_{9/2}$ orbital. Therefore, if the $\Omega = \frac{1}{2}$ orbital were appreciably mixed into the $\frac{3}{2}$ + ²²³Ra bandhead, considerable $j_{15/2}$ orbital content could be understood theoretically. However, particular-rotor calculations of the type presented in Ref. 5 give a very pure (95–100%) $\Omega = \frac{3}{2}$ content to the bandhead in ²²³Ra. This is true if the Fermi level is placed on the $\Omega = \frac{3}{2}$ orbital, as was done in Ref. 5, or, as suggested by BCS calculations, it is placed in between this orbital and the $\Omega = \frac{1}{2}$ orbital.

The low $j_{15/2}$ content of the Fermi level $\frac{3}{2}$ orbital seems present also in other single-particle potentials. Thus, the positive-parity content of this orbital is approximately 80% in standard Folded-Yukawa calculations (Fig. 4 of Ref. 8) and it is more than 90% in the modified oscillator if the parameters of Ref. 10 are applied at the appropriate ϵ_2 deformation. Furthermore, in a recent calculation,¹¹ the bandheads of odd actinide nuclei have been minimized with respect to higher multipoles, β 5, β 6, and β 7 in addition to β_2 , β_3 , and β_4 . In this study, the Woods-Saxon potential with the same parameters as used here was employed while the Coriolis interaction was neglected. However, although rather large values of $\beta 5$, β 6, and β 7 were obtained for the $\frac{3}{2}^+$ ground state of ²²³Ra, the positive-parity content of the corresponding single-particle orbital remains large, 91%.

It would be very valuable to have additional experimental work on the ¹⁴C fine-structure decay of ²²³Ra with higher resolution. This might allow the resolution of the groups populating the $\frac{15}{2}^-$ and $\frac{5}{2}^+$ states in 209 Pb and determine more conclusively the HF for populating the $\frac{15}{2}$ state. It might also determine whether the $\frac{1}{2}$ state at 2032 keV in ²⁰⁹Pb is also populated. Since the data⁴ hint at this population, and the implied HF would be quite low, it could be significant. It would also be of considerable interest to undertake new theoretical studies of the spectroscopy of ²²³Ra. In particular, it would be important to understand why the $\Omega = \frac{1}{2}$ bands are best described with $\beta_3 = 0.15$ while all other bands are best described with $\beta_3 = 0.10$. This understanding could give additional insight into the $j_{15/2}$ content of the ²²³Ra ground state.

When this paper was in preparation, we learned of a

paper¹² somewhat similar to the present one. Thus, the same idea that the spherical components of the ²²³Ra ground state should determine the ¹⁴C HFs is put forward. However, no calculation of the single-particle structure is performed; instead it is assumed that the ground state is a mixture of $j_{15/2}$ and $i_{11/2}$ components of essentially equal amplitudes. The present analysis shows that this is not consistent with standard models which give a much smaller $j_{15/2}$ than $i_{11/2}$ content in the ground state.

In summary, the HFs determined from the fine structure in the ¹⁴C decay of ²²³Ra are strikingly similar to HF patterns in the alpha decay of odd-*A* reflectionasymmetric deformed nuclei. Of special significance in the cluster decays of the ¹⁴C type is the natural occurrence of a deformed mother nucleus and a spherical daughter nucleus. This results in direct experimental information on the spherical components in the deformed mother nucleus. Calculations suggest a low HF for the population of the 779-keV $\frac{11}{2}^+$ state and a high HF for the population of the $\frac{9}{2}^+$ ground state in ²⁰⁹Pb as experimentally observed. However, the low HF for the population of the 1423-keV $\frac{15}{2}^{-}$ state and the related large $j_{15/2}$ in the ²²³Ra ground state does not come out in standard calculations. This disagreement between theory and experiment is further accentuated by the measured small splitting between the $\frac{3}{2}^{+}$ and $\frac{3}{2}^{-}$ bandheads of ²²³Ra, which also suggests large components of $j_{15/2}$ in the ²²³Ra ground band. It is an important task for future studies of the structure of octupole deformed nuclei to explain the reason for this discrepancy and to find some more consistent description. It will also be important to study the fine structure of the ²⁴Ne decay of the $\frac{5}{2}^{+}$ [633] ground state of ²³³U into the $g_{9/2}$ and $j_{15/2}$ shell-model states of ²⁰⁹Pb. In this case, the transition to the $g_{9/2}$ state should be allowed while the degree of forbiddenness of the transition to the $j_{15/2}$ state will indicate residual octupole correlation in ²³³U.

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FIG. 2. Calculation of the shell-model composition of the $\Omega = \frac{3}{2}$ ground-state orbital of ²²³Ra. In the upper figure parts, the Woods-Saxon neutron orbitals are drawn where the dashed line corresponds to the "optimal deformation" of ²²³Ra, $\beta_2=0.129$, $\beta_3=0.10$, $\beta_4=0.075$, $\beta_5=0.01$, and $\beta_6=0.004$. The orbitals are labeled by the spin projection on the symmetry axis, Ω . On the upper left-hand side of the figure, starting from spherical shape, all five deformation parameters increase in a linear way, while, on the upper right-hand side, only the quadrupole deformation varies with all other parameters kept at their "optimal values." In the lower parts, the squared overlap between the $\Omega = \frac{3}{2}$ Fermi level orbitals for N=135 (drawn by a thick line in the single-particle diagrams) and the most important $\Omega = \frac{3}{2}$ spherical orbitals is given. Note that for all deformations shown, the deformed $\frac{3}{2}$ orbital is mainly of $i_{11/2}$ character and that the $g_{9/2}$ content is small. The Woods-Saxon parameters are those of Ref. 10 which were also used in Refs. 5 and 9.

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