Equilibrium Quadrupole and Hexadecapole Deformations in ^{230}Th and ^{238}U†

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Large contributions to the excitation of 4^+ rotational states from $E4$ transitions have been observed in precision Coulomb-excitation experiments with ^{4}He projectiles for even-even targets in the mass range $230 \leq A \leq 252$. The values of $B(E4, 0 \to 4)$ deduced from an analysis of the Coulomb-excitation probabilities for ^{230}Th and ^{238}U are $(1.10 \pm 0.44)e^2$ b^4 and $(1.26 \pm 0.52)e^2$ b^4, respectively. The deformation parameters β_{40} are 0.110 ± 0.027 and 0.100 ± 0.028 for ^{230}Th and ^{238}U, respectively.

Substantial equilibrium hexadecapole deformations β_4 for nuclei in the mass range $220 \leq A \leq 250$ were first proposed by Fröman[1] in order to explain the relatively small α-decay hindrance factors for the $l = 4$ partial wave. More recent estimates of the magnitude of stable β_4 deformation in this mass region using the level schemes of the Woods-Saxon potential and the influence of β_4 on the properties of the single-particle states of deformed nuclei have been reported by Gareev, Ivanova, and Pashkevich.[2] Nilsson and co-workers[3] have also included hexadecapole deformation in the nuclear shape for these nuclei and have extended their calculations, based on a modified harmonic-oscillator potential, into the region of proposed "superheavy" nuclei. It is quite apparent from these theoretical predictions[1-3] that the presence of β_4 distortions exerts a sizable influence on nuclear properties and must be properly accounted for in calculations which are extrapolated into the region of "superheavy" elements.

From the analysis of Coulomb-excitation experiments it is possible to extract the reduced electric multipole matrix elements of the multipole operators. Recently, Stephens *et al.*[4] reported results from a Coulomb-excitation experiment that measured the reduced hexadecapole moment $\langle 4 \| \mathfrak{M}(E4) \| 0 \rangle$ in ^{152}Sm.

Measurements[5,6] of the inelastic scattering of 27.5- to 50-MeV α particles from rare-earth nuclei have been interpreted to yield quantitative determinations of the hexadecapole deformation in the mass distributions of these nuclei in addition to the quadrupole deformation. However, the only experimental estimate of the magnitude of the hexadecapole deformation in the heavy-mass deformed region has recently been reported by Moss *et al.*[7] These authors have analyzed 23-MeV proton inelastic-scattering data for ^{232}Th and ^{238}U and derive β_4 values of 0.050 ± 0.015 and $0.017^{+0.015}_{-0.030}$, respectively.

We have initiated a systematic Coulomb-excitation program in the heavy-mass deformed region, as recently reported,[8,9] and wish to report in this communication our results for the experimental determination of the reduced electric quadrupole and hexadecapole moments for ^{230}Th and ^{238}U and their interpretation in terms of the equilibrium deformation parameters β_{20} and β_{40}.

We have measured the elastically and inelastically scattered α particles from ~20-μg/cm^2 targets of ^{230}Th and ^{238}U at incident energies of 17 and 18 MeV using the Oak Ridge tandem Van de Graaff in conjunction with an Enge split-pole magnetic spectrograph. Measurements were made at a laboratory angle of 150° for both targets and were supplemented by a 90° measurement for the ^{238}U target. Scattered particles were detected at the focal plane of the spectrograph using a position-sensitive gas proportional counter described by Ford, Stelson, and Bemis.[10] The isotopically pure targets were prepared using a 150-cm radius electromagnetic isotope separator.[11] The energy resolution achieved in these experiments was about 15 keV, which is more than sufficient to resolve the rotational states of the ground-state bands in the heavy-mass deformed region. The experimental scattered α-particle spectrum at 150° for ^{230}Th is shown in Fig. 1.

The excitation probabilities for the 2^+ and 4^+ states were determined relative to the elastic scattering peak from the measured peak areas with an accuracy of 1% for the 2^+ state and 3.6 to 6% for the 4^+ state. These uncertainties include the error in background subtraction and the uncertainty in resolving the 2^+ peak from the elastic peak. The transition probabilities, $B(E2, 0 \to 2)$ and $B(E4, 0 \to 4)$, were extracted from the experimental excitation probabilities for the 2^+ and 4^+ states with the aid of the Winther–de Boer computer program[12] for multiple Coulomb excita-

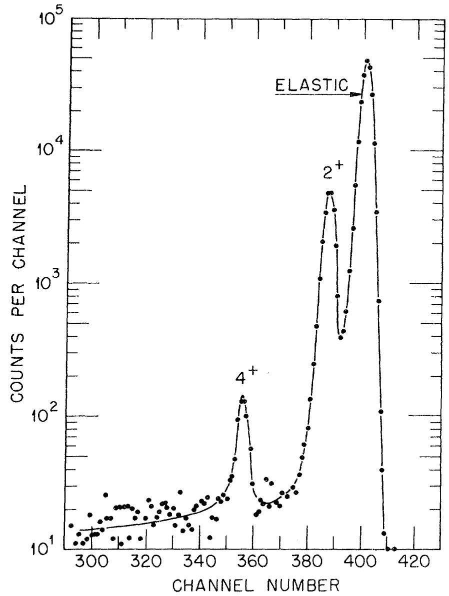

FIG. 1. Spectrum for 17.00-MeV α particles scattered from ^{230}Th (~ 20-μg/cm^2 target) at a laboratory angle of 150°.

tion which includes $E1$, $E2$, $E3$, and $E4$ excitation. The influence of the hexadecapole moments on the Coulomb-excitation cross section for the 4^+ state in ^{230}Th is displayed in Fig. 2 as a function of the reduced $E4$ matrix element $\langle 4\|\mathfrak{M}(E4)\|0\rangle$. These calculations were done in the limit of the rigid-rotor model with states 0^+, 2^+, 4^+, and 6^+, where the five reduced $E2$ matrix elements and the seven reduced $E4$ matrix elements are given by

$$M_{if}(E\lambda) \equiv \langle I_f\|\mathfrak{M}(E\lambda)\|I_i\rangle = (2I_i+1)^{1/2}[(2\lambda+1)/16\pi]^{1/2} \times Q_{\lambda 0}\langle I_i\lambda K0|I_i\lambda I_f K\rangle. \quad (1)$$

The intrinsic quadrupole moment Q_{20} for a prolate shape was obtained from the experimental value[9] for $B(E2,\ 0\to 2)$. The experimental excitation probabilities for the 4^+ state in ^{230}Th and ^{238}U are (22.7 ± 7.4)% and (14.0 ± 4.0)% larger than the values calculated without the inclusion of the hexadecapole moments. We attribute the excess excitation of the 4^+ state to the presence of $E4$ Coulomb excitation. The experimental excitation probabilities for the 2^+ and 4^+ states in ^{230}Th and ^{238}U are listed in Table I together with the deduced values for $B(E2, 0\to 2)$ and $B(E4, 0\to 4)$. These $B(E4, 0\to 4)$ values correspond to a reduced $E4$ matrix element $\langle 4\|\mathfrak{M}(E4)\|0\rangle$ of $(1.05 \pm 0.21)e$ b^2 and $(1.12\pm 0.24)e$ b^2 for ^{230}Th and ^{238}U, respectively. The results were deduced from the analysis with all seven reduced $E4$ matrix elements included in the multiple Coulomb-excitation calculations. The use of reduced $E4$ matrix elements based on the rigid-rotor model seems justified because the $B(E4, 0\to 4)$ values in Table I are very large, being a hundred times $B(E4)_{\text{s.p.}}$. The reduced $E4$ matrix element $\langle 4\|\mathfrak{M}(E4)\|0\rangle$ extracted from the 4^+ excitation probability at 90° for ^{238}U is $1.06e$ b^2 and is consistent with the 150° data although the accuracy of 4^+ excitation at 90° is 11%.

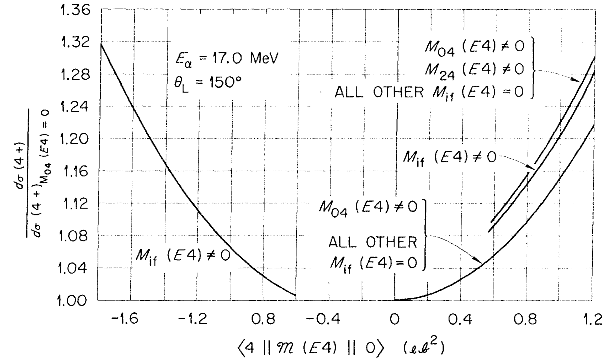

FIG. 2. Ratio of the calculated cross section for excitation of the first 4^+ state in ^{230}Th, including $E4$ excitation, to the cross section without any $E4$ contribution, as a function of $\langle 4\|\mathfrak{M}(E4)\|0\rangle$.

Several other effects which could influence the calculated excitation probabilities of the 2^+ and 4^+ states have been considered in the analysis: (1) quantum mechanical corrections to the excitation probabilities which were obtained from the semiclassical treatment of multiple Coulomb ex-

TABLE I. Experimental excitation probabilities for the 2^+ and 4^+ rotational states of ^{230}Th and ^{238}U together with resultant $B(E2, 0\to 2)$ and $B(E4, 0\to 4)$ values. The excitation probabilities were determined at 150° in the laboratory system.

	E_α (MeV)	$\sigma(2^+)/\sigma(\text{el})$	$\sigma(4^+)/\sigma(\text{el})$	$B(E2, 0\to 2)$ (e^2 b^2)	$B(E4, 0\to 4)$ (e^2 b^4)
^{230}Th	17	0.1054 ± 0.0008	0.002 32 ± 0.000 14	8.01 ± 0.11	1.10 ± 0.44
^{238}U	18	0.1690 ± 0.0017	0.0056 ± 0.0002	11.70 ± 0.15	1.26 ± 0.52

citation; (2) deviation of the reduced $E2$ matrix elements from the rigid-rotor limit; and (3) influence of higher 2^+ states on the excitation probabilities of the 2^+ and 4^+ states in the ground-state rotational band. Alder and co-workers[13] have investigated the quantum mechanical corrections of the Coulomb-excitation process in second-order perturbation theory. We are justified in using these quantum corrections in second-order perturbation theory because ~98% of the excitation of the 4^+ state is pure double $E2$ excitation under the condition $\langle 4 \| \mathfrak{M}(E4) \| 0 \rangle = 0$. The quantal correction reduces the excitation probability for the 4^+ state by the pure double $E2$ process by 3% for ^{230}Th and ^{238}U. This quantal correction is included in the analysis for the results given in Table I. Omitting the quantal correction would decrease our measured value for $\langle 4 \| \mathfrak{M}(E4) \| 0 \rangle$ by 10% for ^{230}Th and by 19% for ^{238}U.

Centrifugal stretching effects could alter the reduced $E2$ matrix elements. The parameter $\alpha \equiv B/A$, where $E(I) = AI(I+1) - BI^2(I+1)^2$, for ^{230}Th is 1.3×10^{-3}.[14] In the extreme point of view, if we attribute this value of α to centrifugal stretching which can be related to the $B(E2,\, I_i \to I_f)$, the measured value for $\langle 4 \| \mathfrak{M}(E4) \| 0 \rangle$ is reduced by 7% for ^{230}Th.

The analysis of the data for ^{238}U was also done including the 2^+ "γ-vibrational-like" state at 1060.2 keV, which is the most strongly excited of the five higher 2^+ states observed[15] in ^{238}U. The measured value for $\langle 4 \| \mathfrak{M}(E4) \| 0 \rangle$ is increased by only 1% for ^{238}U with the inclusion of the higher 2^+ state.

The errors for the measured reduced $E4$ matrix element $\langle 4 \| \mathfrak{M}(E4) \| 0 \rangle$ from an uncertainty in beam energy and scattering angle are negligible. For example, uncertainties of ± 50 keV in the beam energy and ± 1° in scattering angle introduce errors of ± 0.7% in $\langle 4 \| \mathfrak{M}(E4) \| 0 \rangle$.

The measured transition probabilities are related to the intrinsic multipole moments by the relations

$$B(E2,\, 0 \to 2) = (5e^2/16\pi) Q_{20}^2, \tag{2}$$

$$B(E4,\, 0 \to 4) = (9e^2/16\pi) Q_{40}^2. \tag{3}$$

For a homogeneous charge distribution of shape $R = R_0(1 + \beta_{20} Y_{20} + \beta_{40} Y_{40})$, the intrinsic multipole moments are related to the model-dependent deformation parameters β_{20} and β_{40} by the following expressions[16]:

$$Q_{20} = \frac{3ZR_0^2}{(5\pi)^{1/2}} \beta_{20} \times \left(1 + 0.360\beta_{20} + 0.967\beta_{40} + 0.328\frac{\beta_{40}^2}{\beta_{20}}\right), \tag{4}$$

$$Q_{40} = \frac{ZR_0^4}{(\pi)^{1/2}} \beta_{40} \times \left(1 + 0.983\beta_{20} + 0.725\frac{\beta_{20}^2}{\beta_{40}} + 0.411\beta_{40}\right). \tag{5}$$

Using a charge radius parameter $R_0 = 1.2A^{1/3}$ fm and restricting β_{20} to positive values (prolate shape), we have solved the above relationships numerically to yield values for β_{20} and β_{40} which are given in Table II (labeled "uniform"). The deformation parameters for a diffuse-edge charge distribution with axial symmetry are also given in Table II (labeled "diffuse edge"). The radial integrals in the expression for the multipole moments under this assumption have been evaluated numerically[16] for $r_0 = 1.1$ fm and $a = 0.6$ fm, which are best fits for these parameters from electron scattering. If only the first-order terms of $\beta_{\lambda 0}$ were retained in $Q_{\lambda 0}$, the values for $\beta_{\lambda 0}$ deduced from our measured $E\lambda$ moments would be increased by 20% for β_{20} and about 60% for β_{40}.

The excitation probabilities could also be analyzed on the basis of a negative value for $\langle 4 \| \times \mathfrak{M}(E4) \| 0 \rangle$ (see Fig. 2). In the case of ^{230}Th the extracted value for $\langle 4 \| \mathfrak{M}(E4) \| 0 \rangle$ is $-(1.57 \pm 0.21) e$ b^2

TABLE II. A comparison of the theoretical and experimental equilibrium deformation parameters for ^{230}Th and ^{238}U. The theoretical results are those of Nilsson *et al.* (Ref. 3) and of Gareev, Ivanova, and Pashkevich (Ref. 2).

	β_{20}				β_{40}			
	(expt)				(expt)			
	Uniform	Diffuse edge	(Nilsson)[a]	(Gareev)	Uniform	Diffuse edge	(Nilsson)[a]	(Gareev)
^{230}Th	0.204 ± 0.006	0.226 ± 0.006	0.191	0.16	0.110 ± 0.027	0.118 ± 0.030	0.088	0.07
^{238}U	0.235 ± 0.006	0.261 ± 0.007	0.221	0.20	0.100 ± 0.028	0.106 ± 0.031	0.066	0.075

[a]The relationship between ϵ, ϵ_4, and β_{20}, β_{40} was calculated by numerical integration from a formalism given by Nix (Ref. 17).

and $B(E4, 0\rightarrow 4) = 220B(E4)_{\text{s.p.}}$. The model-dependent deformation parameters for a uniform charge distribution are $\beta_{20} = 0.264 \pm 0.003$ and $\beta_{40} = -0.261 \pm 0.032$. This solution seems rather unlikely because the experimental intensities from the α decay of the even-A nuclei of Th and U are explained[1,18] by introducing a positive β_{40}. Also the calculations[2,3] of the sum of single-particle energies as a function of β_{20} and β_{40} indicate that the equilibrium value of β_{40} is positive for these nuclei.

The equilibrium deformation parameters β_{20} and β_{40} from the calculations of Gareev, Ivanova, and Pashkevich[2] and of Nilsson *et al.*[3] are also listed in Table II. Our values for β_{40} are somewhat larger than either of the theoretical predictions. Also, the model-dependent deformation parameters β_{40} deduced from the Coulomb-excitation reaction are considerably larger than the values for the mass distribution inferred from the (p,p') reaction.[7]

We have demonstrated that electric quadrupole and hexadecapole moments in the actinide deformed region may be reliably extracted from precision Coulomb-excitation experiments using ^{4}He-ion projectiles. The measured moments may be used to deduce model-dependent deformation parameters β_{20} and β_{40} which may serve as more reliable input data for the calculation of the stability of superheavy elements. We are currently pursuing the determination of hexadecapole moments for other thorium and uranium isotopes as well as the plutonium and curium isotopes.

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