Sign of the $E4$ moment in ^{236}U by multiple Coulomb excitation with ^{40}Ar ions*

M. W. Guidry,† E. Eichler, N. R. Johnson, G. D. O'Kelley,‡ R. J. Sturm,§ and R. O. Sayer
Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830*
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Multiple Coulomb excitation with ^{40}Ar projectiles has been used to excite the ground state rotational band of ^{236}U through the 12^+ state. A comparison of the experimental and theoretical excitation probabilities confirms a positive value for the $E4$ moment, removing the sign ambiguity inherent in the determination of this moment by α-particle Coulomb excitation.

[NUCLEAR REACTIONS Multiple Coulomb excitation, $^{40}\mathrm{Ar}+^{236}\mathrm{U}$, $E_{\mathrm{lab}}=145.8$, 152.4 MeV; measured excitation probabilities, compared to theoretical predictions; deduced sign of $E4$ moment.]

I. INTRODUCTION

Theoretical calculations which minimize the nuclear energy with respect to the deformation parameters β_k have been used to predict significant β_4 deformation in the rare-earth and actinide regions.[1-8] The presence of such deformations, suggested initially by α-decay properties,[9,10] was firmly established in the rare-earth region by Hendrie *et al.*[11] and Aponick *et al.*,[12] using α-particle scattering at energies above the Coulomb barrier. Recent α-particle experiments at energies below the Coulomb barrier have demonstrated the presence of significant hexadecapole deformation of the nuclear charge for a number of nuclei.[13-20]

McGowan *et al.*[18] and Bemis *et al.*[19] first used the α-scattering (Coulomb excitation) technique in the heavy-element region where large β_4 deformations for many nuclei were found. These results have been corroborated for ^{232}Th in another recent Coulomb excitation experiment,[20] though they are in some disagreement with the higher-order deformations inferred from fitting perturbed band structure[21] in ^{182}W, and from muonic x-ray work on ^{232}Th, ^{238}U, and ^{182}W.[22] Inelastic scattering of α particles[23] and protons[24] from actinide targets at energies above the Coulomb barrier has also been analyzed in terms of higher-order deformations of the optical potential. Though the results disagree considerably with each other in magnitude, both find measurable Y_{40} components in the shape of the nuclear potential.

There are points of disagreement concerning the magnitude of β_4 deformations; and, indeed, it is not yet even clear what the relation among deformations in the Coulomb, nuclear, and optical potentials as measured by the above-mentioned techniques should be. Nevertheless, the presence of measurable hexadecapole deformation is an empirical fact for a large class of nuclei, with attendant consequences for calculations of deformed nuclear structure, fission barriers, electromagnetic transition probabilities, etc. It is therefore of import to know both the magnitude and sign of the $E4$ deformation.

In the α-particle Coulomb excitation method the yield for 4^+ excitation in a ground-state rotational band in excess of that predicted by multiple-$E2$ excitation is related quadratically to a reduced matrix element $\langle 4^+ \| M(E4) \| 0^+\rangle$. Unfortunately, the quadratic relationship excludes a unique solution, and in ^{236}U values of +1.13 and -1.99 for $\langle 4^+ \| M(E4) \| 0^+\rangle$ are consistent with the observed excitation probability of the 4^+ state.[19] These two solutions correspond to positive and negative values, respectively, for the model-dependent deformation parameter β_4. Theoretical calculations[1] predict a positive β_4 deformation as is consistent with α-decay properties in this region.[9] However, it is desirable to determine experimentally the sign of the hexadecapole moment in actinide nuclei.

If $E4$ contributions to excitation are sizable, appreciable differences in calculated excitation probabilities (using matrix elements based on the two different roots for $\langle 4^+ \| M(E4) \| 0^+\rangle$) are expected for high-spin states which are accessible only by higher-order processes. Indeed, such a dependence has been found in ^{236}U for the ground-band excitation probabilities P_8, P_{10}, and P_{12}, and comparison of calculated probabilities with experimental ones confirms a positive sign for the $E4$ moment.

Analogous results have been reported previously for ^{232}Th and ^{238}U by Eichler *et al.*,[25] and a similar method has been proposed by Winkler.[26] A dependence of the Coulomb-nuclear interference effect on the sign of the $E4$ moment has also been exploited to determine the sign of the hexadecapole moment for several rare-earth nuclei,[27] while the scattering of protons[23] and α particles[24] above the Coulomb barrier also indicates a positive β_4

deformation of the (optical) potential for ^{232}Th and ^{238}U.

II. EXPERIMENTAL METHOD

Multiple-Coulomb excitation of ^{236}U was performed using 146-MeV and 152-MeV $^{40}Ar^{8+}$ beams from the Oak Ridge Isochronous Cyclotron (ORIC). The target was a thick, metallic uranium foil of 89% isotopic enrichment in mass 236, mounted at 45° relative to the beam axis. A coaxial Ge(Li) detector was placed at 90° relative to the beam axis to minimize absorption of γ rays in the target material and to avoid any tailing due to the Doppler effect for recoil ions slowing in the target. For the experiment utilizing 146-MeV ^{40}Ar ions, the detector had an efficiency of 6.5% for the ^{60}Co 1333-keV transition while the experiment at 152-MeV utilized a detector with 18% efficiency. The Ge(Li) detector was operated in coincidence with a 100-μm deep, 300 mm^2 annular silicon detector which was shielded by 1.6 mg/cm^2 of gold foil to reduce the detection of secondary electrons. The annular detector spanned a backscattering angular range of 164° to 176° with an average angle of 168°, thereby selecting those events involving backward scattering which favor the multiple excitation process.

The linear signals from the Ge(Li) detector, the silicon detector, and the time relation between them from a time-to-amplitude converter (TAC) were subjected to slow-coincidence requirements, digitized by a fast analog-to-digital converter (ADC) system, and placed into temporary buffer storage on an SEL-840A computer as the several parameters of a three-parameter data word. The data were subsequently transferred to disc storage and finally to magnetic tape. Following the experiment the data were scanned with appropriate windows on the TAC and heavy-ion spectra to yield γ-ray spectra from which random events were subtracted, giving spectra in final form for analysis.

Peak areas were extracted by a manual analysis and also by a Gaussian least-squares fitting routine, but the areas obtained via the computer code have been used exclusively in the analysis. In no case did the intensities obtained by the two procedures differ by more than 5%.

Because the target was of sufficient thickness to stop the beam completely, the effective thickness for Coulomb excitation was determined by a pulse-height discriminator set on the heavy-ion spectrum. In order to calibrate the backscatter spectrum, a short bombardment was made on a ^{162}Dy target in addition to the primary one on ^{236}U. From the upper cutoffs of the ^{162}Dy and ^{236}U backscatter spectra (corresponding to scattering from the face of the target), the average backscatter angle, and the kinematic backscatter factor,[28] the heavy-ion spectrum was calibrated and the lower cut-off determined to correspond to minimum incident projectile energies of 129 and 107 MeV for the 152-MeV and 146-MeV runs, respectively. The absolute efficiencies of the Ge(Li) detectors were determined by calibration with a standard ^{226}Ra source and a ^{182}Ta source.

In experiments of this type care must be taken to maintain a distance of closest approach which insures a purely electromagnetic interaction. In a classical calculation, assuming a radius parameter of $r_0 = 1.2$ fm and spherical nuclei with sharp density cutoffs, the highest energy beam used corresponds to a closest-approach distance of 6.9 fm between nuclear surfaces. Although α-particle Coulomb excitation experiments suggest the onset of interference at distances as large as 8 fm between nuclear surfaces,[18(b)] other recent data[29] indicate that for heavy-ion projectiles the critical region lies much closer to the sharp cutoff radius of the target nucleus. Furthermore, the small (1–2%) effects noted at the larger separations in the α-particle experiments[18(b)] lie well within the scope of our experimental uncertainties. We conclude that a separation of 6.9 fm between spherical nuclear surfaces is adequate to exclude significant Coulomb-nuclear interference in the present work.

These crude considerations do not account properly for any state dependence of the interference, as no published experimental data for such behavior at higher spins ($I > 4$) is available. Neither are effects due to static or dynamical deformations adequately considered in the above arguments. The dynamical deformations are probably small and should, at any rate, raise the barrier. Theoretical consideration is presently being given to the nature of the Coulomb-nuclear interference for high-spin states excited by heavy-ion reactions.[30] These calculations indicate a deformation and state dependence of the interference, but suggest that at the energies considered in this paper deviations from pure Coulomb excitation will be negligible.

III. EXPERIMENTAL RESULTS

In each of the experiments the $E2$ transitions in the ground-state rotational band were easily detected through spin 12^+ ($E_{lab} = 152$ MeV) or 10^+ ($E_{lab} = 146$ MeV). A typical spectrum of the γ-ray transitions in the ground band is shown in Fig. 1. The peak areas were used to compute an experimental excitation probability for the transitions

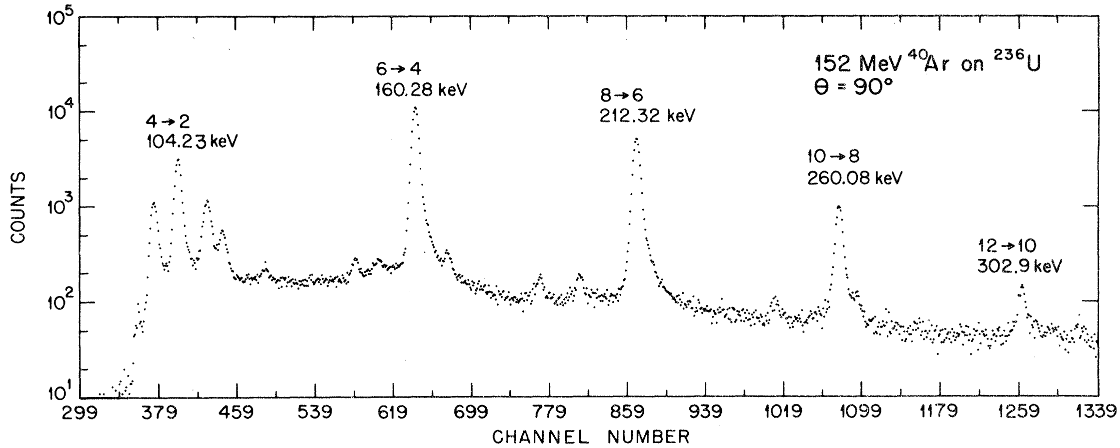

FIG. 1. Spectrum of the ^{236}U ground-state rotational band populated by multiple Coulomb excitation. The spectrum was taken on a thick target with a Ge(Li) detector at 90° to the beam axis, operated in coincidence with backscattered projectiles.

observed from the relation

$$(P_J)_{\rm exp}=\frac{1}{N_B a}\left[\frac{(1+\alpha_T)N}{W(\theta)T_\gamma\epsilon}-T_c\right] , \qquad (1)$$

where N_B is the number of valid heavy ions (the detected backscatter particles which generate an "event" logic pulse); a is the fractional isotopic abundance; α_T is the total internal conversion coefficient; N is the area of the full-energy peak, corrected for summing; $W(\theta)$ is an angular distribution factor for the γ rays; T_γ corrects for γ-ray absorption in the target; ϵ is the efficiency of the γ-ray detector; and T_c subtracts the cascade feeding from higher states. The internal conversion coefficients were taken from the compilation of Hager and Seltzer.[31] Previous experimental work on ^{232}Th and ^{238}U indicates that the angular distribution is described well by Coulomb excitation theory,[32] with the calculated and measured values of $W(\theta)$ agreeing to better than 5%.[25] The calculated values of the statistical tensors have therefore been used to compute the angular distribution function $W(\theta)$, with finite solid-angle corrections to the A_2 and A_4 coefficients based on the estimates of Camp and Van Lehn.[33]

The theoretical transition probabilities corresponding to those determined experimentally were computed using a form of the semiclassical Winther-deBoer coupled-channels code,[32] modified by Holm[34] to accept $E3$ and $E4$ matrix elements, and by Sayer[35] to accommodate thick targets. Therefore, the theoretical excitation probability of a state J is given by[35]

$$(P_J)_{\rm theor}=\frac{\int_{E_i}^{E_{\rm cut}(J)}\frac{dE}{S(E)}\int d\Omega_L\frac{d\sigma_J(\theta_L,E)}{d\Omega_L}}{\int_{E_i}^{E_{\rm cut}(0)}\frac{dE}{S(E)}\int d\Omega_L\frac{d\sigma_R(\theta_L,E)}{d\Omega_L}} , \qquad (2)$$

where $S(E)$ is the stopping power for the projectile in the thick target, $d\Omega_L$ is an element of solid angle in the laboratory system, $d\sigma_J(\theta_L,E)/d\Omega_L$ is the laboratory differential cross section for excitation of the state J, $d\sigma_R(\theta_L,E)/d\Omega_L$ is the Rutherford differential cross section in the laboratory reference frame, E_i is the maximum incident projectile energy, and $E_{\rm cut}(J)$ is the energy of the lowest-energy particle which contributes to the experimental excitation probability of Eq. (2). The quantity $E_{\rm cut}(J)$ is computed from the kinematics of the scattering process, as is the Rutherford cross section, by the program PARTSPEK.[28] The stopping powers are interpolated from the values of Northcliffe and Schilling,[36] and the differential cross section for Coulomb excitation results from numerical solution of the coupled differential equations defining the excitation-probability amplitudes on the nuclear eigenstates.[32]

Since considerable attention has recently been focused on the whole question of stopping power validity, some attention must be given to the effect of any stopping power uncertainties on our calculations. The stopping powers $S(E)$ enter Eq. (2) in two ways: (1) explicitly in the denominator of the integrands, and (2) implicitly in the values of the limit $E_{\rm cut}$. Bearing in mind that we are primarily interested in the excitation of high-spin states in this paper, the following points are salient.

For the excitation of a high-spin state J, only a small range of energy values lying near the maximum energy E_i contributes appreciably to the excitation probability P_J. This is because, in a classical sense, the $r^{-\lambda-1}$ dependence of the λ-pole potential implies that close-approach trajectories impart maximum torque (transfer of "spin quanta") to the deformed target nucleus. (More rigorously, the multipole coupling parameter $\chi^{(\lambda)}$ describing the strength of the interaction goes as $a^{-\lambda}$, where $2a$ is the classical distance of closest approach). Then for the higher-spin states there is an effective energy limit on the integration lying above $E_{\rm cut}(J)$, and P_J for these states is insensitive to the value of $E_{\rm cut}(J)$, and hence to the stopping powers implicit therein.

Because our conclusions ultimately depend only on normalized probabilities (i.e., on ratios), we are interested only in the shape of $S(E)$, not its magnitude, in the integrand of Eq. (2). For the excitation of the 8^+, 10^+, and 12^+ states, on which the strength of our conclusion rests, the smooth variation of the Northcliffe-Schilling stopping powers over the effective energy for excitation is < 5%. It therefore follows from the above considerations that the conclusions in this paper are quite insensitive to the stopping powers used, provided the variation of $S(E)$ with energy is not radically different from the Northcliffe-Schilling prediction.

A. Rotational limit

If the nucleus is a good rotor the matrix elements for a transition $i\to f$ in the ground band are given by

$$\langle I_i \| M(E\lambda) \| I_f\rangle = (-1)^{I_f - I_i}(2I_i+1)^{1/2} \times \langle I_i\,\lambda 0 0 | I_f 0\rangle \langle 0 \| M(E\lambda) \| 2\rangle\,, \qquad (3)$$

where $\langle I_i\,\lambda 00 | I_f 0\rangle$ is a Clebsch-Gordan coefficient. Equation (3) and the values of $M(E\lambda, 0\to\lambda)$ from Bemis, *et al.*[19] have been used to compute the theoretical Coulomb excitation probabilities for three separate cases: (a) $M(E2)$ matrix elements only, (b) $M(E2)$ elements and $M(E4)$ elements based on the positive-root $M(E4,\ 0^+\to 4^+)$, and (c) $M(E2)$ elements and $M(E4)$ elements based on the negative-root $M(E4,\ 0^+\to 4^+)$ of Ref. 19. In all calculations states in the ground band with spins 0–14 were included.

These calculations and their comparison with the experimental excitation probabilities are shown in Table I. The numbers in parentheses represent the absolute value (not normalized) of the experimental/theory ratio for the 146-MeV data, while those numbers not in parentheses are the 152-MeV data, normalized at the 6^+ level. The discrepancy in absolute value between the two sets of data is thought to be due to a malfunction of the scaler for the number of heavy ions in the 152-MeV experiment. Since the conclusions in this paper require only a comparison of relative excitation probabilities, the normalized probabilities of Table I are sufficient. The experimental excitation probabilities are in much better agreement with theory when the positive-root $M(E4)$ matrix elements are used in the calculation than when the negative-root solutions are employed. Before these results can be legitimately interpreted as confirmation of a positive sign for the $E4$ moment, several effects that are not included in the values of Table I must be considered: (1) virtual excitation of other levels, (2) quantum-mechanical corrections to the semiclassical calculations, (3) possible deviations of the matrix elements from rotational predictions, and (4) the inclusion of higher-order moments ($\lambda > 4$) in the excitation process.

B. Inclusion of vibrational matrix elements

In addition to the multipole coupling between ground-band members, there may exist other states having favorable energies and symmetries which are connected to ground-band states through nonnegligible matrix elements. In particular, bands in even-even deformed nuclei built on β, γ, and octupole vibrational modes may be significantly coupled to states of the ground band by the $E2$ or $E3$ component of the Coulomb field. These states then provide virtual channels through which ground-band excitation may proceed, and the corresponding matrix elements must be included in the calculation. To determine the

TABLE I. Ratios of experimental/theoretical Coulomb excitation probabilities for members of the ground-state rotational band in ^{236}U. The numbers in parentheses are absolute values (not normalized) from the 146-MeV experiment, while those not in parentheses are from the 152-MeV experiment, normalized as indicated. All levels in the ground-state band through spin 14 were included in the calculations.

Level	$M(E2)$ only	$M(E_4)$ (+)	$M(E4)$ (−)
4^+	(0.97 ± 0.05)	(1.02 ± 0.06)	(0.91 ± 0.05)
6^+	1.00 ± 0.05 [a]	1.00 ± 0.05 [b]	1.00 ± 0.05 [c]
	(1.04 ± 0.06)	(1.05 ± 0.06)	(1.06 ± 0.06)
8^+	1.16 ± 0.05 [a]	1.07 ± 0.05 [b]	1.31 ± 0.07 [c]
	(1.07 ± 0.07)	(1.01 ± 0.07)	(1.19 ± 0.08)
10^+	1.13 ± 0.08 [a]	0.95 ± 0.07 [b]	1.32 ± 0.10 [c]
	(1.12 ± 0.11)	(0.96 ± 0.10)	(1.29 ± 0.13)
12^+	1.14 ± 0.18 [a]	0.83 ± 0.13 [b]	1.29 ± 0.20 [c]
$\langle 2^+ \| M(E2) \| 0^+\rangle$ =	−3.40	−3.40	−3.36
$\langle 4^+ \| M(E4) \| 0^+\rangle$ =	0.00	1.23	−1.99

[a] Ratios are from 152-MeV data normalized to the 6^+ level $(P_{\rm exp}/P_{\rm theor})_{6^+} = 0.74$.

[b] Ratios are from 152-MeV data normalized to the 6^+ level $(P_{\rm exp}/P_{\rm theor})_{6^+} = 0.80$.

[c] Ratios are from 152-MeV data normalized to the 6^+ level $(P_{\rm exp}/P_{\rm theor})_{6^+} = 0.78$.

magnitude of this effect, vibrational matrix elements derived from the work of McGowan *et al.*[37] were included in a 20-state version of the Coulomb-excitation computer program, with other requisite information on vibrational states in ^{236}U taken from Refs. 37–40. In related work on ^{238}U, Eichler *et al.*[25] found that inclusion of the β band in the calculations had only a small influence on the ground-state probabilities. As transitions from the ground to the β band are not observed for ^{236}U in α-particle Coulomb excitation,[37] it was concluded that the β band has negligible effect on the ^{236}U ground-band probabilities. Therefore, two cases were calculated for the ^{236}U problem: the ground band with (a) the complete γ band through the 13^+ state, and (b) the complete $K=0$ octupole band to the 15^- state. The appropriate matrix elements were derived from the rotational model [Eq. (3)] and the ground-to-vibrational matrix elements of McGowan *et al.*[37] The percentage change induced in the excitation probability for various ground-band states is shown in Table II, along with an estimated correction for the inclusion of all vibrational states connected to the ground band. The adopted corrections are based on the effect of the two cases considered and previous experience with other combinations of vibrational states in ^{238}U.[25] The corrections are seen to range from -3% at the 4^+ level to $+13\%$ at the 12^+ state.

C. Quantum-mechanical effects

The semiclassical formalism of Winther and deBoer[32] treats projectile motion as the classical dynamics of a point charge scattered in a spherically symmetric Coulomb field. This approximation is a good one as long as two general conditions are satisfied: (1) the distance of closest approach $2a$ should be much larger than the deBroglie wavelength $\lambda\!\!\!^{-}$ for the projectile, in which case the wave packet describing the projectile is approximately spatially confined to a classical trajectory, and (2) dynamical distortion of the classical orbit during the scattering process must be negligible.

The wavelength condition is usually specified by the inequality

$$\eta \equiv a/\lambda\!\!\!^{-} \gg 1. \tag{4}$$

This condition is fulfilled rather well for heavy-ion Coulomb excitation and quantal corrections are small for first-order processes. However, the quantal corrections introduce large changes in matrix element phases which can have considerable effect when multiple processes (second-order and higher) are important.[41,42] Therefore, even for very heavy ions, quantal effects become important for the excitation of high spin states proceeding by multi-step processes. For the typical situations discussed in this paper, the majority of any dynamical effects on the classical trajectory are accounted for by the use of energy-symmetrized hyperbolas in the Winther-de Boer program.[30]

Attempts at treating the quantal corrections have involved perturbation theory,[41,42] various symmetrizations of the orbit parameters,[32,43] coupled-channel integration of the exact quantum-mechanical equations of motion,[44] or a recent adaptation of a classical limit S-matrix approach.[45,46,30] The exact coupled channels approach requires excessive computer time for large values of η, while the S-matrix formalism and sophisticated orbital symmetrizations are still in a developmental stage. Therefore, we have used coupled-channel calculations[47] for low η, extrapolated by $1/\eta$ to large values of η, to estimate the quantal corrections. The adopted quantal corrections to the theoretical excitation probabilities range from 0% for the 4^+ to -10% for the 12^+ state and are displayed in Table III. It must be strongly emphasized that the validity

TABLE II. The effect on P_{theor} of including vibrational states in the calculation.

State	γ band $I^\pi=2^+-13^+$ (%)	$K=0$ octupole $I^\pi=1^--15^-$ (%)	Adopted correction (%)
4^+	−2.5	−0.3	−3
6^+	−0.7	−2.4	−3
8^+	+1.0	−1.8	−1
10^+	+2.7	+1.8	+5
12^+	+4.9	+8	+13

TABLE III. The adopted quantal corrections to the semiclassical value of P_{theor}.

State	Quantal correction to P_{theor} (%)
4^+	0
6^+	0
8^+	−4 [a]
10^+	−7 [a]
12^+	−10 [b]

[a] Extrapolated by $1/\eta$ from calculations at lower values of η using a coupled-channels quantal code (Ref. 47).

[b] Estimated from trends for lower-spin states.

of the $1/\eta$ extrapolation for high spins is not unequivocally established at present. For this reason large uncertainties in the quantal corrections for P_{10} and P_{12} have been included in the uncertainties quoted in this paper. Fortunately, the vibrational and quantal corrections act in opposite directions on the theoretical probabilities for high spins, and one hopes for some cancellation in these corrections. However, a better understanding of these corrections is mandatory if the interpretation of Coulomb excitation probabilities for high-spin states is to yield precise quantitative information.

D. Deviations from the rotational model

The ground-band matrix elements used in this calculation have been derived from the experimental $B(E2, 0^+ \to 2^+)$ and $B(E4, 0^+ \to 4^+)$ values[19] using the predictions of the rotational model [Eq. (3)]. The even-even actinides are known to show small deviations from $I(I+1)$ energy spacing in the ground-state rotational band, with the first few levels being reasonably well-described by an expansion in powers of $I(I+1)$:

$$E(I) = AI(I+1) - BI^2(I+1)^2 . \qquad (5)$$

The quantity B/A measures the deviation from rotational behavior and has a value of 0.58×10^{-3} for this nucleus.[48] If this deviation from rotational energy spacing is attributed to centrifugal stretching, the $E2$ matrix elements are given by the formulas of Symons and Douglas.[49] For B/A ratios of this magnitude the corresponding increase in the calculated excitation probabilities is as large as 10% and 20% for the 10^+ and 12^+ states, respectively.[25]

In a separate publication lifetime measurements by the recoil-distance method in the ^{236}U ground-state band for spins 4^+ through 12^+ are described.[50] From these lifetimes the $B(E2)$ values and the transition quadrupole moments $Q_{20}(I_i \to I_f)$ were extracted. The constancy within experimental error of these moments indicates that ^{236}U is a good rotor for those states considered. This suggests that the use of the rotational model to compute the ground-band matrix elements in ^{236}U is a good approximation. The further assumption that the $E4$ matrix elements also exhibit rotational behavior seems a reasonable one, but it remains an assumption, as no experimental test of the behavior of the $E4$ moments at higher angular momentum is yet available.

E. Inclusion of higher-order deformation

The theoretical calculations described in the previous sections have included contributions to the excitation probability from $E2$ and $E4$ matrix elements only. One should consider the possible contributions of $E6$ and higher-order moments to the transition probability. Potential deformation parameters for ^{238}U have been determined from (α, α') work above the Coulomb barrier.[23] These data indicate that the magnitudes of the β_2, β_4, and β_6 parameters for that nucleus are in the approximate ratios 20 : 6 : 1. Assuming that the potential deformation parameters of Hendrie *et al.*[23] for ^{238}U are a reasonable indication of the charge deformation parameters in ^{236}U, we conclude that β_6 deformations are small compared to β_4 and β_2 deformations. This, in conjunction with the $r^{-\lambda-1}$ dependence of the $E\lambda$ potential, suggests that the omission of $E6$ moments does not introduce appreciable uncertainty into the calculations.

F. Corrected ratios

The ratios $P_{\rm exp}/P_{\rm theor}$, corrected for quantum-mechanical effects and for the inclusion of virtual excitation through vibrational states, are displayed in Fig. 2. The ratios are much closer to unity for the positive-root $M(E4)$ solutions than for the negative-root values. The differences between the two sets of points is somewhat obscured by the normalization at the 6^+ state and it is even more instructive to compare the normalized χ^2 for deviation of the two sets of ratios from unity: $\chi^2 = 0.17$ for the positive-root set; $\chi^2 = 2.61$ for the negative-root set. These results support the choice of the positive-root solution for the $E4$ moment in ^{236}U.

The error bars in Fig. 2 represent a folding of both experimental and theoretical uncertainties

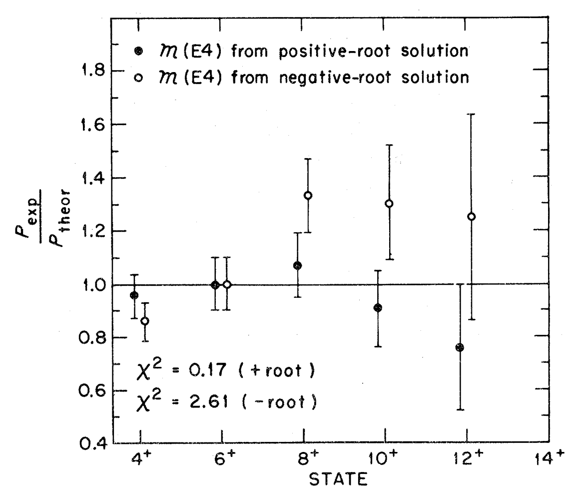

FIG. 2. The normalized ratios $P_{\rm exp}/P_{\rm theor}$ including estimated corrections for all effects discussed in text. The error bars represent absolute uncertainties.

in P_{exp}/P_{theor}. Major contributions to the uncertainty in the relative experimental probabilities include those in total conversion coefficients (~3%), relative efficiencies (2–3%), and feeding intensities (5–10%). The uncertainties in P_{theor} arise from estimated uncertainties in the effect of vibrational states (2–10%), quantal corrections (2–10%), and deviations from rotational $B(E2)$ values lying within the experimental uncertainties of the recoil-distance data (3–25%).[50] They do not reflect uncertainties in the assumed rotational behavior of the $E4$ moments or neglect of the $E6$ moments. Total errors in P_{exp} range from 7% to 9%, while those in P_{theor} vary from 4% to 30%. It should be noted that the very large uncertainties in P_{12} are due primarily to large uncertainty in P_{theor}. This is a consequence of uncertainties in the large quantal and vibrational corrections for this state, and of the lack of sufficiently precise information on the behavior of the 12^+ excitation probability relative to the predictions of the rotational model.

IV. CONCLUSIONS

The critical dependence of the excitation probabilities on the sign chosen for the hexadecapole moment has been demonstrated for high-spin states in ^{236}U. Probabilities calculated using the positive-root value of Bemis *et al.*[19] agree well with the experimental excitation probabilities, whereas those calculated using the negative-root solution are in much poorer agreement with the experimental results. This supports the assignment of a positive $E4$ moment to ^{236}U, a conclusion which is predicted by theory[1] and is consistent with α-decay properties.[9] It is also in agreement with previous results on ^{232}Th and ^{238}U obtained by a similar method,[25] and with inelastic scattering from the optical potential in the actinide region.[23]

As has been pointed out, the greatest uncertainty in this type of experiment is associated with the calculation of P_{theor}. Because the roots for the $E4$ moment in ^{236}U are large, the effect of this moment on higher-spin excitation is of sufficient magnitude to allow a clear choice for its sign, despite the uncertainties currently inherent in P_{theor}. For nuclei where the magnitude of the hexadecapole moment is not as large this may no longer be true, and a more precise calculation of P_{theor} would be necessary. An improved calculation of P_{theor} depends primarily upon (1) a more rigorous treatment of quantal effects for higher-order processes, (2) a more thorough understanding of the contribution of various virtual excitation channels to ground-band excitation, and (3) more precise life-time measurements for high-spin states to determine the nature of the $E2$ matrix elements connecting ground-band levels. These advances are necessary not only for the particular applications of Coulomb excitation discussed in this paper, but for any detailed understanding of the interaction of heavy ions with nuclei at energies near or below the Coulomb barrier.

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† Oak Ridge Graduate Fellow from the University of Tennessee under appointment with Oak Ridge Associated Universities. Present address: Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720.

‡ Dual appointment: Oak Ridge National Laboratory and University of Tennessee.

§ Max Kade Foundation Fellow. Permanent address: University of Marburg, Marburg, Germany.

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