

CONCLUSIONS

The elastic-scattering data only serve to confirm what was obviously expected. When one moves into the more classical region, as attested by the larger values of the parameter η , and restricts the discussion to sufficiently forward angles, the elastic cross section is essentially Rutherford. Further, the radius parameter, extracted from fitting mainly this forward-angle region, as opposed to the diffraction-like extrema, are reminiscent of low-energy alpha-particle-scattering results. These radii appear to be systematically, but not significantly, smaller than those obtained in the presence of diffraction-like effects. This is somewhat at odds with the tendency toward larger radii usually observed as

one goes to lower energies of the same scattering system.^{2,3}

The processes involved in the fragmentation experiment are, admittedly, rather poorly understood. The transfer reactions appear to be strong contributors at the forward angles, as is to be expected. One can only speculate on the origin of the remaining structure, however. Work is being continued on this analysis.

ACKNOWLEDGMENTS

The authors would like to acknowledge the participation of Dr. E. Newman, Dr. P. G. Roll, and Dr. A. M. Smith at various stages of the elastic-scattering work. Without their assistance it could not have been completed. Thanks are also due to Dr. P. G. Roll and Dr. C. E. Anderson for assistance in the fragmentation work.

Isotope Shift in the Spectrum of Osmium*

ARTHELLA P. HINES AND JOHN S. ROSS

Department of Physics, Rollins College, Winter Park, Florida

(Received October 27, 1961; revised manuscript received February 9, 1962)

Using enriched isotopes, the isotopic shift in five lines of the osmium spectrum, involving transitions of the type $5d^6s_1p - 5d^6s_2$, was measured with a Fabry-Perot interferometer. The average relative isotope shift for the even isotopic pairs, $\Delta\nu(192-190):\Delta\nu(190-188):\Delta\nu(188-186):\Delta\nu(186-184)$, was found to be 1.00:1.12:1.29:1.31, showing a leveling off in the isotope shift as the last neutron pair is subtracted. This results in a predicted intrinsic quadrupole moment for Os^{184} of $6.0 \times 10^{-24} \text{ cm}^2$. The odd-isotope shift $\Delta\nu(192-190):\Delta\nu(189-187)$ was determined to be 1.00:1.27. The average value for $\Delta\nu(190-188):\Delta\nu(190-189)$ and $\Delta\nu(188-186):\Delta\nu(188-187)$ was found to be 1.00:0.69, illustrating the usual even-odd staggering effect.

INTRODUCTION

RESULTS from experimental measurements of atomic isotope shifts provide information about the variation in the nuclear charge distribution, which results from a change in the number of neutrons in a nucleus. Osmium lies between the heavy elements lead and mercury, where the even isotopes exhibit¹ a progressively decreasing shift with decreasing mass, and hafnium and tungsten, which show^{2,3} an inversion in their even-isotope shifts. Due to its location, therefore, the isotope shift of osmium is of particular interest.

Optical examination of the osmium spectrum by several investigators,^{1,4-6} using natural samples, has shown that there is an increasing isotope shift between the even isotopes as one proceeds from atomic mass number 192 down to 186, and a pronounced even-odd

staggering effect in 189. Murakawa and his co-workers determined the nuclear spin of Os^{189} to be $\frac{3}{2}$ and found a value of the magnetic moment in agreement with the value $\mu(\text{Os}^{189}) = +0.65065 \text{ nm}$, determined⁷ by the method of nuclear induction. Murakawa⁸ assumed the nuclear spin of Os^{187} to be $\frac{1}{2}$. Steudel and his co-workers,^{9,10} using a radiogenic 100% enriched sample of Os^{187} , confirmed this value of the nuclear spin, determined $\mu(\text{Os}^{187}) = +0.0653 \text{ nm}$, and observed even-odd staggering for 187.

The availability of the enriched isotopes of osmium provides samples for better measurement of the hyperfine components due to the odd isotopes, for obtaining more accurate even isotope positions, especially 186, and for determining the location of the 184 isotope.

* Supported by a grant from the National Science Foundation.

¹ J. Blaise, theses, University of Paris, 1958.

² W. L. Barr, *J. Opt. Soc. Am.* **48**, 658 (1958).

³ J. Blaise and G. Gluck, *J. phys. radium* **20**, 466 (1959).

⁴ S. Suwa, *Phys. Rev.* **83**, 1258 (1951).

⁵ K. Murakawa and S. Suwa, *Phys. Rev.* **87**, 1048 (1952).

⁶ K. Murakawa and T. Kamei, *Phys. Rev.* **105**, 671 (1957).

⁷ H. R. Loeliger and L. R. Sarles, *Phys. Rev.* **95**, 291 (1954).

⁸ K. Murakawa, *Phys. Rev.* **98**, 1285 (1955).

⁹ G. Guthöhrlein, H. Kopfermann, G. Nöldeke, and A. Steudel, *Naturwissenschaften* **46**, 598 (1959).

¹⁰ G. Guthöhrlein, G. Nöldeke, and A. Steudel, Program of Atomic Spectroscopy Symposium, Argonne National Laboratory, June, 1961.

TABLE I. Isotopic constitution of samples of osmium, in percent.

Sample \ Isotope	192	190	189	188	187	186	184
Natural	41.00	26.40	16.10	13.30	1.64	1.59	0.018
189	2.50	8.50	87.30	1.70	0.05	0.05	0.05
187	8.43	9.38	9.27	25.74	45.76	1.43	0.01
186	9.80	8.77	7.31	9.54	3.31	61.27	0.10
184	32.50	24.20	16.80	16.90	2.52	4.79	2.25

EXPERIMENTAL DETAILS

Natural osmium and the four enriched samples,¹¹ listed in Table I, were excited, using neon as the carrier gas, in liquid-nitrogen-cooled hollow-cathode discharge tubes, similar in design to that described by Murakawa.¹² The samples were photographed with a Fabry-Perot interferometer mounted external to a Hilger E-495 spectrograph. The interferometer plates utilized a 5-layer dielectric coating, with a reflectance of 90% from 4000 Å to 5000 Å. Part of the data from the comparator readings was reduced on an IBM 1620 computer.

RESULTS

It was possible to observe directly the isotope 184 in the three lines 4794, 4261, and 4112, and to detect its presence in two other lines, 4420 and 4136. In the latter cases, the increased intensity in the high-frequency component (*A*), due to 189, indicated the presence of 184. In neither case was it possible to resolve the pattern, although in 4420 it was possible to detect a shift of the center of gravity, due to the two components, compared to the position of the 189 component alone.

In order to have compatible intensities for measurement, the 190–186 isotope shift values were determined from short exposures on natural osmium which had been enriched with the 186 isotope. After the components, due to 189 and 187, had been located relative to the even isotope 190, their presence was corrected for, when necessary, in the final values of the positions of the other even isotopes. The values for the observed positions of the even isotopes are listed in Table II. The classification of the osmium I lines is that given by Van Kleef.¹³ The error values represent three times the probable error, derived from readings taken on spec-

trograms with different spacers and isotopic samples. Since there are hyperfine structure components due to 189 close to all of the strong even isotopes except 190, measurements were made relative to this isotope and are so listed in the tables.

It was possible to detect the splitting of 187 into two components except in 4112. This particular line showed a strong, narrow component, unresolved by the interferometer with a 30-mm spacer, indicating that the splitting of 187 is very small. In the other lines the presence of a considerable amount of 188, which lies close to the low-frequency component of 187(β), makes a determination of the 187 splitting somewhat difficult with the available enrichment. In 4136 it was not possible to resolve 188 and 187(β). Due to the difficulty in obtaining high resolution without overlapping of components, the 187 values do not have a high degree of accuracy, but are in agreement with the more precise values obtained by Steudel.^{9,10} The 187 components were located relative to the even isotopes by measurements to either 188 or 190 on the spectrograms taken with the enriched samples.

Examination of the enriched 189 sample showed four strong components in all of the lines except 4112, which exhibited two strong, sharp components plus a third broad one with medium intensity. In all cases the intensities showed a flag pattern with the high-frequency component being the strongest. The 189 components were located relative to the even isotopes by measuring directly to 190 on the enriched 189 spectrograms, which confirmed measurements from either 188 or 190 to the outer components (*A* or *D*) of 189 on the spectrograms taken with natural osmium. Table III lists the observed positions for the 189 and 187 hyperfine components, relative to the position of 190.

Some weak components, due to the splitting of the $5d^66s^2$ configuration, were detected in 4261, 4420, and 4794 on the spectrograms using enriched 189. Due to their faintness and proximity to strong lines, these components could not be accurately measured. Therefore, all the center-of-gravity calculations for the two odd isotopes were carried out assuming that the splittings of the lower even-energy states with the 5^2d^66s configuration are small, compared to the higher

TABLE II. Isotope positions in the spectra of Os I (in units of 10^{-3} cm^{-1}).

Wavelength (Å)	Classification	Isotope						
		192	190	189 (c.g.)	188	187 (c.g.)	186	184
4793.994	$5d^66s(^6D)6p^7D_3^{\circ} - 5d^66s^2^5D_3$	-47.9±0.5	0	32.2±2.2	53.8±0.8	101.0±1.6	118.1±0.9	181.3±0.5
4420.468	$5d^66s(^6D)6p^7D_4^{\circ} - 5d^66s^2^5D_4$	-53.4±0.5	0	40.5±1.2	59.1±0.5	108.2±1.0	126.7±0.6	(198.5±1.7) ^a
4260.854	$5d^66s(^6D)6p^7D_5^{\circ} - 5d^66s^2^5D_4$	-56.2±0.5	0	43.9±1.5	64.2±0.5	114.5±1.2	137.8±1.0	208.0±1.9
4135.784	$5d^66s(^6S)6p^7P_4^{\circ} - 5d^66s^2^5D_3$	-46.7±0.7	0	34.4±1.0	54.2±0.7	93.8±2.3	113.6±1.1	(170.7±3.0) ^a
4112.018	$5d^66s(^6D)6p^7F_2^{\circ} - 5d^66s^2^5D_1$	-55.5±0.6	0	43.1±2.5	60.6±0.6	112.7±1.0	131.7±1.0	206.1±0.8

^a Not resolved from *A* component due to 189.

¹¹ Obtained from Union Carbide Nuclear Company, Oak Ridge National Laboratory.

¹² K. Murakawa, J. Phys. Soc. Japan 9, 391 (1954).

¹³ Th. A. M. Van Kleef, Koninkl. Ned. Akad. Wetenschap. Proc., Ser. B, 63, 501 (1960).

TABLE III. Positions of hyperfine components in Os¹⁸⁹ and Os¹⁸⁷ (in units of 10⁻³ cm⁻¹).

Wavelength (Å)	190	189(D)	189(C)	189(B)	189(A)	187(β)	187(α)
4794	0	-99.1±1.2	-49.0±1.0	29.8±1.8	135.5±1.3	87.1±1.1	111.4±1.1
4420	0	-152.9±1.4	-65.0±1.6	52.5±0.9	195.8±0.8	88.1±0.5	124.3±1.0
4261	0	-270.1±0.6	-87.5±0.9	99.4±1.2	269.4±0.9	85.6±1.0	138.5±0.7
4136	0	-162.8±0.6	-55.9±0.9	60.0±0.7	170.7±0.6	(69.1±2.0) ^a	112.3±1.2
4112	0	-53.7±1.8		37.9±1.8	119.7±1.7		112.7±1.0

^a Not resolved from 188.

odd-terms having the 5d⁶6s6p configuration. The resulting center-of-gravity positions are listed in Table II.

DISCUSSION

Although there are variations in the shifts of the individual lines, they all appear to show the same general even-isotope shift pattern and, except for the 189 center-of-gravity value, the same odd-isotope shift. The resulting average isotope shifts are shown in Fig. 1. Here, the 192-190 shift has been taken as unity and a weighted average, according to accuracy, is obtained for the relative positions of the other isotopes and the centers of gravity.

In osmium, the general trend of the relative isotope shift between even isotopic pairs is to progressively increase as the mass decreases. However, it is found within experimental limits that the 186-184 shift is of the same order of magnitude as that of 188-186. There is no decided tendency for the isotope shift between consecutive neutron pairs to continue to increase or to decrease as the last neutron pair is subtracted.

The only neighboring element which has overlapping neutron pairs for comparison is tungsten. In Table IV the relative isotope shifts in osmium and tungsten³ are shown, normalized to the neutron pair 110-112. With reference to the shift in osmium for the isotopic pair with mass 184-186, it is seen that in tungsten there is a decided increase in the relative shift for the corresponding neutron pair 108-110.

Comparison of experimental isotope shift measurements and theoretical calculations has shown¹⁴⁻¹⁸ that the most important contributions to the isotope shift

phenomena in the heavy elements comes from the nuclear volume and nuclear deformation effects.

The deformation of a nucleus is given by $e = (a - b) / r_0$, where a and b are the major and minor radii of the nuclear spheroid and r_0 is the radius of a sphere of the same volume. This deformation is related to the intrinsic quadrupole moment Q_0 by the relationship

$$e(1 + 0.17e + \dots) = 5Q_0 / 4Zr_0^2.$$

Meligy¹⁸ has computed a theoretical isotope shift constant which is the sum of C_v , a volume-dependent constant based upon a spherical nucleus in which the charge distribution has a trapezoidal shape as determined by electron-nucleus scattering experiments, and C_e , a deformation-dependent isotope shift constant. He has shown that if the experimental isotope shift constant C_{exp} , as defined by Kopfermann,¹⁶ is substituted for this theoretical constant, the following expression results:

$$e_2^2 - e_1^2 = -\frac{3\delta A}{2A} \left(\frac{C_{exp}}{C_v} - 1 \right),$$

where e_1 and e_2 are the different nuclear deformations of the lighter and heavier isotopes, respectively, and A is the atomic mass of the lighter isotope.

If the intrinsic quadrupole moment is known for one isotope of an element, a knowledge of C_v , of C_{exp} , and of the relative isotope shift will allow the deformations and intrinsic quadrupole moments of the other isotopes to be predicted.

Table V shows these isotope shift constants for osmium. The intrinsic quadrupole moments of the isotopes 186, 188, and 190 have been determined^{19,20}

TABLE IV. Comparison of even-even isotope shift ratios for osmium and tungsten.

Element	Atomic number	Neutron pair				
		106-108	108-110	110-112	112-114	114-116
W	74	0.79	1.13	1.00		
Os	76		1.02	1.00	0.87	0.78

¹⁴ L. Wilets, D. L. Hill, and K. W. Ford, Phys. Rev. **91**, 1488 (1953).

¹⁵ A. R. Bodmer, Proc. Phys. Soc. (London) **A67**, 622 (1954).

¹⁶ P. Brix and H. Kopfermann, Revs. Modern Phys. **30**, 517 (1958).

¹⁷ A. S. Meligy, Nuclear Phys. **14**, 248 (1959).

¹⁸ A. S. Meligy, S. Tadros, and M. A. El-Wahab, Nuclear Phys. **16**, 99 (1960).

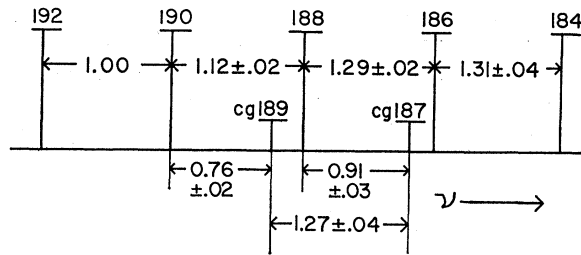


FIG. 1. Relative isotope positions in the Os I spectrum.

¹⁹ K. Alder, A. Bohr, T. Huus, B. Mottelson, and A. Winther, Revs. Modern Phys. **28**, 432 (1956).

²⁰ E. Ye. Berlovich, K. Grotovski, M. Bonitz, V. E. Breslav, and B. K. Preobrazhensky, Nuclear Phys. **6**, 672 (1958).

TABLE V. Isotope shift constants for osmium.

Isotopic pair	C_v (cm^{-1})	C_{exp} (cm^{-1})	C_{exp}/C_v	Relative shift
184-186	0.291	0.170	0.584	1.31
186-188	0.289	0.168	0.581	1.29
188-190	0.287	0.146	0.509	1.12
190-192	0.286 ^a	0.130 \pm 0.20 ^b	0.455	1.00

^a See reference 18.^b See reference 1.

from transition probabilities based on observed cross sections for Coulomb excitation of rotational levels. From the determination of $Q_0(\text{Os}^{190})$ Meligy deduced a value of $Q_0(\text{Os}^{192}) = 3.06 \times 10^{-24} \text{ cm}^2$ from isotope shift results.

Table VI shows these intrinsic quadrupole moments and deformations together with those predicted from our relative isotope shifts (Fig. 1) for the even-even isotopic pairs in osmium. The predicted Q_0 values for osmium 186 and 190 are based on $Q_0(\text{Os}^{188})$ determined from transition probabilities. Those Q_0 values predicted for osmium 184 and 192 are based on the transition probability values for $Q_0(\text{Os}^{186})$ and $Q_0(\text{Os}^{190})$, respectively. The Q_0 values predicted from isotope shift measurements by this procedure are in good agreement with Coulomb excitation results, and lead to a prediction of $Q_0(\text{Os}^{184}) = 6.0 \times 10^{-24} \text{ cm}^2$. The major error in the

TABLE VI. Intrinsic quadrupole moments and deformations for osmium.

Isotope	From transition probabilities		Predicted from isotope shift	
	e	Q_0 (10^{-24} cm^2)	e	Q_0 (10^{-24} cm^2)
184			0.197	6.0
186	0.179	5.5	0.184	5.6
188	0.165	5.1
190	0.136	4.2	0.139	4.3
192			0.099	3.1

values predicted from isotope shift measurements comes from the uncertainty of $\pm 15\%$ in C_{exp} .

It can be seen that as one subtracts neutron pairs in the even-massed isotopes of osmium there is a corresponding increase in the intrinsic quadrupole moment, with a leveling off in the rate of change of the nuclear deformation.

The relative shift of 1.27 for 189-187 as shown in Fig. 1 indicates that in osmium the odd-isotope shift is of the same order of magnitude as that for the even-isotopic pairs which show the largest relative shift. Isotope shift data are available on mercury¹ and hafnium,² which are other heavy elements with two odd isotopes. Table VII compares the relative shifts for the even-even pairs to that for the odd-odd pair. The relative shifts have been normalized to that of the odd-odd pair. In each case, the odd-isotope shift is

TABLE VII. Comparison of even-even and odd-odd relative isotope shifts in hafnium, osmium, and mercury.

Element	Atomic number	Isotopic pairs of even mass			Isotopic pair of odd mass
Hf	72	(174-176)	(176-178)	(178-180)	(177-179)
		0.79	0.75	0.90	1.00
Os	76	(184-186)	(186-188)	(188-190)	(187-189)
		1.03	1.02	0.88	1.00
Hg	80	(196-198)	(198-200)	(200-202)	(199-201)
		0.69	0.83	0.93	1.00

closest to the even-isotopic pairs which show the largest relative shift.

The even-odd staggering effect is observed in osmium 189 and 187 in the usual sense, with the center of gravity of the isotope with odd mass shifted toward the even isotope of lighter mass. It is found that $\Delta\nu(190-189)/\Delta\nu(190-188)$ is 0.68 ± 0.02 and $\Delta\nu(188-187)/\Delta\nu(188-186)$ is 0.71 ± 0.04 .

ACKNOWLEDGMENTS

The authors wish to thank National Science Foundation Undergraduate Research Participants Robert Fox, Paul Haynes, and Merton VanPelt for their assistance on this project. In addition they are grateful to Dr. Jean Blaise, Maitre de Recherches au C.N.R.S., Bellevue, France, for supplying a copy of his unpublished thesis work on osmium.