Static Quadrupole Moments of the First 2⁺ States of the Even-Even Osmium Nuclei^{*}

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Measurements of the total thick-target yields of γ rays deexciting the first 2⁺ excited states of Os^{192, 190, 188, 186, 184} following Coulomb excitation by helium, oxygen, and sulfur ions have been made. These measurements were compared with theoretically calculated yields in order to extract linear combinations of the static electric quadrupole moments of the 2⁺ states. The results were compared with the measurements of Pryor and Saladin of the absolute moments of Os^{192, 190, 188} in order to obtain the best combined values for these three isotopes, as well as absolute values for Os^{186, 184}. These values are -0.50 ± 0.20 , -0.95 ± 0.19 , -1.32 ± 0.43 , -1.47 ± 0.54 , and -2.4 ± 1.1 , respectively. For the isotopes Os^{192, 190, 188, 186} the $B(E2, 0^+ \rightarrow 2^+)$ values determined in the experiment agree with published values to within 10%. The $B(E2, 0^+ \rightarrow 2^+)$ value of Os¹⁸⁴ was previously unknown and was determined to be $3.20 \pm 0.62 \ e^2 b^2$. The results are also compared with the model calculations of Kumar and Baranger and found to be in good agreement.

INTRODUCTION

The isotopes of Os and Pt cover a particularly interesting region within the Periodic Table. The Os isotopes span an interval which extends from the highly deformed, rotational Os¹⁸⁴ nucleus to the nearly spherical Os¹⁹² nucleus. This transition is gradual in contrast to the low-mass end of the deformed rare-earth region, where the transition is much sharper. The theory of Kumar and Baranger¹ predicts and experiments by Glenn, Pryor, and Saladin² and Pryor and Saladin³ verified that a fairly smooth change in shape occurs from prolate ellipsoidal (intrinsic quadrupole moment $Q_0 > 0$) to oblate ellipsoidal ($Q_0 < 0$) as one goes from the light Os isotopes to the heavy Pt isotopes.

The purpose of this paper is two-fold: First to provide an independent determination of the relative quadrupole moments of $Os^{188, 190, 192}$ and, second, to extend the measurements to the isotopes $Os^{184, 186}$ which, because of the small isotopic enrichments available, were not amenable to the inelastic-scattering-type experiments described in Ref. 3.

METHOD

The experimental method consisted in a very precise determination of the relative $2^+ \rightarrow 0^+ \gamma$ ray yields from all isotopes present in the target. Measurements of this type have previously been described by several authors.⁴⁻⁷ For completeness we define and summarize in the following some necessary quantities and equations. It is convenient to use the convention that all theoretically calculated or assumed quantities be labeled with small letters, while experimentally given or determined quantities are designated by capital letters. Mixed quantities (ratios of experimental and theoretical values, etc.) will be designated by Greek symbols.

The γ -ray yield at an angle θ_{γ} to the incident beam can be calculated from

$$y(\theta_{\gamma}) = 2\pi \int_{0}^{E_{B}} \int_{0}^{\pi} \frac{1}{S(E)} \frac{d^{2}\sigma}{d\Omega_{p} d\Omega_{\gamma}} \sin\theta_{p} d\theta_{p} dE ,$$
(1)

where

S(E) = dE/dx

is the stopping power of the target material.

The differential cross section $d^2\sigma/d\Omega_{p}d\Omega_{r}$ which takes into account the feeding of the 2^+ states from decays of higher-lying states, as well as virtual excitation via these states, is calculated by means of the coupled-channels code of Winther and deBoer.⁸ In order to perform the double integration over particle angle and energy, the calculations were performed in intervals $\Delta \theta_{b} = 4^{\circ}$ from 4 to 180° for at least eight energies. The stopping power dE/dx was calculated by means of a computer program based on a Bethe-type formula, whose parameters were adjusted to give good agreement with experimental data. Agreement to within a few percent was obtained with other calculations, notably those of Northcliffe and Schilling.9

The theoretical yield given by (1) can be expanded in a form suggested by second-order perturbation theory, i.e., in terms of the reduced transition probability $b = B(E2, 0^+ + 2^+)$ and the spectroscopic quadrupole moment q of the first 2^+ state:

$$y = f \boldsymbol{b} (1 + c \boldsymbol{q}) , \qquad (2)$$

613

6

where f and c are functions depending on the experimental conditions, i.e., projectile type, bombarding energy, and excitation energy of the first 2^+ state, θ_{γ} , etc. Both f and c are obtained by calculating the thick-target yield for various values of b and q. The experimental yield can be expanded accordingly:

$$Y = fB(1+cQ), \qquad (3)$$

where *B* and *Q* are now the experimental values for the reduced transition probability and the spectroscopic quadrupole moment of the first 2⁺ state, i.e., $B = B(E2, 0^+ \rightarrow 2^+)_{exp}$, $Q = Q_{exp}$. It is, furthermore, convenient to introduce the calculated and experimental ratios of γ -ray yields of two isotopes *i* and *j*:

$$r_{ij} = \frac{y_i}{y_j} = \frac{f_i}{f_j} \frac{b_i}{b_j} \frac{1 + c_i q_i}{1 + c_j q_j}, \qquad (4)$$

$$R_{ij} = \frac{Y_i}{Y_j} = \frac{f_i}{f_j} \frac{B_i}{B_j} \frac{N_i}{N_j} \frac{\epsilon_i}{\epsilon_j} \frac{1 + c_i Q_i}{1 + c_j Q_j}, \qquad (5)$$

and the double ratio

$$\tau_{ij} = R_{ij} / r_{ij} \,. \tag{6a}$$

In Eq. (5), N_i designates the relative abundance of isotope *i* in the target and ϵ_i the efficiency of the γ -ray detector for the radiation from the 2⁺ $\rightarrow 0^+$ decay of isotope *i*. Of particular interest is the quantity

$$\tau^{0}_{ij} = R_{ij} / \gamma^{0}_{ij}, \qquad (6b)$$



FIG. 1. γ -ray spectrum resulting from bombardment of an enriched target with 56-MeV S³² ions.

where $r_{ij}^0 = r_{ij}(q_i = 0, q_j = 0)$ is the yield ratio calculated under the assumption $q_i = q_j = 0$. Making use of the fact that $c_j Q_j \ll 1$, one obtains from Eqs. (6), (4), and (5):

$$\tau_{ij}^{0} = \frac{R_{ij}}{r_{ij}^{0}} = \frac{B_{i}b_{j}}{B_{j}b_{i}}\frac{N_{i}}{N_{j}}\frac{\epsilon_{i}}{\epsilon_{j}}\left[1 + c_{i}\left(Q_{i} - \frac{c_{j}}{c_{i}}Q_{j}\right)\right]$$
$$= \alpha_{ij}\left[1 + c_{i}\left(Q_{i} - \frac{c_{j}}{c_{i}}Q_{j}\right)\right], \qquad (7)$$

where

$$\alpha_{ij} = \frac{B_i b_j}{B_j b_i} \frac{N_i}{N_j} \frac{\epsilon_i}{\epsilon_j}$$

The ratio c_j/c_i is very weakly dependent upon the bombarding energy E_b , detection angle θ_γ , and projectile type, changing by only about 1% under widely differing bombarding conditions and may thus be treated as a constant. The coefficients $c_i(A_p)$ alone, however, are roughly proportional to the projectile mass A_p . Thus, if experiments with various particles are carried out, it is possible to plot $\tau_{ij}^0 = R_{ij}/r_{ij}^0$ as a function of $c_i(A_p)$. The intercept of the ensuing straight line with the τ_{ij}^0 axis is equal to α_{ij} and thus yields ratios of B(E2) values. Its slope *m* is given by

$$m = \alpha_{ij} [Q_i - (c_j/c_i)Q_j],$$

and since c_j/c_i is known, the linear combination $Q_i - (c_j/c_i)Q_j$ of the quadrupole moments of any two isotopes *i* and *j* in the target for which the relative yields have been determined can be calculated. It should be pointed out, that the determination of quadrupole moments by this method is independent of B(E2) values, relative detector efficiencies, and isotopic abundances. The latter two quantities are only of importance if relative B(E2) values are extracted from the value of the intercept α_{ij} with the τ_{ij}^0 axis.

EXPERIMENTAL CONSIDERATIONS

Ion beams of helium-4 at 10.0, 12.0, and 14.0 MeV, oxygen at 36.0, 42.0, and 48.0 MeV, and sulfur at 48.0, 52.0, and 56.0 MeV were produced by The University of Pittsburgh's three-stage tandem Van de Graaff accelerator. A true coaxial Ge(Li) detector with a nominal active volume of 40 cm³ was used to determine the relative γ -ray yields. The detector was located at a distance of 10.0 cm from the target at an angle $\theta_{y} = 125^{\circ}$ for all measurements. The finite solid-angle correction factors for the detector were determined by doing a spline interpolation on energy of the correction factors calculated by Camp and van Lehn¹⁰ for the geometry present in this experiment. The results were, furthermore, corrected for selfabsorption in the target. The efficiency of the dePARTIAL DECAY SCHEMES



FIG. 2. Partial level scheme of the even-even Os isotopes. Energies are in keV.

tector was measured to a precision of 2% using several calibrated γ -ray sources.¹¹ The resolution of the detector was 2.4 keV full width at half maximum in the energy region of interest.

Two thick targets were used during the course of the experiment, a 250-mg/cm²-thick natural osmium target and a 95-mg/cm²-thick enriched Os¹⁸⁴ target. The isotopic enrichment of the latter was Os¹⁸⁴: 2.25%, Os¹⁸⁶: 4.79%, Os¹⁸⁷: 2.52%, Os¹⁸⁸: 16.9%, Os¹⁸⁹: 16.8%, Os¹⁹⁰: 25.2%, and Os^{192} : 32.5%. A typical γ -ray spectrum resulting from bombardment of our enriched target with a 56-MeV S³² beam is shown in Fig. 1. The areas under the full-energy peaks of interest were obtained by means of the computer program AUTOFIT by Milner.¹² This program performs a least-squares fit to the data using a functional form which consists of a sum of Gaussian curves representing the contributions of all photopeaks plus a background term.

It is well known that experiments involving γ ray-yield determinations can be influenced through the so-called deorientation effect which consists in a deorientation of the excited nucleus in the time interval between excitation and deexcitation.¹³ This deorientation is due to the interaction of the magnetic moment of the recoiling nucleus in flight or after it has been stopped with extranuclear magnetic fields and results in an attenuation of the γ -ray angular distribution. Angular distribution of γ rays deexciting the first 2⁺ states of Os¹⁹² and Os¹⁹⁰ were determined for He⁴ ions, O¹⁶ ions, and S³² ions. No deorientation was found within the limits set by the precision of the measurement (1%). Thus, if deorientation effects are present, they are sufficiently small to be negligible in the analysis.

CALCULATIONS

The γ -ray yield calculations require as input the energies and spins of all levels which contribute significantly to the population of the first 2⁺ state, as well as all E2 and M1 matrix elements connecting these states, and the corresponding internalconversion coefficients. The states considered in

TABLE I. Matrix elements used in the calculations. $\mathfrak{M}(E2)$ matrix elements are in units e b, $\mathfrak{M}(M1)$ matrix elements are in units $e\hbar/2mc$.

	Os ¹⁹²	Os ¹⁹⁰	Os ¹⁸⁸	Os ¹⁸⁶	Os ¹⁸⁴
$\langle 0^+ $ M (E2) $ $ 2+ $ angle$	-1.443	-1.559	-1.683	-1.817	-1.955
$\langle 0^+ $ M (E2) $ 2^+ ' angle$	0.442	0.497	0.488	0.46	0.19
$\langle 2^+ \mathfrak{M}$ ($E2 angle 4^+ angle$	-2.197	-2,371	-2.655	-2.764	-3.199
$\langle 2^{+\prime} \mathfrak{M}(E2)] 2^{+} angle$	1.391	1,282	0.895	0.766	0.046
$\langle 4^+ \mathfrak{M} (E2) 6^+ \rangle$	•••	•••	•••	•••	-4.035
$\langle 2^{+\prime} \mathfrak{M}(M1) 2^{+} \rangle$	0.0671	0.0374	0.0092	•••	•••



FIG. 3. Plot of $\tau_{i,192}^0$ versus c_i . The straight lines represent least-squares fits to the data.

the various isotopes are shown in Fig. 2. The reduced matrix elements $\langle 0^+ || \mathfrak{M}(E2) || 2^+ \rangle$ and $\langle 2^+ \| \mathfrak{M}(E2) \| 2^+ \rangle$ were treated as variable parameters, whereas all other E2 matrix elements and all M1 matrix elements were taken from experiment wherever possible and from considerations of systematics otherwise. In particular the E2matrix elements of Os¹⁸⁶ through Os¹⁹² were taken as weighted averages of the measurements of Prvor and Saladin,³ Casten et al.,¹⁴ and Stelson and Grodzins.¹⁵ For Os¹⁸⁴, the B(E2) values 0⁺ to 2⁺ and 0^+ to $2^{+\prime}$ were assigned on the basis of systematics in this region. The remaining E2 matrix

616

elements were obtained using the rotational-model prediction. The only transitions where M1 admixtures are of importance are $2^{+\prime}$ to 2^{+} decays. For Os^{192} , Os^{190} , and Os^{188} , the relevant $B(M1, 2^{+\prime} - 2^{+})$ values have been determined by Milner et al.¹⁶ The B(M1) values for Os¹⁸⁶ and Os¹⁸⁴ have not been measured, but from the systematics in the osmium region it is evident that these matrix elements are sufficiently small so that they can be neglected. The E2 and M1 matrix elements used in this calculation are summarized in Table I. The internalconversion coefficients were taken from the tables of Hager and Seltzer.¹⁷

TABLE II. Summary of the quadrupole moments determined in this experiment and comparison with the results of other authors. All moments are quoted in units e b.

	Present expt.	Pryor and Saladin (Ref. 3)	Wagner <i>et al.</i> (Ref. 18) (Mössbauer)	Kumar and Baranger (Ref. 1)	Rotational model
Os ¹⁹²	-0.50 ± 0.20	-0.41 ± 0.20		-0.359	-1.31
Os ¹⁹⁰	-0.95 ± 0.21	-0.99 ± 0.13		-0.891	-1.41
Os ¹⁸⁸	-1.32 ± 0.23	-1.31 ± 0.34	-1.81 ± 0.24	-1.160	-1.52
Os ¹⁸⁶	-1.47 ± 0.54		-1.80 ± 0.22	-1.409	-1.65
Os ¹⁸⁴	-2.4 ± 1.1				-1.77

	Present expt.	Pryor and Saladin (Ref. 3)	Casten <i>et al.</i> (Ref. 14)	Stelson and Grodzins (Ref. 15)	Kumar and Baranger (Ref. 1)
Os ¹⁹²	2.09 ± 0.21	2.04 ± 0.06	2.22 ± 0.34	2.15 ± 0.20	2,576
Os ¹⁹⁰	2.48 ± 0.25	2.39 ± 0.06	2.50 ± 0.37	2.55 ± 0.25	2.595
Os ¹⁸⁸	2.69 ± 0.27	2.90 ± 0.08	2.70 ± 0.40	2.75 ± 0.15	2.731
Os^{186}	2.88 ± 0.39		3.10 ± 0.40	3.11 ± 0.11	2.950
Os ¹⁸⁴	3.20 ± 0.62				

TABLE III. Summary of B(E2) values in units of $e^2 b^2$ obtained in the present experiment and comparison with other experiments.

RESULTS AND DISCUSSION

6

The results of least-squares fits to the points $\tau_{i,j=192}^{0}$ for i=190, 188, 186, and 184 are shown in Fig. 3. Due to the very low isotopic abundances of Os^{186} and Os^{184} in the target (4.79 and 2.25%) and the small excitation probability with low-mass projectiles, it was not possible to extract with sufficient precision γ -ray yields resulting from bombardment of Os^{186} and Os^{184} with α particles. This accounts for the rather large uncertainties in the quadrupole moments of these isotopes. The slope determines the linear combination of the quadrupole moments Q_i and Q_{192} as indicated in the figure. The number near each point specifies the bombarding energy in MeV. It should be noted that there is nothing unique about the choice of Os^{192} as the reference nucleus. The absolute quadrupole moments were determined by combining these relative moments with those measured by Pryor and Saladin.³ This combination was done by selecting a value for the quadrupole moment of Os^{192} which minimizes the sum of the differences between the two sets of data.

From the intercepts of the straight lines one obtains relative B(E2) values. In contrast to the quadrupole moments, these intercepts and hence the relative B(E2) values depend on the relative isotopic abundance and detector efficiency.

Table II summarizes the quadrupole moments determined in this experiment. Also shown for

comparison are the results of Pryor and Saladin,³ the Mössbauer results of Wagner *et al.*,¹⁸ the predictions of the Kumar and Baranger¹ calculations, and the rotational-model predictions. There is excellent agreement between the present results and those of Pryor and Saladin.³ The experimental results are, furthermore, in very good agreement with the theory of Kumar and Baranger.¹ The results for Os¹⁸⁶ are consistent with the Mössbauer measurements of Wagner *et al.*,¹⁸ but for Os¹⁸⁸ the agreement is marginal.

Table III summarizes the B(E2) values obtained in the present experiment and compares them with the results of other experiments and the Kumar-Baranger¹ prediction. There is again excellent agreement between the various sets of data and the predictions of Kumar-Baranger.¹

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PHYSICAL REVIEW C

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VOLUME 6, NUMBER 2

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Independent Yields of Krypton and Xenon Isotopes in Thermal-Neutron Fission of ²³⁵U. Observation of an Odd-Even Effect in the Element Yield Distribution*

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The fractional and independent fission yields of krypton isotopes (masses 87 to 94) and xenon isotopes (masses 137-143) produced in thermal-neutron fission of 235 U were measured using a mass separator operating in an on-line mode with a fission source exposed to timecontrolled neutron fluxes. The noble-gas isotopes were collected simultaneously at given times and each mass was measured separately. The distribution of the isotopic yields of each element was found to be Gaussian, in general agreement with Wahl's estimates. The total independent yields of krypton and xenon, as well as their complementary elements, barium and strontium, were found to indicate an odd-even effect in the element yield distribution. Even-Z elements are formed in higher yields than their odd-Z neighbors (20-50% higher); the even-Z yields are also higher than Wahl's "normal" element yields (~12% higher for Kr and Ba and ~40% for Xe and Sr). Half-life values of 20 nuclides produced in the decay of krypton and xenon isotopes were measured as well.

I. INTRODUCTION

A major question bearing on the understanding of the mechanism of low-energy fission is the relation between the nuclear structure of the primary fission products and the distribution of nuclear charge and mass in the fission process. The dispersion of independent fission yields along an isobaric chain seems to be represented by a normal Gaussian equation,¹ given by

$$P(Z) = (C_Z \pi)^{-1/2} \exp \left[-\frac{(Z - Z_P)^2}{C_Z} \right],$$

where P(Z) is the fractional independent yield of a nuclide of nuclear charge Z, Z_P is the most probable charge, and C_Z is related to the width of the dispersion.

By compiling the measured independent (or partially cumulative) fission yields²⁻⁹ of at least two members of 19 mass chains, Wahl *et al.*¹⁰ were able to obtain a width parameter $\sigma_z [C_z = 2(\sigma_z^2 + \frac{1}{12})]$ which was nearly constant for those masses (σ_z = 0.56±0.06). The Z_P function adopted by Wahl et al.¹⁰ was a strictly empirical one, based on the 19 values of Z_p found experimentally. By using this Z_p function, assuming a constant width of the isobaric dispersions, and using the experimental fission chain yields, Wahl calculated independent yields of all the nuclides produced in the thermal fission of ²³⁵U.¹¹

The only doubt as to the constancy of σ_z was raised by the results of Strom *et al.*,⁸ who observed a systematic decrease in this parameter in the mass range 131 to 133. However, the yields of ¹³²Sn and ¹³³Sb have recently been remeasured,¹²⁻¹⁴ and the results are in much better agreement with the estimated values. The observed high yield of ¹³²Sn was explained^{12, 15} by a shell effect favoring the production of this doubly closed-shell nucleus. However, because of the difficulty of making accurate experimental measurements in this region, it is too early to draw final conclusions and more experiments are called for.

From the estimated values of the independent yields, the isotopic yield distribution can be con-