Sign of the Hexadecapole Moments of ^{232}Th and ^{238}U Nuclei*

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The ambiguity in the sign of the hexadecapole moments of ^{232}Th and ^{238}U, as determined from α-particle Coulomb-excitation experiments, has been resolved by experiments using 145-MeV ^{40}Ar to multiply Coulomb excite these nuclei up to their 12^+ states. Excellent agreement with the experimental results was obtained with predictions from the Winther–de Boer program when positive $M(E4)$ values were used, while significant disagreement was found when the negative $M(E4)$ values were used.

Within the past year several groups have been able to demonstate the presence of significant hexadecapole deformation in some deformed nuclei by means of α-particle Coulomb excitation. Stephens *et al.*[1,2] utilized γ-ray measurements in studies of ^{152}Sm and ^{154}Sm. With a high-resolution magnetic spectrograph to measure the inelastically scattered ^{4}He, McGowan *et al.*[3] and Bemis *et al.*[4] found very large $E4$ moments in a number of heavy deformed nuclei. Similar measurements have been made by Erb *et al.*[5] for rare-earth nuclei. These experiments utilize the same rationale: The excitation of the 4^+ state is calculated assuming multiple $E2$ excitation; the experimental cross section is extracted; and the excess in the experimental cross section to the 4^+ state is ascribed to $E4$ excitation. All of these measurements suffer from the same ambiguity; namely, they yield two solutions since there is a quadratic relationship between the excess 4^+ yield and the $E4$ matrix element. From the results of McGowan *et al.*[3,4] on ^{238}U, for example, $\langle 4^+\|\mathfrak{M}(E4)\|0^+\rangle$ values of $+1.22e$ b^2 or $-1.86e$ b^2 are both consistent with the computed probabilities. The profound effect of the choice between the positive and negative values of $\langle 4^+\|\mathfrak{M}(E4)\|0^+\rangle$ for ^{238}U is illustrated by Fig. 1. The shapes are the two possibilities obtained by calculating β_2 and β_4, assuming a homogeneous charge distribution. Although theoretical treatments of the nuclear shapes[6,7] and α decay properties[8,9] suggest positive values for most of the nuclei under study, it seems important to establish experimentally the correct sign of the hexadecapole moment.

We have been able to resolve the ambiguity present in the α-particle results by performing multiple Coulomb excitation of the ground-state rotational band of ^{232}Th and ^{238}U with ^{40}Ar. The calculated probabilities P_8, P_{10}, and P_{12} show a strong dependence on the $M(E4)$ values, permitting an unequivocal choice of sign. A somewhat related conclusion was reached by Winkler.[10]

The Coulomb-excitation experiments were performed with 145-MeV ^{40}Ar beams from the Oak Ridge isochronous cyclotron. At this bombarding energy the distance of closest approach between the surfaces of the ^{40}Ar nucleus and the ^{232}Th or ^{238}U nucleus would be 7.5 fm, assuming a sharp cutoff and a radius parameter r_0 of 1.2 fm. This is considered to be more than adequate to make nuclear interference with the Coulomb-excitation process unimportant.

We used the traditional backscattered heavy-ion γ-ray coincidence approach, complemented by simultaneous storage of singles γ-ray spectra. The heavy-ion detector was a 100-μm-deep, 200-mm^2 annular surface-barrier Si diode placed at a geometry which yielded an average scattering angle of 168°. The 6.5% coaxial Ge(Li) detector was placed at 90°, 2.5 cm from the target; its absolute efficiency was measured with calibrated sources to an accuracy of ~ 5%.

The targets of ^{232}Th and ^{238}U were metallic, 140 and 85 mg/cm^2, respectively, thick enough to stop the recoiling target nuclei as well as the

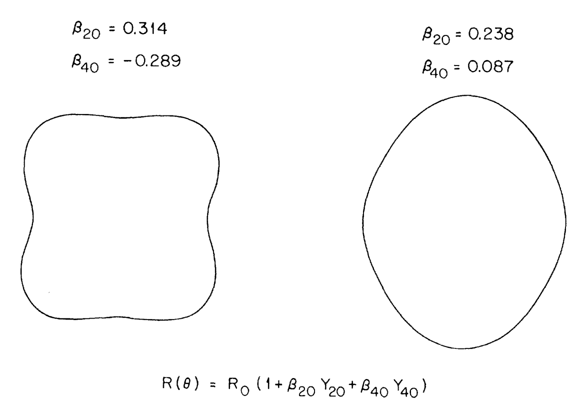

FIG. 1. The two shapes of the ^{238}U nucleus obtained by using the positive and negative $E4$ moments and by assuming a homogeneous charge distribution.

incident projectiles. The low-energy cutoff in the backscattered ^{40}Ar spectrum was set at ~20 MeV which corresponds to ~110 MeV in the incident frame.

In the coincidence spectra we observed transitions between members of the ground-state band up to and including the 12^+ state for both nuclei. Since the Ge(Li) detector was at 90° to minimize Doppler broadening, sizable angular-distribution corrections were necessary. To test for possible attenuation of the γ-ray angular distributions, we made separate measurements at 0°, 45°, and 90° with the Ge(Li) detector 6 cm from the ^{232}Th and ^{238}U targets. For the 4^+, 6^+, and 8^+ states the distributions agreed to within 5% with the results obtained by utilizing the theoretical parameters from the Coulomb-excitation program.[11] Thus, in the results quoted here the theoretical parameters are used. The finite solid-angle corrections were made using the estimates of Camp and Van Lehn.[12]

To correct for internal conversion of the observed transitions we used the calculations of Hager and Seltzer.[13] The major source of uncertainty in the absolute experimental probabilities is in the efficiency values of the Ge(Li) detector. In the case of the 10^+ and 12^+ states, the statistical uncertainty of the areas of the corresponding γ-ray peaks also contributes in an important way.

All of the experimental-to-theoretical comparisons utilized the Winther–de Boer[11] coupled-equations Coulomb-excitation program in a form modified by Holm[14] to accept multipolarities from 1 to 4, and by Sayer[15] to handle thick targets. The stopping-power values are from Northcliffe and Schilling.[16] We assumed that the rotational model is obeyed rigorously and that given the matrix elements $\langle 2^+\|\mathfrak{M}(E2)\|0^+\rangle$ and $\langle 4^+\|\mathfrak{M}(E4)\times\|0^+\rangle$, the elements joining all other states could be obtained by ratios of Clebsch-Gordan coefficients with appropriate spin normalization. The variation of Coulomb-excitation probabilities P_8, P_{10}, and P_{12} as a function of the value of $E4$ matrix element is shown graphically for ^{238}U in Fig. 2. The cross-hatching represents the experimental values and uncertainties, and the arrows near the abscissa indicate the values of McGowan *et al.*[3,4] Clearly, the positive value satisfies both the α-particle and ^{40}Ar results. No plot was made for P_6 since the variation with $M(E4)$ is less than 1% and in excellent agreement with the experimental data.[4] The calculated possibilities using the data of McGowan *et al.*[3,4] for

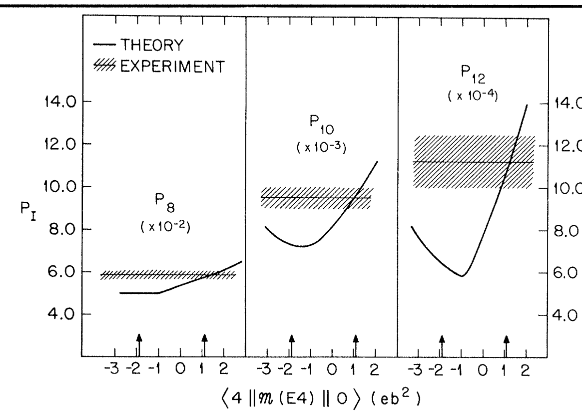

FIG. 2. Theoretical Coulomb-excitation probabilities for the 8^+, 10^+, and 12^+ states in ^{238}U as a function of $\langle 4^+\|\mathfrak{M}(E4)\|0^+\rangle$ values compared to the experimental results. The arrows indicate the two values of $\langle 4^+\|\mathfrak{M}(E4)\times\|0^+\rangle$ from Refs. 3 and 4.

^{232}Th and ^{238}U are compared with our experimental results in Table I. There is rather good agreement between the experimental results and the theoretical values using the positive hexadecapole moments as compared with the large disagreements for the 8^+, 10^+, and 12^+ states using the negative $M(E4)$ values. The P_4 value for ^{232}Th was extracted from the singles spectrum and its probability normalized to the 6^+ experimental/theoretical ratio. A similar computation for P_8 in ^{232}Th gave 0.99 ± 0.05.

We must answer three questions about the theoretical results used to obtain the values in Table I: (1) What is the effect of including ground- to vibrational-state matrix elements in the Winther–de Boer program? (2) How large are quantal effects? (3) What is the effect of centrifugal stretching?

We performed calculations to test the effect of including vibrational states in the Winther–de Boer program. We used the 5.5-MeV proton and 18-MeV α-particle Coulomb-excitation measurements of McGowan *et al.*[17] for the $B(E2)$ and $B(E3)$ values for excitation of β, γ, and three octupole bands in ^{238}U. We ran four cases: (1) the complete β band to the 16^+ state; (2) the complete γ band to the 13^+ state; (3) the complete $K=0$ octupole band to the 15^- state; and (4) a case including the states actually observed in our measurements, the 1^-, 3^-, and 5^- of the $K=0$, the 1^-, 2^-, and 3^- of the $K=1$, the 2^- and 3^- of the $K=2$ octupole bands, the 0^+ and 2^+ of the β band, and the 2^+ of the γ band. The effects observed were from 1 to 3% for P_6, P_8, and P_{10} and from 1 to 8% from P_{12}.

TABLE I. Ratios of experimental to theoretical Coulomb excitation probabilities for the ground-state rotational bands in ^{238}U and ^{232}Th.

Level	Experiment/Theory		
	$M(E2)$ only	$M(E4)$ (+)	$M(E4)$ (−)
		^{238}U + 145.8 MeV ^{40}Ar	
6^+	1.00±0.05[a]	1.00±0.05	1.00±0.05
8^+	1.08±0.05	1.01±0.05	1.21±0.06
10^+	1.21±0.08	1.05±0.07	1.40±0.09
12^+	1.30±0.20	1.00±0.14	1.76±0.25
-	-	-	-
$\langle 2^+\|\mathcal{M}(E2)\|0^+\rangle=$	3.421	3.421	3.377
$\langle 4^+\|\mathcal{M}(E4)\|0^+\rangle=$	0	1.12	-1.86
		^{232}Th + 145.8 MeV ^{40}Ar	
4^+		1.05±0.05[b]	
6^+	1.01±0.05	1.01±0.05	1.04±0.05
8^+	1.18±0.05	1.08±0.05	1.29±0.07
10^+	1.18±0.08	0.92±0.07	1.24±0.09
12^+	1.65±0.25	1.01±0.15	1.70±0.25
-	-	-	-
$\langle 2^+\|\mathcal{M}(E2)\|0^+\rangle=$	3.033	3.033	3.008
$\langle 4^+\|\mathcal{M}(E4)\|0^+\rangle=$	0	1.22	-1.91

[a] Indicated errors reflect only the experimental uncertainty.
[b] Extracted from singles data by normalization to P_6.

Quantum-mechanical corrections to the probabilities calculated by the Winther–de Boer semiclassical treatment can be very significant for the 2^+ and 4^+ states as shown by Alder, Morf, and Roesel.[18,19] This group[20] has recently extended their calculated quantal corrections to the 6^+ and 8^+ states for $E2$ only. Extrapolation of their results to our case would lower the theoretical probabilities by about 2%, and 4% for the 6^+ and 8^+ states. The trends observed for the lower states lead us to expect corrections in the range of 10 to 15% for the 10^+ and 12^+ states.

Changes in the $M(E2)$ values due to centrifugal stretching would increase the theoretical probabilities. If stretching is estimated from the α parameter extracted from a least-squares fit to the ground-state–band energy levels, P_8, P_{10}, and P_{12} would increase by about 5, 10, and 20%, respectively. If the stretching was estimated from the β-band branding ratios,[17] somewhat smaller effects would be observed.

Since the quantal effects reduce the probabilities, while stretching and inclusion of vibrational states increases them, there is a cancelation. The estimated net effects from these three sources are 4% or less for P_8, P_{10}, and P_{12}.

Even higher-order contributions to the ground-state nuclear shape, such as β_6, are theoretically possible. However, since we cannot include the corresponding $E6$ matrix elements in the current version[14] of the Winther–de Boer program, a test of their effect on the Coulomb-excitation process is not possible.

In addition to the determination of the sign of the $E4$ moment described here, we have obtained new spectroscopic information concerning ^{238}U and ^{232}Th, which extends the earlier measurements of Diamond and Stephens,[21] McGowan *et al.*,[17] and Milner *et al.*[22] This information will be covered in a forthcoming publication.

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Multipole Deformation of ^{238}U†

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Elastic and inelastic scattering of 50-MeV helium ions from ^{238}U was studied with a high-resolution magnetic spectrometer. We measured angular distributions for members of the ground-state rotational band up to the 8^+ level. Coupled-channels calculations yield 0.022, 0.060, and −0.012 for the respective deformation parameters β_2, β_4, and β_6 for an optical radius of $1.44A^{1/3}$ fm. These values are compared with other experimental results.

The ability to measure details of nuclear shapes by means of α-particle inelastic scattering has been first demonstrated for permanently deformed rare-earth nuclei.[1] Attempts to extend these measurements to the interesting actinide region of permanent deformations have been thwarted by experimental difficulties, mainly the inability of solid-state detectors to resolve the more closely spaced energy levels in these nuclei. Meanwhile, several theoretical predictions for the hexadecapole moments of actinide nuclei and a few experimental results using other techniques have been published. The interest in the problem is intensified, however, because both the theoretical predictions[2-4] and the experimental results[5,6] show large variations for the Y_{40} moment of uranium. These experiments determine rather large values for the deformation