Molecular Alpha-Particle Clustering in ^{218}Ra; Dipole Collectivity in the Vicinity of Nuclear Shell Closures

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Evidence for the presence of alpha-particle clustering in ^{218}Ra comes from a number of observables: binding energy, S_{2n}, Q_α, $E_x(J^\pi)$, θ_α^2, and F_α. That this clustering is a signature for the new dipole collectivity suggested by Iachello and Jackson follows from observation of simultaneous enhancement of selected $E1$, $E2$, and $E3$ deexcitation transition matrix elements; of these the $E1$ enhancement is most pronounced as would be expected for dipole collectivity.

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The existence of alpha clustering in the low-lying states of light nuclei, particularly in the ^{16}O–^{20}Ne region, is now well established.[1] Recently published[2] systematics for ground-state alpha-particle widths for heavy nuclei also indicate large reduced alpha-particle widths in the vicinity of the $Z=50$ and 82 shell closures. It bears noting that the largest ground-state reduced alpha-particle width for any heavy nucleus yet reported is that[2] for ^{218}Ra; it exhausts 75% of the Wigner sum-rule limit. These large ground-state reduced widths (θ_α^2) suggest that configurations such as $\alpha + A_2$ may indeed exist in the vicinity of shell closures. If the overlap between the alpha cluster and the core nucleus (A_2) is small (as suggested by the large θ_α^2), it may be appropriate to view these states as physically real *molecular* alpha-particle cluster states. Such states could then be viewed as the nuclear equivalents of the HBr or HI diatomic molecules which also invoice consitituents of very different sizes.

Recently, the possible importance of such alpha-particle clustering in heavy nuclei was emphasized and a phenomenological description was proposed[3] in which the cluster states are associated with a new molecular dipole degree of freedom.[4] The model is developed within the context of a spectrum-generating algebra to emphasize the intrinsic symmetries involved; in this case the cluster is characterized by the length and orientation of the vector separating the α and A_2 centers of mass and thus is associated with a dipole degree of freedom and with S and P_μ ($-1 \leqslant \mu \leqslant 1$) bosons—the generators of U(4).

In addition to such molecular states it would be expected, of course, to find the normal quadrupole collective states in the low-energy excitation spectra of these heavy nuclei—and indeed dipole and quadrupole states having the same J^π would be expected to mix. The result of such mixing of the normal quadrupole ground-state band having a sequence $0^+, 2^+, 4^+, 6^+, \ldots$, and the molecular dipole band[3] having sequence $0^+, 1^-, 2^+, 3^-, 4^+, \ldots$ then leads to low-lying 1^- states, as observed,[5] a sequence $0^+, 2^+, 1^-, 4^+, 3^-$, again as observed, as well as large ground-state alpha-particle reduced widths and small relative alpha-particle hindrance factors for excited states. In addition, higher-lying states—$0_2^+, 2_2^+, \ldots$—with small hindrance factors are expected. For *high-spin states* it remains possible that the mixing of the ground-state band and the cluster band may not be as important, leaving the interleaved sequence $4^+, 5^-, 6^+, 7^-, \ldots$.

We report herein our experimental studies on ^{218}Ra; these were designed to test the validity of the molecular alpha-particle clustering picture of heavy nuclei[3] and to search for evidence for the postulated dipole collectivity.

The expected electromagnetic deexcitation width (Γ_γ) of molecular states can be estimated by evaluating molecular sum rules,[6] as well as by considering a classical geometric model[6] for molecular states. It would be expected that molecular states having large θ_α^2 should also have Γ_γ values that exhaust a large fraction of this molecular radiative sum rule. Such a simple classical model requires that all three of $B(E1)$, $B(E2)$, and $B(E3)$ deexcitation matrix elements are enhanced for transitions within a molecular dipole band; enhancement of $B(E1)$ arising from the displacement of the center of charge from the center of mass is predicted to be the most striking.

We recently reported[7] on the possibility of collective dipole states comprising an $\alpha + ^{14}$C molecular band in ^{18}O having spin sequence $0^+, 1^-, 2^+, 3^-, (4^+)$, large alpha widths ($\theta_\alpha^2 \approx 20\%$), and enhanced $E1$ matrix elements [$B(E1) \sim 10^{-2}$ W.u.];

these $B(E1)$ values are the largest thus far reported in even-even nuclei; they correspond to 13% of the molecular $E1$ sum rule.[6] The bandhead in this case is the well-known four-particle, two-hole (4p-2h) 0_2^+ state at 3.63 MeV in ^{18}O.

In this Letter we report on our studies of an alpha-particle–cluster molecular band in ^{218}Ra involving the nonyrast low-spin and *high-spin states* in ^{218}Ra, and their $E1$ and $E2$ deexcitation modes. We have found that the molecular states in these bands display enhanced electric deexcitation matrix elements of many multipolarities: $E1$, $E2$ (and $E3$) simultaneously. The enhancement of the $B(E1)$ values has been found to be the most prominent.

We have studied ^{218}Ra via the reaction ^{208}Pb(^{13}C, $3n$)^{218}Ra through measurements of gamma-ray excitation functions, alpha-particle excitation functions, gamma-gamma coincidences, gamma-ray angular distributions, and alpha-gamma delayed coincidences. All these studies were carried out with use of ^{13}C beams from our model MP1 tandem accelerator at 67.0 MeV where the $3n$ channel cross section maximizes, as well as at 59.0 and 59.5 MeV, near the Coulomb barrier, where we have attempted to minimize the angular momentum in the entrance channel of interest and thus emphasize the population of low-spin (1^-, 3^-), nonyrast, high-lying states systematically appearing in this region.[8] A Doppler-shift recoil distance measurement (RDM) was used to measure the $B(E2)$ decay matrix elements of the 2^+ and 4^+ states. A similar measurement[9] on only the high-spin states of ^{218}Ra shows more limited but consistent results. Our RDM experiment and excitation curve near the barrier (58.5 to 60.0 MeV) yield ordering of the $4^+ \to 2^+$ and $2^+ \to 0^+$ transitions opposite to that suggested in Ref. 9.

The level spectrum of ^{218}Ra obtained from our work is shown in Fig. 1. The high-spin structure of ^{218}Ra indeed shows the interleaved spin sequence $4^+, 5^-, 6^+, 7^-, 8^+, 9^-, \ldots, 15^-, 16^+, 17^-$. Individual $E1/E2$ branching ratios exhibit a very similar ratio: $7.0\times10^{-5} \leq B(E1)/B(E2) \leq 9.5\times10^{-5}$ (in Weisskopf units). Note that these branching ratios do not vary as much as those reported in Ref. 9. The RDM measurement yields $B(E2{:}2^+\to0^+) = 80\pm20$ W.u. and $B(E2{:}4^+\to2^+) = 125\pm30$ W.u. as shown in Fig. 2. Several experimental difficulties are reflected in the large error bars. To avoid the large background produced by commonly used metallic stopper foils we used Mylar stopper foil. A much preferable experimental arrangement could be achieved by use of a ^{208}Pb beam and a reversed reaction; however, if these $B(E2)$ values are extrapolated to the high-spin states we find $B(E1) \approx 10^{-2}$ W.u. for these as plotted in Fig. 2. These would clearly reflect very enhanced $E1$ matrix elements; indeed these would be the fastest $E1$ transitions yet found in heavy nuclei. We thus clearly find in the high-spin structure of ^{218}Ra evidence for alpha-particle clustering through the interleaved spin sequence $4^+, 5^-, 6^+, \ldots, 15^-, 16^+, 17^-$ and through the enhancement of the $B(E1)$ matrix elements.

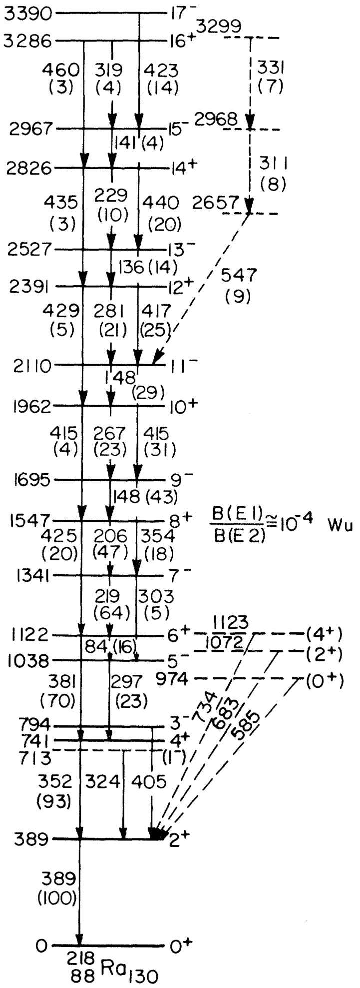

FIG. 1. High- and low-spin states of ^{218}Ra. Individual $B(E1)/B(E2)$ branching ratios, deduced from the shown intensities, are almost constant. The molecular dipole cluster band comprises alternating parity states with enhanced intraband $E1$ transitions.

As already noted[5] the excitation energy of the $J^\pi = 1^-$, 3^- states in this region reaches a low value of ~200 keV. However, additional increase of the excitation energy of the $J^\pi = 1^-$ states as N

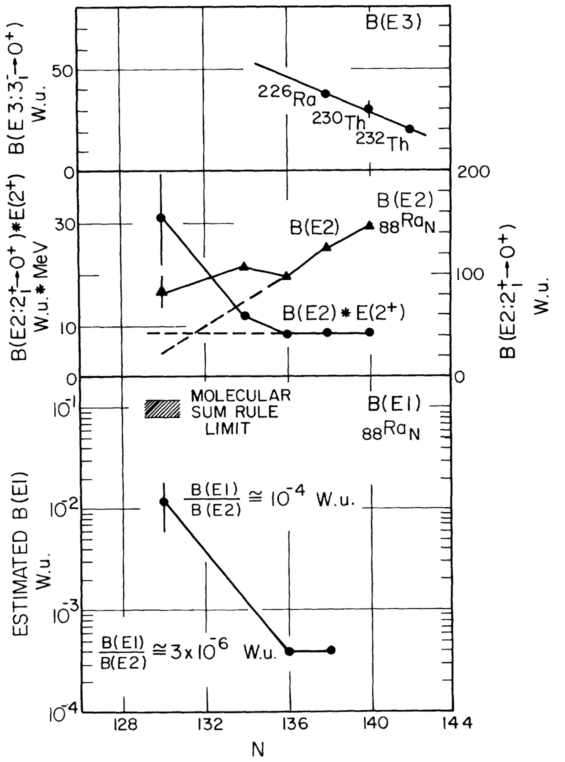

FIG. 2. Enhanced $E1$, $E2$, and $E3$ transitions in ^{218}Ra. The extracted $B(E1)$ matrix elements are the largest known in heavy nuclei. Also, the reduced alpha widths in ^{218}Ra are the largest (Ref. 2) known in heavy nuclei ($\theta_\alpha^2 = 75\%$). The $E2$ enhancement is more than expected for regular collective states (Ref. 10) [characterized by $B(E2)\,E(2^+) = \mathrm{const}$]. The molecular dipole cluster states show extra enhancement of their $E1$, $E2$, and $E3$ deexcitation matrix elements as discussed in Ref. 6.

approaches 126 is observed[8]; it arises from the decrease in the energy of the ground states (i.e., larger binding energies) rather than from an increase in the intrinsic excitation of the 1^- state itself. In the vicinity of the $N = 126$ shell closure, we find evidence for extra binding of the Nth nucleus with respect to the $(N+2)$nd nucleus. That extra binding is clearly observed as a deviation from linearity of the two-neutron separation energy (S_{2n}), which is usually linear in N. This extra binding is, in fact, equal to the increase in the excitation energy of the $J^\pi = 1^-$ state. The observed extra ground-state binding could possibly reflect mixing of the closely lying 0^+ alpha-particle-cluster bandhead and the 0^+ quadrupole ground state with consequent depression of the latter and elevation of the former. In contrast the alpha-particle separation energies (Q_α) exhibit a linear dependence on N. In the cluster model this can be understood as a cancellation of the extra binding evident for ^{218}Ra and ^{222}Th. The rise in the excitation energy of the $J^\pi = 1^-$ state (toward $N = 126$) is also correlated with the well-known decrease[5,8] in the hindrance factor (F_α) by ~1000 and the increase in θ_α^2 for the ground state,[2] all suggesting that in these light actinide nuclei the molecular cluster band lies low, close to the ground-state band with which it mixes.

It has been suggested previously[11,12] that the low energy of the $J^\pi = 1^-, 3^-$ states in this region is evidence for the existence of permanent octupole shapes. We note first that, as shown in Ref. 6, alpha-particle-cluster states yield permanent (pear-shape) octupole moments, but also yield polarization (dipole moment) and deformation (quadrupole moment). It will be of interest to study further the detailed connection between alpha-particle cluster and permanent octupole deformation. The large ground-state reduced alpha width for ^{218}Ra ($\theta_\alpha^2 = 75\%$), however, provides support for the reality of the cluster interpretation.[3]

We have already noted that the $B(E1)$ matrix elements in ^{218}Ra are large. In Fig. 2, we compare these $B(E1)$ values to those of ^{226}Ra[13] and ^{224}Ra.[8] We clearly observe an enhancement of $B(E1)$ in moving toward the light Ra isotopes. The extrapolated $B(E1{:}0^+ \to 1^-)$ in ^{218}Ra exhausts 15% of the molecular sum rule[6] for $E1$ transitions. It is also well known[10] that for regular quadrupole collective states of even-even nuclei the product $B(E2{:}2_1^+ \to 0^+)E(2^+)$ is a constant (proportional to Z^2/A), as observed for the heavier Ra (and Th) isotopes, as shown in Fig. 2. We observe an additional enhancement of $B(E2{:}2^+ \to 0^+)$ for ^{218}Ra, as well as ^{222}Ra (and ^{226}Th),[8] over that predicted for normal collective states[10]; a rotational cluster model,[6] on the other hand, predicts an enhancement of a factor of 4 beyond that of the collective model.[10] Unfortunately, no data are available as yet on $B(E3{:}3_1^- \to 0^+)$ in ^{218}Ra. The only three known $B(E3)$ for stable targets in the region[13,14] suggest an increase in $B(E3)$ toward $N \geqslant 126$; a rotational cluster model predicts an enhanced $B(E3)$ of 58 W.u. for ^{218}Ra.[6] As shown then in Fig. 2, all the low electric multipoles are enhanced in ^{218}Ra, but the enhancement of the $B(E1)$ is most striking; similar behavior is observed[15] for ^{220,222}Th where only the yrast high-spin states were measured and only relative $B(E1)$'s are reported. We emphasize that the nonyrast low-lying 1^- and 3^- states are, however, essential for the molecular cluster interpretation.

To conclude, we find evidence for alpha-particle clustering in the light Ra isotopes and in ^{218}Ra, in particular. The alpha-particle–cluster states are low lying near shell closures and a signature for them is found in incremental and simultaneous enhancement of $E1$, $E2$, and possibly $E3$ transition rates. The molecular alpha-particle–cluster states exhibit a new collective dipole behavior near shell closures. A complete description of these heavy actinide nuclei, however, will require that the cluster dipole degree of freedom be treated in coexistence with the other well recognized quadrupole and octupole collective degrees of freedom, much as in the coexistence observed[7] in ^{18}O.

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[1] A. Arima, in *Clustering Aspects of Nuclear Structure and Nuclear Reactions*, edited by W. T. H. Van Oers *et al.*, AIP Conference Proceedings No. 47 (American Institute of Physics, New York, 1978), p. 1.

[2] E. Roecki, in Proceedings of the International Conference on Nucleus-Nucleus Collisions, Michigan State University, September 1982, Nucl. Phys. A400, 131c (1983).

[3] F. Iachello and A. D. Jackson, Phys. Lett. 108B, 151 (1982); F. Iachello, Nucl. Phys. A396, 233c (1983).

[4] F. Iachello, Phys. Rev. C 23, 2778 (1981).

[5] F. S. Stephens, Jr., F. Asaro, and I. Perlman, Phys. Rev. 100, 1543 (1955).

[6] Y. Alhassid, M. Gai, and G. F. Bertsch, Phys. Rev. Lett. 49, 1482 (1982).

[7] M. Gai, M. Ruscev, A. C. Hayes, J. F. Ennis, R. Keddy, E. C. Schloemer, S. M. Sterbenz, and D. A. Bromley, Phys. Rev. Lett. 50, 239 (1983).

[8] C. M. Lederer and V. S. Shirley, *Table of Isotopes* (Wiley, New York, 1978).

[9] J. Fernandez Niello, H. Puchta, F. Riess, and W. Trautmann, Nucl. Phys. A391, 221 (1982). Y. Gono *et al.* (private communication) report the 4^+ and 2^+ as suggested in Fig. 1.

[10] L. Grodzins, Phys. Lett. 2, 88 (1962).

[11] A. Gyurkovich, A. Sobiczewski, B. Nerlo Pomorska, and K. Pomorski, Phys. Lett. 105B, 95 (1981).

[12] G. A. Leander, R. K. Sheline, P. Möller, P. Olanders, I. Ragnarsson, and A. J. Sierk, Nucl. Phys. A388, 452 (1982).

[13] C. Mittag, R. Zimmerman, and J. de Boer, unpublished; J. de Boer, private communication (to be published).

[14] F. K. McGowan, C. E. Bemis, Jr., W. T. Milner, J. L. C. Ford, Jr., R. L. Robinson, and P. H. Stelson, Phys. Rev. C 10, 1146 (1974).

[15] D. Horn, G. D. Dracoulis, A. P. Byrne, and C. H. Fahlander, private communication; D. Ward, G. D. Dracoulis, J. R. Leigh, R. J. Charity, D. J. Hinde, and J. D. Newton, to be published; W. Bonin, M. Dahlinger, S. Glienke, E. Kankeleit, M. Kramer, D. Habs, B. Schwartz, H. Backe, Z. Phys. A 310, 249 (1983).