

## THE ISOTOPE EFFECT IN THE SPECTRA OF THE HEAVY ELEMENTS

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## INTRODUCTION

STUDIES of the isotope effect in the atomic spectra of the heavy elements have continued to grow at a faster pace over the last decade. We see from the review by A. R. Striganov and Yu. P. Dontsov<sup>[1]</sup> published in 1955 that about 200 theoretical and experimental papers had been published on this subject in the entire previous period beginning in 1927. About 120 papers have been devoted to the isotope effect in the spectra of the heavy elements in the period since 1955. Such a heightened concern with this problem involves the pertinence of the isotope effect in atomic spectra, not only in studying the electron cloud of the atom, but also in studying the properties of the nucleus and in creating models for it. This is understandable, since the isotope effect results from interaction of the electron cloud of the atom with its nucleus. Hence this phenomenon makes it possible to establish the electron configurations for the energy levels of the atom. In addition, it permits one to find the deformation parameters of the nucleus and to get valuable information on the filling of the neutron and proton shells.

The original theory of Racah, Breit, and Rosenthal on the isotope shift developed for the spectra of the heavy elements was based on a drop model of the nucleus. It implied that the displacements of the spectral lines of the isotopes of a particular element must be equidistant with increasing mass number. However, experiment has shown that the isotope shifts are not equidistant in the spectra of some elements.<sup>[1,2,3]</sup>

Anomalously large shifts have been found in the spectra of neodymium, samarium, and gadolinium. In these three cases, a marked increase in the shift was observed between the lines of the isotopes having neutron numbers  $N = 88$  and  $N = 90$ . Jumps in the isotope shift have also been found in the spectra of strontium, zirconium, and molybdenum between the lines of the isotopes having  $N = 50$  and  $N = 52$ , in the spectrum of cerium between the lines of the isotopes having  $N = 82$  and  $N = 84$ , and in the spectrum of lead between the lines of the isotopes having  $N = 126$  and  $N = 128$ . It has also been established that the isotope shift between the lines of other isotopes is not

equidistant, although this effect is generally less clearly marked.

As we know, the isotope shift in the spectrum of a particular element is determined by the variation in the mean-square radius of the charge distribution of the nucleus in its ground state.<sup>[5,6]</sup> The mean-square radius depends on the filling of the neutron shells, on the deformation of the nucleus, and on surface oscillations of the nucleus. Each of these can be the determining factor in the isotope shift of the levels of an atom, or two or all three factors can give comparable contributions.<sup>[7]</sup>

The jumps in the isotope shifts in the spectra of Sr, Zr, Mo, Ce, and Pb involve the shell structure of the nucleus.<sup>[1,2]</sup> They occur between the lines of even-even isotopes having completely-filled neutron shells in the nuclei and the lines of subsequent isotopes in which a new pair of neutrons begins to occupy a higher shell. The jumps in the isotope shift near the "magic" numbers show that the effective radius of the nucleus increases relatively greatly when the new neutron pair is added. This implies that the effective radius depends on the details of filling of the shells and on the extent to which they are filled by neutrons. This is manifested correspondingly in the isotope shift.<sup>[7]</sup> A. B. Migdal and his associates<sup>[8,9]</sup> have recently made a theoretical study of this problem, based on their theory of the Fermi fluid in finite systems, taking into account the individual properties of nuclei as the number of neutrons and protons in them is varied.

The anomalous shift in the spectra of Nd, Sm, and Gd involves a transition from spherical to deformed nuclei.<sup>[2]</sup> The region of statically-deformed nuclei begins at  $N = 90$  and ends near  $N = 116$ . An apparent increase in the effective radius occurs as the deformed nucleus rotates, owing to averaging over all directions. This is manifested in an extra isotope shift in the direction of increasing volume effect. This extra shift, which has been termed the quadrupole shift, can be appreciable, since the deformation of the nucleus when paired neutrons are added can vary greatly from isotope to isotope. Hence, non-equidistant isotope shifts in the region of deformed nuclei are explained by the uneven variation in the deformation of the nuclei of the isotopes of a particular element.

Studies show that non-equidistant isotope shifts also appear among the spherically-symmetric nuclei. If we base ourselves on the generalized model of the nucleus, we can assume that the reason for non-equidistant shifts in the region of spherical even-even nuclei is nuclear surface oscillations, or as they are usually called, zero-point oscillations in the nuclear electric quadrupole moment.<sup>[6,7,10]</sup> These vibrations blur the boundaries of the nucleus, and this increases the effective radius of the nucleus. The uneven variation of the amplitude of these vibrations in the nuclei of different isotopes produces an uneven variation in the effective radius. This is manifested in an extra isotope shift.

This article will discuss the isotope shift in the spectra of the intermediate and heavy elements. The treatment begins with strontium, zirconium, and molybdenum. Along with the mass effect, the spectra of these elements appreciably manifest a volume effect, which is of the same order of magnitude as the former. Since in such cases the measured quantities are the algebraic sum of the mass and volume shifts, the problem arises of distinguishing the component characteristic of the heavy elements from the total isotope shift.

The experimental data on the isotope shift in the spectra of the heavy and some intermediate elements published since 1954 are presented below. The fundamental advances in the theory of the isotope shift are discussed, and the theoretical data are compared with the experimental results. We show that the isotope shift can be used to determine the deformation parameters, the intrinsic quadrupole moments, and the reduced probabilities of electric quadrupole transitions of deformed nuclei. A complete résumé of the literature on the isotope effect in the atomic spectra of the discussed elements is given at the end of the review.

A review by Stacey<sup>[4]</sup> has appeared since this article was submitted for publication, discussing thoroughly and in detail the isotope effect in the atomic spectra of the heavy elements. The author gives a summary of the experimental data on isotope shifts and isotope-shift constants. However, we should note that outdated results are given for the isotope shifts in the spectra of cerium, ytterbium, and hafnium, while later studies have not been taken into account, e.g.,<sup>[56,79,81]</sup>.

## I. EXPERIMENTAL DATA ON THE ISOTOPE SHIFT

The experimental technique of studying the isotope shift has undergone no substantial changes in the last decade. As before, the Fabry-Pérot interferometer is used to resolve the complex structure of the spectral lines. However, the quality of the measurements has been greatly improved by increase in the resolving power of this instrument and also by use of isolated and enriched isotopes. The resolving power of the interferometer, its transmission, and its degree of contrast have increased considerably, owing to use of reflective dielectric coatings having smaller absorption losses. The use of isolated and enriched isotopes has made it possible to eliminate or at least to decrease considerably the mutual overlap of the components of successive even-even isotopes, and also to reduce interference from the hyperfine structure of the even-odd isotopes. In addition, for every element

for which enriched isotopes were used, it was possible to detect the components of the isotopes whose concentrations in natural samples weren't sufficient for spectroscopic detection. All of this has made it possible to study the isotope shift in a large number of intervals between even-even isotopes. It has made the measurements more reliable, and has made it possible to measure the shifts for many spectral lines. The latter has permitted averaging of the results over a large number of measurements.

As we know, the overall isotope shift can be represented as the sum of three terms:<sup>[1,2]</sup>

$$\Delta\nu = \Delta\nu_n + \Delta\nu_{sp} + \Delta\nu_v,$$

where  $\Delta\nu_n$  is the normal mass shift,  $\Delta\nu_{sp}$  is the specific mass shift, and  $\Delta\nu_v$  is the volume shift. The normal mass shift characteristic of the light elements is explained by the Bohr theory by taking account of the finiteness of the mass of the nucleus with respect to that of an electron. Then the energy levels of the atom prove to depend on the mass of the nucleus. The specific mass shift occurs in the spectra of atoms having two or more electrons. It is due to the exchange interaction of the electrons. The volume shift, which is manifested in the spectra of the heavy elements, is due to the finite dimensions of the nucleus, which cause the nuclear field to deviate from the purely Coulombic field of a point charge.<sup>[1,2]</sup>

In the normal mass shift, the energy levels of the atom of a lighter isotope of a given element lie closer to the series limit than those of the heavier isotopes. Conversely, in the volume shift the levels of the atom of the lightest isotope are farther from the series limit. It has been provisionally agreed to consider the former type of shift in the levels to be negative, and the latter type positive. Correspondingly, the spectral lines also show shifts in different directions. As most studies have done, we shall take a shift in a line to be positive when the component due to the heavier isotope is shifted to higher wave numbers. This direction of shift is characteristic of the normal mass shift. The opposite direction is considered negative, and it is characteristic of the volume shift. If we know the shifts in the lower ( $\Delta T_l$ ) and upper ( $\Delta T_u$ ) levels, we can find the isotope shift by the formula

$$\Delta\nu = \Delta T_u - \Delta T_l,$$

which gives the correct sign, irrespective of the nature of the shift.<sup>[1]</sup>

### 1. The Isotope Shift in the Spectra of Sr, Zr, Mo, and Other Intermediate Elements

Strontium, zirconium, and molybdenum belong to the group of intermediate elements in the periodic table. The isotope shifts in their atomic spectra are very small, and hence they remained unstudied until recent years. However, it was of interest to study the isotope shift in this region of nuclei in connection with the development of the shell structure of the nucleus. This is because each of the cited elements has an isotope containing the magic number of neutrons  $N = 50$ , at which  $1g_{9/2}$  shell becomes filled. By analogy with the isotope shifts in the spectra of lead and cerium, one might expect a jump to be found here in the volume component of the shift.

The first studies were performed by Yu. P. Dontsov.<sup>[36,38]</sup> Using enriched isotopes, he studied in detail the isotope shifts between the components of the even-even isotopes in the spectra of zirconium and molybdenum. He measured the isotope structure for many spectral lines belonging to different transitions. The obtained experimental data showed that the isotope shift between the components of the isotopes Zr<sup>90</sup> and Zr<sup>92</sup> and between Mo<sup>92</sup> and Mo<sup>94</sup> was about 35% greater than for the neighboring pair of heavier isotopes. In both cases, this somewhat enhanced isotope shift occurs between isotopes having neutron numbers  $N = 50$  and  $N = 52$ . That is, it happens when two extra neutrons are added to the completely filled  $lg_{9/2}$  shell, and enter the  $2d_{5/2}$  shell. The spectrum of molybdenum also showed a jump in the isotope shift between the isotopes Mo<sup>98</sup> and Mo<sup>100</sup> having  $N = 56$  and  $N = 58$ , at which the  $2d_{5/2}$  shell becomes filled, and the two neutrons enter the next shell  $lg_{7/2}$ . The non-equidistant isotope shifts indicate that the volume effect plays an appreciable role for this group of elements.

Other investigators<sup>[37,40]</sup> using more highly enriched isotopes have obtained the same type of results on the isotope shift in the spectra of zirconium and molybdenum for nuclei having  $N \geq 50$ . For a quantitative estimate of the jump in the isotope shift between the isotopes having  $N = 50$  and  $N = 52$ , it was important to compare the shift over this interval with that for isotopes having  $N < 50$ . The only element suitable for this purpose is strontium. The difficulty in solving this problem consisted in the fact that here they had to use not only the stable isotopes of mass numbers 84, 86, and 88, but also the radioactive isotope Sr<sup>90</sup>. The study of the isotope shift from this point of view was performed by Heilig and Steudel.<sup>[35]</sup> The obtained results definitely demonstrated a jump in the isotope shift in this region of nuclei.

A complicated problem in studying the isotope shift among the intermediate elements is to distinguish the component due to the volume effect in the experimentally measured shift. The normal mass shift can be easily calculated by Bohr's formula. The specific effect is very difficult to calculate for atoms having multielectron shells. Hence, one usually limits one's self to rough estimates found by indirect reasoning. However, this method doesn't always give an exact enough result. We can consider King's method<sup>[42]</sup> to be the most general way of estimating the mass shift (the algebraic sum of the normal and specific effects). This method is based on the fact that the isotope shift in the spectral lines when volume and mass shifts are present can be expressed by the formula:

$$\Delta\nu = \Delta\nu_v + \Delta\nu_m = EV + M, \quad (1.1)$$

where  $E$  is a factor depending on the electron cloud of the atom,  $V$  is a factor depending on the properties of the nucleus, and  $M$  is the mass effect. For two lines (a) and (i), the shift in the interval (n) between the components of two consecutive even-even isotopes can be represented as follows:

$$\begin{aligned} \Delta\nu_{an} &= E_a V_n + M_a, \\ \Delta\nu_{in} &= E_i V_n + M_i. \end{aligned}$$

We can derive the following relation from these equations:

$$\Delta\nu_{in} = \frac{E_i}{E_a} \Delta\nu_{an} + M_i - \frac{E_i}{E_a} M_a, \quad (1.2)$$

which can be represented in the form

$$\Delta\nu_{in} = B_i \Delta\nu_{an} + A_i, \quad (1.3)$$

where

$$B_i = \frac{E_i}{E_a} \quad \text{and} \quad A_i = M_i - \frac{E_i}{E_a} M_a.$$

When plotted on coordinate axes  $\Delta\nu_{in}$  and  $\Delta\nu_{an}$ , Eq. (1.3) will form a straight line of slope  $B_i$  and intercept  $A_i$  on the  $\Delta\nu_{in}$  axis. In order to construct such a straight line from the experimental data, one must plot against these axes the shifts shown by two lines in corresponding intervals. To determine the coefficients  $B_i$  and  $A_i$ , one should construct a number of such lines, plotting the isotope shifts of different lines along the  $\Delta\nu_{in}$  axis, and the shifts in one particular line along the  $\Delta\nu_{an}$  axis. These lines will give a system of  $k$  equations of the type

$$M_i + B_i M_a = A_i \quad (1.4)$$

with  $k + 1$  unknowns, with  $1 \leq i \leq k$ . We need an additional condition to solve this system. It can be obtained from the known fact that the specific shift in the lines varies both in sign and in magnitude. Hence, one can assume that the specific shift satisfies the condition  $\sum M_{sp}^2 = \min$ . Together with the system of equations (1.4), this condition permits one to obtain the overall mass shifts for all the measured lines. One can thereupon extract the volume component of the shift for each line from the experimentally measured shifts in all the intervals. Here one assumes that the mass shift between the components belonging to isotopes having a mass-number difference  $\Delta A = 2$  remains constant.

Table I gives the results of measuring the isotope shifts in the spectrum of strontium<sup>[35]</sup> for the isotopes of mass numbers 84, 86, 88, and 90. The latter three columns give the shift  $\Delta\nu$  between the components belonging to the respective isotopes. The sign before the magnitude of the shift gives the direction of the shift. The root-mean-square error amounts to  $\pm 0.2 \times 10^{-3} \text{ cm}^{-1}$ .

Figure 1 shows schematically the isotope structure as represented by the mean of the two spectral lines of Sr II. Each component is labeled with its respective mass number and number of neutrons. The positions of the components are plotted as the abscissa. We see from the diagram that the component for the isotope Sr<sup>90</sup> lies between the isotopes Sr<sup>84</sup> and Sr<sup>86</sup>. If there were a mass shift plus an equidistant volume shift, it would lie to the right of the component for Sr<sup>88</sup> toward larger wave numbers. The position of the Sr<sup>90</sup> component in Fig. 1 is explained by the jump in the volume isotope shift in the interval  $\Delta\nu$  (88-90).

Table I. Isotope shift in the spectrum of Sr

$\lambda, \text{ \AA}$	Transition	$\Delta\nu(84-88),$ $10^{-3}\text{cm}^{-1}$	$\Delta\nu(86-88),$ $10^{-3}\text{cm}^{-1}$	$\Delta\nu(88-90),$ $10^{-3}\text{cm}^{-1}$
Sr I 4607.3	$5s^2 1S_0 - 5s5p 1P_1^0$	+ 8.6	+ 4.2	- 7.6
Sr II 4215.5	$5s^2 S_{1/2} - 5p^2 P_{1/2}^0$	+ 12.6	+ 5.9	- 11.8
Sr II 4077.7	$5s^2 S_{1/2} - 5p^2 P_{1/2}^0$	+ 12.2	+ 5.6	- 11.1

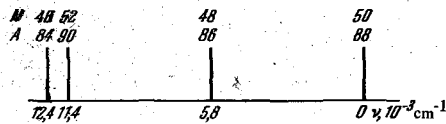


FIG. 1. Isotope structure in the spectrum of Sr II.

In order to extract the volume shift from the experimentally found shift, the authors considered the mass shift in the spectra of the light elements, and concluded that the specific effect for alkali-like  $s \rightarrow p$  transitions lies in the range  $(-0.2 \text{ to } +1.5)\Delta\nu_n$ , while for  $s^2 \rightarrow sp$  transitions it lies within the range  $(-0.3 \text{ to } +0.5)\Delta\nu_n$ . They extracted the volume shift for the two transitions from the total isotope shift on the basis of these considerations. The obtained data, which are given in Table II, show that the component for the isotope  $\text{Sr}^{90}$  occupies the correct position in this case, and that there really is a jump in the isotope shift in the interval  $\Delta\nu(88-90)$ .

The isotope shift in the Sr II lines is completely due to the s electrons, and is referred to the series limit, since the shift in the  $5p^2P$  level is negligibly small. Thus we obtain the experimental value  $\Delta T_S = 18.4 \times 10^{-3} \text{ cm}^{-1}$ . We can get the same value from the data on the isotope shift in the Sr I line. The experimental isotope-shift constant was determined from the found value of  $\Delta T_S$ , and the values of the former for the three intervals are given in Table I.

The isotope shift for 45 lines in the Zr I spectrum has been measured in two studies.<sup>[36,37]</sup> Comparison of the results for a number of lines shows rather good agreement if the experimental errors are taken into account. Table III gives the data of <sup>[37]</sup>, which are characterized by higher reliability and precision. The first column gives the wavelengths of the lines, and then their classifications are given, and then the isotope shifts for the three ranges, with a root-mean-square error of  $\pm 0.2 \times 10^{-3}$ ,  $\pm 0.3 \times 10^{-3}$ , and  $0.5 \times 10^{-3} \text{ cm}^{-1}$ , respectively.

We have estimated the mass shift for the given lines by King's method. Then we isolated the volume com-

Table II. Volume shift in the spectra of Sr

Spectrum	Transition	$\Delta\nu(84-86)$ , $10^{-3} \text{ cm}^{-1}$	$\Delta\nu(86-88)$ , $10^{-3} \text{ cm}^{-1}$	$\Delta\nu(88-90)$ , $10^{-3} \text{ cm}^{-1}$
Sr II	$5s-5p$	from 0 to -3	from 0 to -3	$-18.4 \pm 1.9$
Sr I	$5s^2-5s5p$	from 0 to -3	from 0 to -3	$-12.1 \pm 0.8$
Constant	$6C_{\text{exp}}$	$< 7$	$< 7$	$47 \pm 5$

Table III. Isotope shift in the spectrum of Zr I

$\lambda, \text{ \AA}$	Transition	$\Delta\nu(90-92)$ , $10^{-3} \text{ cm}^{-1}$	$\Delta\nu(92-94)$ , $10^{-3} \text{ cm}^{-1}$	$\Delta\nu(94-96)$ , $10^{-3} \text{ cm}^{-1}$
4035.89	$4d^2 5s^2 a^3 F_4 - 4d^3 5p y^3 G_4^3$	-12.5	-7.0	-3.4
3968.26	$a^3 F_4 - y^3 G_4^3$	-12.7	-5.4	-2.7
3929.53	$a^3 F_3 - y^3 G_3^3$	-12.1	-6.4	-3.1
4633.98	$4d^2 5s^2 a^3 F_3 - 4d^2 5s 5p z^3 G_4^3$	-9.8	-6.3	-3.2
4321.17	$a^3 F_4 - y^5 D_3^3$	-8.7	-6.8	-4.8
4883.60	$4d^2 5s a^5 F_4 - 4d^3 5p y^3 G_3^3$	-8.0	-7.0	-6.4
4805.87	$a^5 F_4 - y^5 G_3^3$	-10.8	-7.4	-5.4
4784.92	$a^5 F_4 - y^3 G_3^3$	-7.4	-7.1	-6.4
4739.48	$a^5 F_3 - y^5 G_3^3$	-10.8	-7.2	-5.4
4687.80	$a^5 F_5 - y^5 G_3^3$	-12.0	-7.2	-5.0
5407.62	$4d^2 5s a^5 F_5 - 4d^2 5s 5p y^5 D_3^3$	-5.3	-6.2	-6.8

Table IV. Mass and volume isotope shifts in the spectrum of Zr I

$\lambda, \text{ \AA}$	Mass shift		Volume shift			Relative shift		
	$M_n$	$M_{sp}$	$\Delta\nu(90-92)$ , $10^{-3} \text{ cm}^{-1}$	$\Delta\nu(92-94)$ , $10^{-3} \text{ cm}^{-1}$	$\Delta\nu(94-96)$ , $10^{-3} \text{ cm}^{-1}$	$\Delta\nu(90-92)$ , $10^{-3} \text{ cm}^{-1}$	$\Delta\nu(92-94)$ , $10^{-3} \text{ cm}^{-1}$	$\Delta\nu(94-96)$ , $10^{-3} \text{ cm}^{-1}$
4035.89	+3.2	+1.4	-17.1	-11.6	-8.0	1.47	1.00	0.69
3968.26	+3.2	+4.1	-20.0	-12.7	-10.0	1.57	1.00	0.79
3929.53	+3.2	+1.8	-17.1	-11.4	-8.1	1.50	1.00	0.71
4633.98	+2.9	-0.6	-12.1	-8.6	-5.5	1.41	1.00	0.64
4321.17	+2.9	-4.7	-6.9	-5.0	-3.0	1.38	1.00	0.60
4883.60	+2.7	-7.7	-3.0	-2.0	-1.4	1.50	1.00	0.70
4805.87	+2.7	-3.4	-10.1	-6.7	-4.7	1.51	1.00	0.70
4784.92	+2.7	-8.5	-1.6	-1.3	-0.6	1.23	1.00	0.46
4739.48	+2.7	-3.2	-10.3	-6.7	-4.9	1.54	1.00	0.73
4687.80	+2.8	-1.3	-13.5	-8.7	-6.5	1.55	1.00	0.75
5407.62	+2.4	-10.4	+2.7	+1.8	+1.2	1.50	1.00	0.67

ponent of the shift and found the relative shift, taking the shift in the interval  $\Delta\nu(92-94)$  to be unity. The results are given in Table IV. The mean relative shift calculated for all the lines is  $\Delta\nu(90-92): \Delta\nu(92-94): \Delta\nu(94-96) = (1.47 \pm 0.03): 1.00: (0.68 \pm 0.03)$ . The obtained data show convincingly that the isotope shift declines in the two intervals following the jump in the interval  $\Delta\nu(90-92)$ .

Table IV shows that, upon extracting the volume shift by King's method, the relative shift in all the lines was found to be about the same. However, the absolute volume shifts in the lines belonging to transitions between levels of the same electronic configurations ( $4d^2 5s^2 - 4d^2 5s 5p$ ,  $4d^3 5s - 4d^3 5p$ ) differ rather strongly, whereas we know that the shifts in a particular interval for similar lines must be identical in the absence of perturbations. If we compare the experimentally-measured isotope shifts (Table III) with the volume shifts (Table IV), we can see that the cited difference in the isotope shift has been greatly magnified upon treatment by King's method. This shows that the estimate of the mass shift (especially in the lines 4321.17, 4883.60, and 4784.92) obtainable by this method is not very exact, in connection with a certain arbitrariness in King's method.

We can use the 4687.80 Å line to determine the shift in the lower level. This line belongs to a transition between unperturbed terms giving good agreement between the measured and theoretical values of the  $g_J$  factor. The electronic configuration of the upper level of the 4687.80-Å line has no s electron. Hence, we can assign all of the volume shift to the level  $4d^2 5s a^3 F_5$ . Further, we can determine from the 5407.62- and 4321.17-Å lines the volume effect for the configurations  $4d^2 5s 5p$  and  $5d^2 5s^2$  with respect to the limit of the  $4d^3 4F$  series. These data are given in Table V for the interval  $\Delta T(90-92)$ . Using these results, we can refine the numerical values of the experimental isotope-

Table V. Volume effect for certain configurations of Zr I (90-92)

Configuration	$\Delta T(90-92)$ , $10^{-3} \text{ cm}^{-1}$
$4d^2 5s$	$13.5 \pm 1.5$
$4d^2 5s 5p$	$16.2 \pm 2.7$
$4d^2 5s^2$	$23.1 \pm 5$

Table VI. Isotope shift in the spectrum of Mo I

$\lambda, \text{ \AA}$	Transition	$\Delta\nu(92-94),$ $10^{-3} \text{ cm}^{-1}$	$\Delta\nu(94-96),$ $10^{-3} \text{ cm}^{-1}$	$\Delta\nu(96-98),$ $10^{-3} \text{ cm}^{-1}$	$\Delta\nu(98-100),$ $10^{-3} \text{ cm}^{-1}$
6030.66	$4d^4 5s^2 a^5 D_4 - 4d^5 5p z^5 P_3$	-25.3	-18.5	-12.0	-26.3
5888.33	$a^5 D_3 - z^5 P_3$	-25.0	-16.6	-12.5	-21.5
5858.27	$a^5 D_3 - z^5 P_3$	-25.0	-18.0	-12.1	-23.3
5791.85	$a^5 D_2 - z^5 P_3$	-24.0	-17.6	-10.8	-25.5
5751.40	$a^5 D_2 - z^5 P_3$	-24.5	-18.5	-11.2	-26.4
5722.73	$a^5 D_2 - z^5 P_3$	-25.6	-18.3	-11.3	23.0
5689.14	$a^5 D_1 - z^5 P_3$	-24.0	-16.5	-11.8	18.9
5650.17	$a^5 D_1 - z^5 P_3$	-24.1	-16.7	-12.0	21.1
5632.43	$a^5 D_0 - z^5 P_3$	-24.8	-17.2	-12.3	21.2
5570.45	$4d^4 5s a^5 S_2 - 4d^5 5p z^5 P_1$	-6.7	-5.3	-3.0	-6.5
5533.05	$a^5 S_2 - z^5 P_1$	-7.2	-5.7	-3.0	-7.2
5506.49	$a^5 S_2 - z^5 P_3$	-8.3	-6.4	-3.6	-7.7
	$4d^4 5s^2 a^5 D - 4d^5 5p z^5 P_0$	-24.7	-17.5	-11.8	-24.9
	$4d^4 5s a^5 S - 4d^5 5p z^5 P_0$	-7.4	-5.8	-3.2	-7.1
	$4d^4 5s^2 a^5 D - 4d^5 5s a^5 S$	-17.3	-11.7	-8.6	-17.8
	$\Delta T_s$	17.3	11.7	8.6	17.8
	Relative shift	1.48 $\pm 0.16$	1.00	0.73 $\pm 0.12$	1.52 $\pm 0.20$
	Experimental isotope-shift constant	41	28	20	43

shift constant  $\beta C_{\text{exp}}$  on the basis of this work. For the intervals  $\Delta\nu(90-92)$ ,  $\Delta\nu(92-94)$ , and  $\Delta\nu(94-96)$ , this constant is found to be  $(37 \pm 3) \times 10^{-3}$ ,  $(25 \pm 3) \times 10^{-3}$ , and  $(17 \pm 2) \times 10^{-3} \text{ cm}^{-1}$ , respectively.

Yu. P. Dontsov<sup>[38]</sup> has obtained very complete results on the isotope shift in the spectrum of molybdenum. Upon comparing with the data of<sup>[40]</sup>, we see that the existing discrepancies involve the fact that Yu. P. Dontsov took into account the overlap of the contours of the impurity isotopes. Table VI gives the fundamental experimental results without account of overlap for 12 lines belonging to two types of transitions. The same table gives the mean isotope shift for each transition. If we subtract these shifts, we obtain the difference between the shifts in the levels  $4d^4 5s^2 a^5 D$  and  $4d^5 5s a^5 S$ . In extracting the volume shift from this quantity, it would seem that we would have to take into account only the normal mass shift, since we can consider the specific shift for the given levels to be zero.<sup>[2]</sup> The normal mass shifts for the levels  $4d^4 5s^2 a^5 D$  and  $4d^5 5s a^5 S$  are  $1.4 \times 10^{-3}$  and  $1.3 \times 10^{-3} \text{ cm}^{-1}$ , respectively. Hence,  $\Delta T_N = 0.1 \times 10^{-3} \text{ cm}^{-1}$ , which amounts to about 1% of the difference between the overall shifts. Hence, we can also neglect the normal shift. As we know, the isotope shift for  $s^2$  electrons is 1.6 times as large as that for one  $s$  electron. Hence, taking into account the shielding of a  $d$  electron (0.8), we can easily obtain the volume shift  $\Delta T_S$  due to one  $s$  electron. All these data are given in Table VI, which also gives the relative shift. The latter agrees well with the relative shift in the zirconium spectrum, if we consider isotopes having the same neutron number for the two elements. The experimental isotope-shift constants are given at the bottom of Table VI. We found these from the shift obtained for an  $s$  electron by comparison with the isotope-shift constant for  $\text{Mo}^{95}$  and  $\text{Mo}^{97}$ .\*

Among the other intermediate elements in whose spectra the isotope shift has been studied, we should mention ruthenium,<sup>[41,42]</sup> palladium,<sup>[43]</sup> cadmium,<sup>[44-49]</sup> tin,<sup>[50,51]</sup> tellurium,<sup>[52]</sup> and barium.<sup>[53-55]</sup> Most of the studies were performed with enriched isotopes. This

permitted fixing the positions of the components belonging to the rare isotopes in all the spectra. One defect in all the studies is the small bulk of experimental data, since the investigators restricted themselves to studying only one spectral line of the element. In addition, in extracting the volume isotope shift, they often didn't take into account the specific mass shift, which can attain values comparable with the experimentally-observed isotope shift in this region of nuclei. The latter circumstance casts doubt on the values obtained by some authors for the relative isotope shifts and experimental constants, which are determined by the volume component of the isotope shift.

Our discussion of the isotope shift in the intermediate elements shows that reliable experimental results have been obtained for a number of elements using enriched isotopes, in spite of difficulties of measurement. These results are given in collected form below in Table XII.

## 2. The Isotope Shift in the Spectra of Isotopes of Heavy Elements of Neutron Numbers 82-126

It is appropriate to start the discussion of the isotope shift in the spectra of the heavy elements with the group of rare-earth elements, where a number of features involving nuclear deformation and filling of neutron shells have been revealed in recent years. As we have mentioned, anomalous shifts in lines were discovered long ago in the spectra of Ce, Nd, Sm, and Gd. These indicated non-equidistance in the shifts of the atomic energy levels. More thorough investigations of the isotope shift in the spectra of the rare-earth elements have been made in recent years, using isolated and enriched isotopes. They have made it possible to study non-equidistance throughout the range of nuclei of neutron numbers from  $N = 78$  to  $N = 106$ .

L. A. Korostyleva<sup>[56]</sup> has remeasured the isotope shift in the spectrum of cerium. The previous studies had established a jump in the isotope shift in the interval between the  $\text{Ce}^{140}$  and  $\text{Ce}^{142}$  components ( $N = 82$  and  $84$ ), where an extra pair of neutrons is added to the completely filled  $1h_{11/2}$  shell. However, the shift between the components of the cerium isotopes had been measured very crudely.<sup>[1]</sup> Table VII gives the more reliable results<sup>[56]</sup> on four Ce II lines. The shift in the intervals  $\Delta\nu(136-138)$  and  $\Delta\nu(138-140)$  proved to be about the same. In comparison, the shift in the interval  $\Delta\nu(140-142)$  was 3.82 times as great. The measured value of the isotope shift has been taken to be the volume shift in the spectrum of cerium, as for all the subsequent elements. The normal mass shift here is actually small, and amounts to about  $0.001 \text{ cm}^{-1}$  (for 5000 Å). However, nothing is known

Table VII. Isotope shift in the spectrum of Ce

$\lambda, \text{ \AA}$	$\Delta\nu(136-138) =$ $=\Delta\nu(138-140),$ $10^{-3} \text{ cm}^{-1}$	$\Delta\nu(140-142),$ $10^{-3} \text{ cm}^{-1}$	$\frac{\Delta\nu(140-142)}{\Delta\nu(138-140)}$
4471.24	14±2	52±1	3.71±0.60
4460.21	14±2	54±1	3.86±0.63
4391.66	15±1	57±1	3.80±0.31
4289.94	10±1.5	39±1	3.90±0.67
	Mean relative shift		3.82±0.28

\*A. Stuedel, Z. Physik 132, 429 (1952).

about the size of the specific shift. It is not ruled out that it could be several times as large as the normal shift. Then the value of the volume relative shift in the spectrum of cerium would be considerably altered. It has been shown in<sup>[9]</sup> that the experimental results of Table VII agree well with the theory of the isotope shift taking the influence of the shell structure of the nucleus into account.

Anomalous isotope shifts in the spectra of neodymium and samarium have been found using natural samples. For gadolinium, they used enriched samples, since the concentration of  $Gd^{152}$  and  $Gd^{154}$  in natural samples is insufficient for spectroscopic detection. An anomalously large shift occurs in the spectra of the three cited elements in the interval between the isotopes having  $N = 88$  and  $N = 90$ , i.e., at the transition from spherically-symmetric to deformed nuclei. New measurements have been made in recent years in order to determine more reliably the shifts between all of the even-even isotopes of these elements.

Following the well-known study of Klinkenberg, the isotope shift in the spectrum of neodymium has been studied by a number of authors. Nöldeke and Steudel have obtained especially detailed data in the Nd I<sup>[57]</sup> and Nd II<sup>[58]</sup> spectra. They determined the experimental isotope-shift constants from the Nd II lines. For the intervals  $\Delta\nu(142-144)$ ,  $\Delta\nu(144-146)$ ,  $\Delta\nu(146-148)$ , and  $\Delta\nu(148-150)$ , these proved to be:  $187 \pm 35$ ,  $169 \pm 35$ ,  $171 \pm 35$ , and  $260 \pm 50$ , respectively, in units of  $10^{-3} \text{ cm}^{-1}$ . In<sup>[61]</sup>, the isotope shift in the interval  $\Delta\nu(142-144)$  was measured for a large number of lines; the obtained results were used to classify the spectrum of Nd I. The cited studies were performed with natural neodymium samples, which contained the even-odd isotopes. Hence the correctness of measurement of the intervals of the isotope structure remained somewhat uncertain. This was confirmed by the rather great deviations in the relative positions of the components of certain isotopes in a series of Nd I and Nd II lines.

Yu. P. Dontsov, V. A. Morozov, and A. R. Striganov<sup>[60]</sup> have measured the isotope shift in the spectrum of neodymium, using enriched samples in which the concentration of the even-odd isotopes did not exceed 1%. The isotope structure was measured on 16 lines, three of them belonging to Nd II. All the arc lines showed a negative shift. It turned out that the difference in the relative shift in the Nd I and Nd II lines found in<sup>[57, 58]</sup> is not observed when enriched samples are used. Figure 2 shows the mean relative shift for all the lines, and gives the root-mean-square error. The interval  $\Delta\nu(142-144)$  is taken as unity. Each component in the isotope structure is labeled with its corresponding mass number and the number of neutrons. The obtained data show that the spectrum of neodymium has a jump in the isotope shift in the interval  $\Delta\nu(142-144)$ , just like that of cerium. It is due to

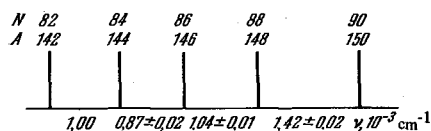


FIG. 2. Relative shifts in the spectrum of Nd.

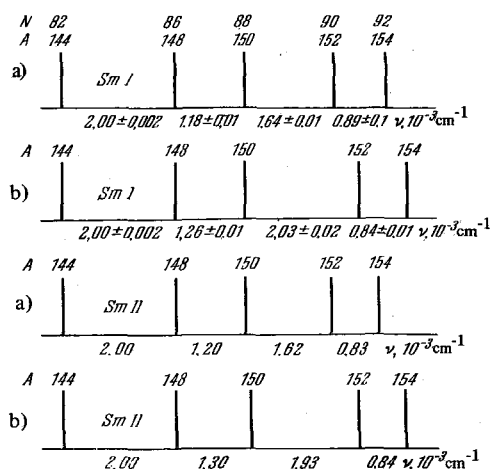


FIG. 3. Relative shifts in the spectra of Sm I and Sm II: a) lines showing negative shifts; b) lines showing positive shifts.

the filling of the new neutron shell  $2f_{7/2}$ . The shift diminishes in the second interval, and then increases again, and attains an anomalously large value in the interval  $\Delta\nu(148-150)$ .

Recently Gerstenkorn and his associates<sup>[62]</sup> have studied anew the isotope shift in the spectrum of neodymium, using a Fabry-Perot interferometer with photoelectric recording, with enriched isotopes. They measured three lines with negative and three lines with positive shifts. The obtained results will be discussed below.

V. V. Eliseev, V. A. Katulin, N. A. Kulazhenkova, and A. R. Striganov<sup>[63, 66]</sup> have studied the isotope shift in the spectrum of samarium, using enriched isotopes. The shift between the even-even isotopes was measured on 59 Sm I lines and eight Sm II lines. Both spectra contain lines having negative and positive shifts. Figure 3 shows the mean relative shifts for the four groups of lines individually. In addition to the anomalously large shift between the components due to the isotopes with  $N = 88$  and  $N = 90$ , the obtained results indicate that other intervals distinctly show non-equidistance outside the limits of experimental error. Attention is called to the fact that the trend in the relative shift differs rather strongly for negatively- and positively-shifted lines in both the Sm I and the Sm II spectra. The relative displacement of the positively-shifted Sm I lines in the interval  $\Delta\nu(148-150)$  is 0.08 greater on the average than for the negatively-shifted lines. This difference is eight times as great as the random errors of measurement, and does not involve superposition of the hyperfine structure of the odd isotopes  $Sm^{147}$  and  $Sm^{149}$ , since no extra components appear on the spectrograms. In the interval  $\Delta\nu(150-152)$  having a large quadrupole effect, the difference in the relative shift between the positively- and negatively-shifted lines becomes as much as 0.39 on the average. This is 20 times the error of measurement. The difference in the relative shift in the interval  $\Delta\nu(150-152)$  for Sm II lines having negative and positive shifts is 0.31 on the average.

In the Sm I spectrum, all the lines having negative shifts belong to transitions of the type  $4f^6s^2-4f^6s6p$ , while the lines having positive shifts belong to  $4f^6s^2-$

$4f^5 5d6s^2$  transitions. Since the lower electron configuration is the same in these transitions, the positive shift in the lines arises from a large isotope shift in the upper levels belonging to the electron configuration  $4f^5 5d6s^2$ . The large isotope shift in these levels can be explained by lessened shielding of the two  $s$  electrons upon transition of one  $4f$  electron to the  $5d$  shell. This increases the charge density of the electrons at the site of the nucleus. In the Sm II spectrum, the negatively-shifted lines belong to  $4f^6 6s-4f^6 6p$  transitions, while the positively-shifted lines belong to  $4f^6 6s-4f^5 5d6s$  transitions. Here also, the upper levels having the  $4f^5 5d6s$  configuration undergo a large isotope shift due to the lessened shielding of the  $s$  electron.

The experimental data on samarium show<sup>[64]</sup> that the upper energy levels of electronic configuration  $4f^6 6s6p$  belonging to negatively-shifted lines lie in the region  $16112-22632 \text{ cm}^{-1}$ . Some of these levels are interspersed with levels of the electronic configuration  $4f^5 5d6s^2$  occupying the region  $18075-23996 \text{ cm}^{-1}$ . Hence we can assume that the upper closely-spaced levels perturb one another. The size of the isotope shift for such levels will depend on the admixture of the perturbing level. If the given level belongs to the electronic configuration  $4f^6 6s6p$ , then a perturbation by a  $4f^5 5d6s^2$  level must increase its isotope shift. Consequently, the shift in the line will diminish. Such a phenomenon is observed in the lines having negative shifts. Here the shift in all the lines in the long-wavelength region is about the same, but it starts to decline with the  $5157.04\text{-}\text{\AA}$  line.<sup>[63]</sup> Perturbation of levels of electronic configuration  $4f^5 5d6s^2$  by  $4f^6 6s6p$  levels must decrease their isotope shifts. The isotope shift will differ for different levels, since the fraction of the wave function arising from the perturbing electronic configuration will differ. This is observed in the lines having positive shifts, where the isotope shift assumes differing values.<sup>[63]</sup> This means that the long-wavelength lines having negative shifts apparently belong to unperturbed transitions, as do the lines having maximum positive shifts. It is precisely these lines that have been used to determine the variation of the wave function of the electronic configurations  $4f^6$  and  $4f^7$  from the isotope shift.\*

The data on the isotope shift in the Sm I and Sm II spectra show that the extra isotope shift in the interval  $\Delta\nu$  (150-152) in going from a spherical to a deformed nucleus turns out to be considerably larger when the nucleus interacts with  $4f^5 5d6s^2$  and  $4f^5 5d6s$  shells. One might suppose that some new effect is being manifested here, which leads to an additional shift of levels of  $4f^5 5d6s^2$  and  $4f^5 5d6s$  configurations when they interact with the deformed nucleus. Then the difference in energy of the  $s$  electrons for two isotopes of the same element could not be considered to consist of two factors, one determined by the electron shell and the other by the nuclear parameters.

King<sup>[65]</sup> explains the observed difference in the relative isotope shift in terms of a large mass effect. For samarium, we must assume the existence of a mass effect amounting to as much as  $0.010 \text{ cm}^{-1}$  for

certain lines. The normal-mass shift for the studied lines of samarium does not exceed  $0.001 \text{ cm}^{-1}$ . Hence, the specific mass effect must be about ten times as large as the normal mass shift. Apparently, this quantity is very large in the samarium spectrum, if we consider that the specific shift is inversely proportional to  $A^2$ , just as the normal mass shift is. To solve this problem requires that we estimate by theory the size of the specific shift in the upper levels for the two types of transitions in atoms of neutral and singly-ionized samarium.

The results on the isotope shift in the spectrum of samarium obtained in<sup>[63]</sup> have been thoroughly checked by Hansen, Stuedel, and Walther.<sup>[67]</sup> Using a recording Fabry-Perot spectrometer and highly enriched samarium isotopes, they measured two lines having positive shifts. It turned out that the results of the measurements agreed well. Upon applying King's method, the authors of<sup>[67]</sup> state that the discrepancy in the trend of the relative isotope shift between the negatively- and positively-shifted samarium lines arises from a large specific effect.

On the same level, Gerstenkorn and his associates<sup>[62]</sup> have studied the isotope shift in the spectrum of neodymium. The obtained results distinctly showed the same phenomenon in the relative shift as was observed in the spectrum of samarium. Table VIII gives the results of the highly-accurate measurement of the isotope shift of two neodymium lines.

The difference in the relative shift of the positively- and negatively-shifted lines in the interval  $\Delta\nu$  (148-150) is as much as 0.49, which amounts to 35%. The lines with positive shifts belong to the  $4f^4 6s^2-4f^3 5d6s^2$  transition, while the lines with negative shifts belong to  $4f^4 6s^2-4f^4 5d6p$  or  $4f^4 5d6s-4f^4 5d6p$  transitions. Thus, the same situation occurs here as in the samarium spectrum. That is, a large shift occurs in the upper level of electronic configuration  $4f^3 5d6s^2$ , owing to lessened shielding of the  $s$  electron upon transition of a  $4f$  electron to the  $5d$  shell. It was shown in<sup>[62]</sup> that the found difference in the relative isotope shift of positively- and negatively-shifted lines can be explained by the specific shift, which for one of the lines can become at least ten times the normal mass shift.

We should note that King's method<sup>[42,65]</sup> is apparently suitable for estimating the specific mass effect in the isotope shift of lines, provided that we know that the difference in the relative shift is specifically due to this effect. One cannot make this assumption a priori in the spectra of the heavy elements. Hence, the statement that there is a large specific effect in the spectra

Table VIII. Isotope shift in two Nd I lines

$\lambda, \text{\AA}$		$\Delta\nu(142-144)$	$\Delta\nu(144-146)$	$\Delta\nu(146-148)$	$\Delta\nu(148-150)$
5291, 67	Shift, $10^3 \text{ cm}^{-1}$ Relative shift	$-74.1 \pm 0.5$ $1.00 \pm 0.007$	$-69.7 \pm 0.5$ $0.94 \pm 0.007$	$-75.7 \pm 0.5$ $1.02 \pm 0.007$	$-104.9 \pm 0.5$ $1.42 \pm 0.005$
5675, 97	Shift, $10^3 \text{ cm}^{-1}$ Relative shift	$+20.4 \pm 0.4$ $1.00 \pm 0.02$	$+19.0 \pm 0.3$ $0.93 \pm 0.02$	$22.2 \pm 0.5$ $1.09 \pm 0.02$	$+39.0 \pm 0.5$ $1.91 \pm 0.01$

\*S. Hüfner, P. Kienle, D. Quitmann, and P. Brix, Z. Physik 187, 67 (1965).



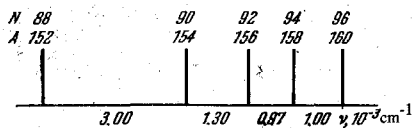


FIG. 4. Relative shifts in the spectrum of Gd.

of neodymium and samarium remains a hypothesis. We might assume the existence of a new phenomenon which would also produce an additional shift, and would be described by the same type of formulas. The question can be answered only by a theoretical treatment of this problem.

In 1956, Speck,<sup>[69]</sup> followed by Kopfermann and his associates,<sup>[70]</sup> was able to study the complete isotope structure in the spectrum of gadolinium using enriched isotopes, including the components of the isotopes  $Gd^{152}$  and  $Gd^{154}$ . Figure 4 shows the mean relative shift according to<sup>[70]</sup>. As we might expect, an anomalously large shift occurs in the interval  $\Delta\nu(152-154)$ . It is pertinent to note that the enriched samples used contained concentrations of the even-odd isotopes  $Gd^{155}$  and  $Gd^{157}$  as much as 8% and 22%, respectively. Hence, hyperfine structure could introduce considerable errors into the measurements. In<sup>[68]</sup>, the isotope shift was measured in the Gd II spectrum, and the experimental isotope shift constant was determined:  $\beta C_{exp}(158-160) = (125 \pm 20) \times 10^{-3} \text{ cm}^{-1}$ .

The isotope shift in the spectrum of dysprosium was first studied on the natural mixture of isotopes,<sup>[71]</sup> and then on enriched isotopes.<sup>[72,73]</sup> In the latter work, 30 spectral lines of Dy I were studied. For 17 of them, the shifts between the components were measured for all five even-even isotopes, including the three rare isotopes  $Dy^{160}$ ,  $Dy^{158}$ , and  $Dy^{156}$ . The spectrograms were later subjected to an additional study.<sup>[82]</sup> Figure 5 shows the obtained results on the relative isotope shift in the spectrum of dysprosium. Just as in the spectra of samarium and neodymium, there is a difference in the relative shifts in the lines shifted in different directions. This difference proves to be greatest in the interval showing the greatest deviation from equidistance in the positions of the components of the isotopes.

Two studies have recently been concerned with the isotope shift in the spectrum of erbium.<sup>[74,75]</sup> They were performed on enriched isotopes. This made it possible to measure the intervals between the components for all the even-even isotopes. The most complete study was conducted by D. A. Volkov and A. F. Golovin,<sup>[74]</sup> who measured 19 spectral lines having negative shifts, and 10 lines having positive shifts. The trend in the relative shift is shown in Fig. 6. The root-

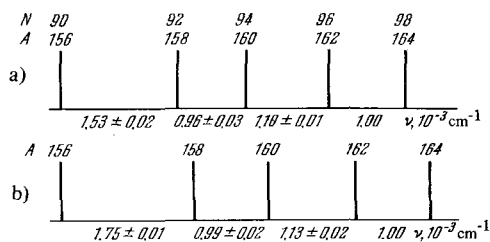


FIG. 5. Relative shifts in the spectrum of Dy: a) lines showing negative shifts; b) lines showing positive shifts.

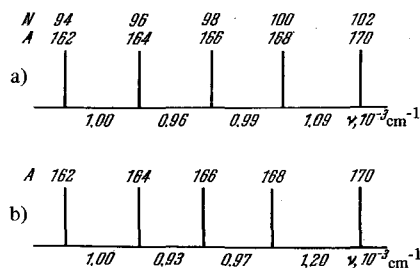


FIG. 6. Relative shifts in the spectrum of Er: a) lines showing negative shifts; b) lines showing positive shifts.

mean-square error of the cited values amounts to 1%. The lines of erbium shifted in different directions show an appreciable difference in the relative shifts.

Krebs and Nelkowski<sup>[76]</sup> have studied the isotope effect in the spectrum of ytterbium, using the natural mixture of isotopes. The authors found the experimental isotope-shift constant, which proved for the most abundant pair of isotopes  $Yb^{172}$  and  $Yb^{174}$  to be  $\beta C_{exp} = (99 \pm 8) \times 10^{-3} \text{ cm}^{-1}$ . The structure of the arc lines of ytterbium at 5556.48 and 3987.99 Å was studied in<sup>[77,78]</sup> using enriched isotopes. The most complete data on the isotope shift in the spectrum of ytterbium were obtained in<sup>[79]</sup>. The highly-enriched samples (the content of the rare isotope  $Yb^{168}$  was as much as 28.7%) made it possible to measure the shifts between the components of all the even-even isotopes for ten spectral lines of Yb I and Yb II. The trend of the relative shift for negatively- and positively-shifted lines proved to be the same within the limits of accuracy of the measurements. The mean value of the relative shift found from all the lines was  $\Delta\nu(168-170) : \Delta\nu(170-172) : \Delta\nu(172-174) : \Delta\nu(174-176) = (1.48 \pm 0.006) : (1.37 \pm 0.004) : (1.10 \pm 0.011) : 1.00$ . These results agree well with those of the previous study.<sup>[77]</sup>

The isotope shift in the spectrum of hafnium has been investigated in two studies using enriched isotopes.<sup>[81,82]</sup> In the former study, the shift was measured for 16 spectral lines of Hf I, and the experimental isotope-shift constant was estimated:  $\beta C_{exp}(178-180) = (104 \pm 23) \times 10^{-3} \text{ cm}^{-1}$ . However, the author concedes preference to the value  $(114 \pm 13) \times 10^{-3} \text{ cm}^{-1}$  obtained in<sup>[80]</sup> from Hf II lines belonging to three different electronic transitions. In the latter study<sup>[82]</sup>, the isotope shift was subsequently measured for 20 spectral lines of Hf I. The position of the component belonging to the rare isotope  $Hf^{174}$  was reliably determined for 15 of these lines. All the lines are shifted in the negative direction. The relative isotope shift was  $\Delta\nu(174-176) : \Delta\nu(176-178) : \Delta\nu(178-180) = (0.89 \pm 0.01) : (0.85 \pm 0.01) : 1.00$ . This agrees with<sup>[81]</sup> within the limits of error. A defect in both studies is the use of samples that were not highly enriched in the rare isotope  $Hf^{174}$  (8–10%).

The isotope shift in the spectrum of tungsten has been studied in recent years, both on the natural mixture<sup>[83]</sup> and on enriched isotopes.<sup>[84-86]</sup> The latter studies are characterized by insufficient enrichment of the samples in the rare isotope  $W^{180}$  and the small number of studied spectral lines. The measurements of the relative isotope shifts by different authors are not in complete accord. We can take the results of



**Table IX.** Relative shifts and experimental isotope-shift constants in the spectrum of W I

	$\Delta\nu(180-182),$ $10^{-3} \text{ cm}^{-1}$	$\Delta\nu(182-184),$ $10^{-3} \text{ cm}^{-1}$	$\Delta\nu(184-186),$ $10^{-3} \text{ cm}^{-1}$
Relative shift	$0.79 \pm 0.06$	$1.13 \pm 0.02$	1.00
$\beta C_{\text{exp}}$	$92 \pm 24$	$133 \pm 25$	$117 \pm 20$

Blaise and Gluck<sup>[86]</sup> to be the most reliable. The values that they found for the relative shifts and experimental constants are given in Table IX.

The isotope shift in the spectrum of osmium has been studied by a number of authors.<sup>[87-91]</sup> Studies<sup>[88-90]</sup> were performed on enriched isotopes, with the enrichment in the rare isotope Os<sup>184</sup> amounting to 2.25%. In all, they studied six spectral lines, and noted the Os<sup>184</sup> component for three lines. The relative isotope shifts found in<sup>[88,90]</sup> differ somewhat, and are given in Table X. Gluck et al.<sup>[91]</sup> have used the isotope shift to classify the spectrum of osmium and to calculate the experimental isotope-shift constant:

$$\beta C_{\text{exp}}(190-192) = (114 \pm 14) \times 10^{-3} \text{ cm}^{-1}.$$

Many authors<sup>[92-98]</sup> have measured the isotope shift in the spectrum of mercury. Among these, we should mention Blaise,<sup>[96]</sup> who used enriched samples, and measured the complete isotope structure of the Hg I line at 2536.5 Å, including the component due to the rare isotope Hg<sup>196</sup>. Schweitzer<sup>[98]</sup> has made the most accurate measurements of the isotope shift in the resonance line of mercury. Figure 7 shows the relative isotope shift in the 2536.5 Å line according to<sup>[96]</sup>. The experimental isotope-shift constant has been obtained<sup>[97]</sup> by measuring the isotope shift of the forbidden Hg II line at 2814.93 Å ( $5d^{10}6s^2S_{1/2} - 5d^96s^2D_{5/2}$ ). The constant for the individual intervals between the even-even isotopes beginning with  $\Delta\nu(196-198)$  proved to be  $\beta C_{\text{exp}} = (174 \pm 40), (212 \pm 30), (235 \pm 30),$  and  $(235 \pm 37) \times 10^{-3} \text{ cm}^{-1}$ .

The isotope shift in the spectrum of lead has been measured using enriched samples for lines of the singly-ionized atom.<sup>[100]</sup> The relative isotope shift between the components belonging to the even-even isotopes proved to be  $\Delta\nu(204-206)/\Delta\nu(206-208) = 0.908 \pm 0.008$ . This agrees well with the value determined earlier with natural samples.

In closing the discussion of the isotope shift in this group of elements, we note that extensive and more precise data have been obtained in the last ten years. They have revealed the relation of the isotope shifts in the atomic levels to the shell structure and deformations of the nuclei. Deformation parameters have been determined from the isotope shifts for many nuclei. Intrinsic quadrupole moments and probabilities of electric quadrupole transitions from the ground state of the nucleus to the first excited state have been cal-

culated. Experimental isotope-shift constants have been recalculated from the refined values of the isotope shifts. All these data are given below.

### 3. The Isotope Shift in the Spectra of the Actinide Elements

The fundamental topic in studying the isotope effect in the spectra of the actinide elements has been the use of the isotope shifts to classify spectra. Just as for hydrogen and helium, the isotope shift in these spectra attains considerable values, and can be measured by large diffraction spectrographs having a dispersion of 1 Å/mm. For this reason, extensive data have been obtained for a number of elements on the shifts for a large number of spectral lines.

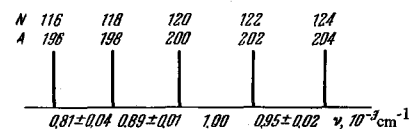
In the spectrum of thorium, the isotope shift has been measured for many lines in the visible and ultraviolet between the natural isotope Th<sup>232</sup> and the isotopes Th<sup>230</sup> and Th<sup>229</sup> obtained from radioactive decay of uranium. The ratio of shifts  $\Delta\nu(232-229)/\Delta\nu(232-230)$  proved to be about 1.7.<sup>[103]</sup> This shows that, in the thorium spectrum, the well-known extra shift of the component for the even-odd isotope Th<sup>229</sup> occurs in the direction toward isotopes of lower mass number. A relation was shown in<sup>[102]</sup> between the size of the isotope shift in the Th II lines and the electronic configurations of the levels.

Detailed data have been obtained for the uranium spectrum on the isotope shift between the components belonging to the isotopes U<sup>238</sup> and U<sup>235</sup>.<sup>[104-109]</sup> The reason for performing these studies was the availability of enriched samples of U<sup>235</sup> and the attempt to use the isotope shifts to classify the many-lined spectrum of uranium. By analyzing the measured shifts, they could establish that 226 lines belonged to the U II spectrum, and find the electronic configurations for 159 of the U II lines.<sup>[106,108]</sup> For the U II line at 4244.37 Å, which shows a large isotope shift, the complete isotope structure was measured using the uranium isotopes of mass numbers 238, 236, 235, 234, and 233.<sup>[105,106]</sup> This structure is shown in Fig. 8. Here the extra shift in the components due to the even-odd isotopes is distinctly manifested in the usual direction, with the ratio  $\Delta\nu(238-235)/\Delta\nu(238-236) = 1.70$ .

Gerstenkorn and his associates<sup>[110]</sup> have obtained the most accurate data on the isotope shift in the spectrum of uranium, using a photoelectric spectrometer with a Fabry-Perot interferometer. They measured the shifts  $\Delta\nu(238-233)$  and  $\Delta\nu(238-235)$  for 20 U I lines with a root-mean-square error of only 1%. Among these, 13 lines showed negative shifts, and belonged to the  $5f^36d7s^2 - 5f^36d7s7p$  transition, while the seven other lines showed positive shifts, and belonged to  $5f^36d7s^2 - 5f^37s^27p$  and  $5f^36d^27s - 5f^36d7s7p$  transitions. The obtained data show that the relative isotope shift  $\Delta\nu(238-233)/\Delta\nu(238-235)$  remains the same for

**Table X.** Relative isotope shifts in the spectrum of Os

Reference	$\Delta\nu(184-186),$ $10^{-3} \text{ cm}^{-1}$	$\Delta\nu(186-188),$ $10^{-3} \text{ cm}^{-1}$	$\Delta\nu(188-190),$ $10^{-3} \text{ cm}^{-1}$	$\Delta\nu(190-192),$ $10^{-3} \text{ cm}^{-1}$
[89]	$1.28 \pm 0.06$	$1.37 \pm 0.04$	$1.15 \pm 0.02$	1.00
[90]	$1.31 \pm 0.04$	$1.29 \pm 0.02$	$1.42 \pm 0.02$	1.00

**FIG. 7.** Relative shifts in the spectrum of Hg I.

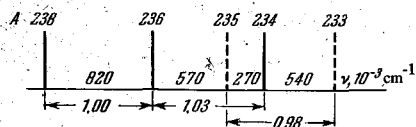


FIG. 8. Isotope structure of the U II line at 4244.37 Å.

negatively- and positively-shifted lines ( $1.554 \pm 0.015$  and  $1.537 \pm 0.015$ ). Practically the same relative shift ( $1.57 \pm 0.02$ ) occurs also in the well-studied U II line at 4244.37 Å ( $5f^{37}s^2-5f^36d7p$ ), which has a negative shift.

On this basis, the authors of<sup>[110]</sup> concluded that for uranium the relative shift remains the same within the limits of error for four different transitions, and that, in distinction from the results on the spectrum of samarium,<sup>[63,66]</sup> no difference is observed in the relative shift of negatively- and positively-shifted uranium lines. However, we should note in this regard that the cited difference was found for samarium in the interval  $\Delta\nu$  (150–152) where the large change in the nuclear deformation between  $\text{Sm}^{150}$  and  $\text{Sm}^{152}$  occurs, and a jump occurs in the isotope shift. For uranium, the variation in the nuclear deformation from isotope to isotope is very small. Hence, the difference in the relative shift (Fig. 8) does not exceed the errors of measurement of the isotope shifts.

In addition, as was correctly noted in<sup>[110]</sup>, there were no lines among those studied that belonged to a transition of a 5f electron to the 6d state. Thus, one could hardly hope to find in the uranium spectrum a difference such as was found in the Sm I and Sm II spectra.

The isotope shift in the spectrum of plutonium was first measured by American and Russian investigators. In order to obtain the spectra of two isotopic samples of plutonium ( $\text{Pu}^{238} + \text{Pu}^{242}$ , and  $\text{Pu}^{239} + \text{Pu}^{240}$ ) in the ultraviolet, Conway and Fred<sup>[111]</sup> used a nine-meter diffraction spectrograph of dispersion 0.92 Å/mm and a spark light source. The complete isotope structure was obtained for four lines (3958.79; 3972.06; 3985.37; and 4021.41 Å). The cited structure for one of these lines is shown in Fig. 9, in which the position of the center of gravity of the component due to the even-odd isotope is given along with the components for the even-even isotopes.

A hollow cathode was used in<sup>[112]</sup> to excite the spectrum, and a three-prism spectrograph combined with a Fabry-Perot interferometer was used for measuring it. The high resolving power of this apparatus made it possible to find not only the isotope shift, but also the hyperfine structure of the even-odd isotope in the spectra of the two-isotope plutonium sample  $\text{Pu}^{239} + \text{Pu}^{240}$ . The isotope shift was measured for 19 lines in the 4100–6500 Å region. A doublet hyperfine structure was found for six of these and its width measured. The authors showed that they could predict the elec-

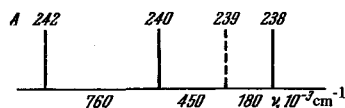


FIG. 9. Isotope structure of the Pu line at 4021.41 Å.

tronic configurations for the levels of many transitions by studying the hyperfine and isotope structure in the spectrum of plutonium.

Further studies of the isotope shift and hyperfine structure in the spectrum of plutonium have basically amounted to using these effects to classify lines and to find the atomic energy levels and their electronic configurations. It was shown in<sup>[113–118]</sup> that one can determine the inner quantum numbers  $J$  for the total angular momentum of the electron cloud of the atom from the relation of intensities of the components of the doublet hyperfine structure of  $\text{Pu}^{239}$ . The hyperfine splitting and isotope shift in the lines of the isotopes  $\text{Pu}^{239}$  and  $\text{Pu}^{240}$  have been measured in detail in a number of studies, and they have been used successfully to identify Pu I and Pu II lines, and also to establish the electronic configurations of the levels of many transitions.<sup>[116–120]</sup>

Conway and McLaughlin<sup>[121]</sup> were the first to measure the isotope shift between the isotopes  $\text{Am}^{241}$  and  $\text{Am}^{243}$  in the spectrum of americium for 28 lines in the 2800–4900 Å region. They used a direct-current arc to excite the spectrum, and measured it with a nine-meter diffraction spectrograph. Since the lines of the studied isotopes of americium show hyperfine structure, the isotope shift was measured between the first (more intense) components of this structure. The maximum  $\Delta\nu$  (241–243) shifts amounted to as much as  $0.80 \text{ cm}^{-1}$ . A more detailed study of the hyperfine and isotope structure was conducted in<sup>[122]</sup>. The authors used the obtained data for a preliminary classification of the spectrum of americium.

The last actinide element in whose spectrum an isotope shift has been found is curium. Conway and McLaughlin<sup>[123]</sup> were able to photograph the spectrum of a sample of the isotopes  $\text{Cm}^{242}$  and  $\text{Cm}^{244}$  in the 3050–5250 Å region with a nine-meter diffraction spectrograph. They found 153 lines in the spectrum having an appreciable isotope shift. They observed both negative and positive shifts. For some lines the shift was as much as  $1.3 \text{ cm}^{-1}$ .

In discussing the isotope shift between the components due to even-even isotopes in the spectra of the actinide elements, we see that no clearly-marked jumps in the isotope shifts have yet been discovered in this region of nuclei. However, one cannot state categorically that jumps in the isotope shifts in the spectra of the actinide elements do not exist. As we know, jumps among the even-even isotopes can be found from the variation in the shift in the spectrum of a given element when the nucleus becomes reorganized through addition of paired neutrons, the charge remaining the same. In the spectra of thorium and curium, the isotope shifts have thus far been studied over one interval each: for Th,  $\Delta\nu$  (230–232); and for Cm,  $\Delta\nu$  (242–244). We do not yet know the shifts in the subsequent intervals upon addition of the corresponding next pairs of neutrons. Hence, we can draw no conclusions at present on the variation of the isotope shift in the spectra of thorium and curium. In the spectra of uranium and plutonium, the isotope shifts have been studied for each element in only two intervals (cf. Figs. 8 and 9). Comparison of these shifts shows that in the uranium spectrum the shifts remain the same in both intervals

within the limits of error of the measurements. In the spectrum of plutonium, the isotope shifts are not equidistant, since the shift in one interval is 16% greater than in the other. We should note that the cited shifts in the spectra of uranium and plutonium were studied with diffraction spectrographs, and hence the root-mean-square error of the measurements was as much as  $\pm 5\%$ . The obtained data given in Figs. 8 and 9 come from measuring only one spectral line per element. Thus we see that the studies of the isotope shift in the spectra of the actinide elements must be broadened. For each element, one must include a larger number of radioactive isotopes, and make the measurements with a Fabry-Perot interferometer on a large number of spectral lines.

## II. THEORY OF THE ISOTOPE SHIFT IN THE SPECTRA OF THE HEAVY ELEMENTS

The original theory of the isotope shift in the spectra of the heavy elements was based on a model of the nucleus as a drop having finite dimensions. The energy of an optical electron moving in the field of the nuclear charge depends on the radius of the nucleus over whose volume the charge is distributed. In going from one isotope to another, the nuclear charge undergoes redistribution as neutrons are added. For spherical nuclei having radii varying according to an  $A^{1/3}$  law, the energy difference of the optical electrons of the even-even isotopes of a given element is constant, and the isotope shift of the levels must be equidistant. Deviations from equidistance observed in the spectra of many elements are explained by non-uniform variation of the nuclear radii as neutrons are added. This can arise from the peculiarities of the filling of the neutron shells in the nuclei (at  $N = 50, 82,$  and  $126$ ) or by variation in the static deformation of the nuclei in the region  $88 < N < 116$ , or by zero-point vibrations of the surface of the nuclei with respect to an equilibrium spherical form for vibrational nuclei. Qualitatively, this theory explains fairly well all the phenomena of the isotope shift. However, it cannot give quantitative agreement with experiment. Representing the nucleus as an incompressible drop of radius  $R = \frac{1}{2}A^{1/3} \times 10^{-13}$  cm gives about  $1\frac{1}{2}$  times as large values of the isotope shift as one observes experimentally.

A number of studies in recent years have been devoted to reconciling the theory of the isotope shift with experiment. They treat the influence on the isotope shift of effects involving deformation of the nuclei, the shape of the charge distribution, compressibility, axial asymmetry, and the shell structure of nuclei. We shall briefly take up the results of some of these studies dealing with theoretical matters.

The most general relations for the isotope shift were derived by Bodmer<sup>[5]</sup> under the assumption that there is a charge distribution within the volume of the nucleus, but without concretizing this distribution. The difference in energies of photons emitted by atoms of two isotopes of the element  $Z$  is determined by the relation

$$\delta E = \Phi \frac{R_\infty}{\Gamma^2(2\rho)} \frac{3-2\rho}{3+2\rho} \left( \frac{2Z}{a_0} R \right)^{2\rho} \frac{\delta \langle r^2 \rangle}{R^2}, \quad (2.1)$$

where  $R_\infty$  is the Rydberg constant,  $\rho^2 = 1 - (Z/137)^2$ ,  $a_0$  is the first Bohr radius,  $R = 1.2 A^{1/3} \times 10^{-13}$  cm; and

$\langle r^2 \rangle$  is the mean-square radius of the charge distribution in the nucleus.

The quantity  $\Phi$  generally depends on the variation during the electronic transition of the sum of the wave functions of all the electrons at the site of the nucleus. It is usually assumed that the isotope shift is completely determined by the optical  $s$  electron that penetrates most deeply to the nucleus (or to a lesser degree, the  $p_{1/2}$  electron). However, the variation of the wave functions of the electron core as the optical electron undergoes transition are considered insignificant. The difficulty of estimating the contribution of the electron core to the isotope shift renders the calculations of the shifts in the energy levels of atoms of different isotopes of an element somewhat indefinite. Hence, one generally uses a quantity not depending on the electron shells of the atom to characterize the isotope shift. This quantity, which is called the isotope-shift constant,

$$C = \frac{5}{3} \frac{R_\infty}{\Gamma^2(2\rho)} \frac{3-2\rho}{3+2\rho} \left( \frac{2Z}{a_0} R \right)^{2\rho} \frac{\delta \langle r^2 \rangle}{R^2}, \quad (2.2)$$

characterizes the difference in the charge distribution in the nuclei of the isotopes, and is determined by the variation in the mean-square radius of the proton distribution in going from isotope to isotope.

Thus, in calculating the isotope-shift constant, a very important factor is the choice of representation of the mean-square radius of the proton distribution. One usually assumes that  $\langle r^2 \rangle$  is a function of  $A^{1/3}$ . For spherical nuclei,

$$\langle r^2 \rangle = sR^2, \quad R = 1.2A^{1/3} \cdot 10^{-13} \text{ cm}$$

where  $s$  is a parameter characterizing the radial charge distribution in the nucleus. For a uniform distribution throughout the nucleus,  $s = \frac{3}{5}$ .

For deformed nuclei ( $150 < A < 190$ ) having a rotational spectrum, the mean-square radius of the proton distribution is<sup>[6]</sup>

$$\langle r^2 \rangle = \frac{3}{5} R^2 \left( 1 + \frac{5}{4\pi} \beta^2 + \frac{3}{8\pi} \beta^4 \right), \quad (2.3)$$

where  $\beta$  is the deformation parameter of the nucleus.

For nuclei having a vibrational spectrum, the vibrator being harmonic,<sup>[6]</sup>

$$\langle r^2 \rangle = \frac{3}{5} R^2 \left( 1 + \frac{5}{4\pi} \langle \beta^2 \rangle \right), \quad (2.4)$$

Apart from a coefficient,  $\langle \beta^2 \rangle$  here is the amplitude of the zero-point vibrations of the surface of the nucleus with respect to its equilibrium shape, or the dynamic deformation.

A. S. Davydov and G. F. Filippov<sup>[11]</sup> interpret the spectra of certain nuclei occurring in the regions  $60 < A < 160$  and  $180 < A < 240$  as being rotational spectra of non-axially deformed nuclei whose surfaces are characterized by parameters  $\beta$  and  $\gamma$ . The stated regions include nuclei near the magic numbers, and also nuclei having an equidistant spectrum of levels, which is usually considered to be a vibrational spectrum.<sup>[12]</sup> In the Davydov-Filippov model, the mean-square radius of the proton distribution is expressed as follows:

$$\langle r^2 \rangle = \frac{3}{5} R^2 \left[ 1 + \frac{5}{4\pi} \beta^2 + \frac{3}{8\pi} \beta^2 f(\gamma) \right], \quad f(\gamma) = \cos \gamma (1 - 4 \sin^2 \gamma). \quad (2.5)$$

The parameter  $\gamma$  varies from  $0^\circ$  for nuclei having a rotational spectrum to  $30^\circ$  for nuclei having an equidis-

tant spectrum of levels. In these limiting cases, the Davydov-Filippov model gives the same results for  $\langle r^2 \rangle$  as Eqs. (2.3) and (2.4) do.

Experiments on scattering of fast electrons by nuclei<sup>[13]</sup> indicate that the proton density within the nucleus is constant except in the boundary region, where it smoothly declines to zero. From this fact we get the following expression for the radius of the nuclear charge:

$$R = (1.115 A^{1/3} + 2.151 A^{-1/3} - 1.742 A^{-1}) \cdot 10^{-13} \text{ cm}$$

The dimensions of the nuclear charge determined in this way practically coincide with  $1.2 A^{1/3} \times 10^{-13}$  cm. However, the relative change in the radius upon adding neutrons to a nucleus takes place more slowly in this case:

$$\frac{\delta R}{R} = \frac{1 - 1.93A^{-2/3} + 4.69A^{-4/3}}{1 + 1.93A^{-2/3} - 1.56A^{-4/3}} \frac{\delta A}{3A} = \zeta \frac{\delta A}{3A}. \quad (2.6)$$

The coefficient  $\zeta = 0.88 \pm 0.02$  in the region  $125 \leq A \leq 218$ .

Thus, the relative change in the mean-square radius of the charge distribution in the nucleus, which determines the value of the constant in (2.2), can be written in the form

$$\frac{\delta \langle r^2 \rangle}{R^2} = \frac{2}{5} \zeta \frac{\delta A}{A} + \frac{3}{4\pi} \delta (\beta^2 + 0.3\beta^2 f), \quad (2.7)$$

$\delta A = A_2 - A_1$ ;  $\delta (\beta^2 + 0.3\beta^2 f) = (\beta_2^2 + 0.3\beta_2^2 f(\gamma_2)) - (\beta_1^2 + 0.3\beta_1^2 f(\gamma_1))$ . The subscripts 1 and 2 refer to the light and heavy isotopes, respectively. The first term in this relation characterizes the volume isotope-shift constant arising from the uniform increase in the nuclear radius as neutrons are added, while the second term is the deformation constant, which determines the contribution to the isotope shift made by the variation in the nuclear deformation.

Thus, the relation of the mean-square radius of the proton distribution to the nuclear deformation makes it possible to understand the deviations observed in the spectra of many elements from equidistance in the positions of the components belonging to the even-even isotopes. However, it leaves unexplained the large jumps in the isotope shift that appear at the nuclei having magic numbers of neutrons. In addition, calculations of the isotope shift made within the framework of these concepts give results that are  $1\frac{1}{2}$  times too high.

Meligy<sup>[14]</sup> has calculated the isotope-shift constant for a trapezoidal charge distribution in the nucleus, and obtained good agreement with experiment. However, in his calculations he made an insufficiently correct choice of the numerical values of the parameters of the trapezoidal distribution<sup>[15,16]</sup> determining the mean-square radius and its variation in going from isotope to isotope. The more exact calculations of F. A. Babushkin<sup>[17]</sup> show that the trapezoidal nature of the form of the charge distribution has very little effect on the size of the isotope-shift constant.

One can gain a certain reduction in the disagreement between the theory of the isotope shift and experiment by assuming that the nuclear material is compressible<sup>[5,18]</sup> This involves a slower increase in the radius of the nucleus as neutrons are added than one would expect from the drop model. E. E. Fradkin<sup>[16]</sup> has proposed explaining the entire discrepancy between

theory and experiment by the compressibility effect by introducing two parameters. One of these is the coefficient of regular compressibility  $\eta = (\delta R/R)/(\delta N/3A) < 1$ , which characterizes the denser packing of the nucleus upon adding neutrons, as compared with the  $A^{1/3}$  law, the other is the coefficient of deformational compressibility  $\xi < 0$ , which indicates the effective increase in the proton density at the center of the nucleus as the latter becomes deformed. The isotope-shift constant derived by Fradkin for an arbitrary charge distribution over the volume of the nucleus is expressed with good accuracy in the region  $Z \leq 80$  as follows:

$$C = \frac{R_\infty}{3} \frac{(1+\rho)^2}{\Gamma^2(2\rho+1)} \left( \frac{2Z}{a_0} R \right)^{2\rho} \frac{\delta \langle r^2 \rangle}{R^2}. \quad (2.8)$$

The coefficient of regular compressibility gives rise to the following expression for the variation of the nuclear radius as neutrons are added:

$$\frac{\delta R}{R} = \eta \zeta \frac{\delta N}{3A}, \quad (2.9)$$

where  $\zeta$  is determined by Eq. (2.6). The deformational compressibility can be explained either by compressibility of the nucleus upon varying the deformation ( $R = R(\beta)$ ), or by variation of the radial charge distribution upon deformation ( $s = s(\beta)$ ). One cannot distinguish these two factors within the framework of the Fradkin model. The expression for the mean-square radius of the proton distribution in the deformed nucleus of (2.5) takes on the form

$$\langle r^2 \rangle = \frac{3}{5} R^2 (1 + \xi \beta^2) \left[ 1 + \frac{5}{4\pi} \beta^2 + \frac{3}{8\pi} \beta^2 f(\gamma) \right] \approx \frac{3}{5} R^2 \left[ 1 + \left( \frac{5}{4\pi} + \xi \right) \beta^2 + \frac{3}{8\pi} \beta^2 f(\gamma) \right]. \quad (2.10)$$

The numerical values of the compressibility coefficients  $\eta = 0.7$  and  $\xi = -5/8\pi$  were selected<sup>[16]</sup> to get best agreement of the theoretical calculations of isotope shifts with the experimental data in the region  $56 \leq Z \leq 82$ .

Under these assumptions, the relative variation of the mean-square radius of the charge distribution in a deformed nucleus as neutrons are added takes on the form

$$\frac{\delta \langle r^2 \rangle}{R^2} = \frac{2}{5} \eta \zeta \frac{\delta A}{A} + \frac{3}{8\pi} \delta (\beta^2 + 0.6\beta^2 f(\gamma)). \quad (2.11)$$

Analogous relations in a somewhat more general form have been derived by F. A. Babushkin,<sup>[17]</sup> who used the same values of the compressibility coefficients, and by Ionesco-Pallas.<sup>[19]</sup>

Thus, introduction of the hypothesis of compressibility of nuclei as neutrons are added considerably lessens the volume isotope-shift constants calculated by Eqs. (2.8) and (2.11). However, compressibility of nuclei with increasing deformation diminishes the role of deformation in the isotope shift, and permits better explanation of deviations from equidistance in the positions of the components belonging to even-even isotopes. However, the large jumps observed in the isotope shift when a pair of neutrons is added to nuclei having a magic number of neutrons cannot be explained by representing  $\langle r^2 \rangle$  as a function of  $A^{1/3}$ ,  $\beta$ , and  $\gamma$ , since nuclei show no appreciable deformations near the magic numbers. This fact indicates that the mean-square radius of the proton distribution must depend on the individual properties of the nuclei, as manifested

in the sequence of filling of the neutron levels. The role of the shell structure in the jump in the isotope shift at the magic number  $N = 126$  was first estimated by Breit.\* A systematic theoretical treatment of the role of the shell structure of the nucleus in the isotope shift has been carried out by G. G. Bunatyan and M. A. Mikulinskiĭ,<sup>[9]</sup> and also by V. A. Belyakov,<sup>[20]</sup> based on the theory of the Fermi fluid in finite systems, as developed by A. B. Migdal.<sup>[8]</sup>

The essence of these concepts consists in an interaction between nuclear quasiparticles, each of which moves in the self-consistent field of the rest of the quasiparticles. The variation in the mean-square radius of the charge distribution is determined by the variation in the proton density as neutrons are added:

$$\delta \langle r^2 \rangle = \sum_{\lambda\lambda'} (\delta\sigma)_{\lambda\lambda'} \langle r^2 \rangle_{\lambda\lambda'}$$

Here  $\lambda$  is the set of quantum numbers  $n, l, j$ , and  $m$  of the quasiparticles; and  $\delta\sigma$  is the variation in the density matrix of the protonic quasiparticles, which is due to the interaction of the added neutrons with the rest of the quasiparticles. The nature of the interaction of the nuclear quasiparticles is determined by the interaction amplitudes  $f^{np}$ ,  $f^{nn}$ , and  $f^{pp}$ , which are not calculated in the theory, but found by comparison with experiment. The calculations performed<sup>[8,9,20]</sup> have shown that the values of the interaction amplitudes determined from various physical processes (quadrupole moments of nuclei, isomer and isotope shifts, etc.) agree quite well with one another. It turned out that  $f^{pp}$  and  $f^{np}$  within the nucleus are positive, and several times smaller in absolute value than at the surface of the nucleus, where they become negative. This indicates an effective repulsion of the quasiparticles within the nucleus, but an attraction at the boundary of the nucleus. Owing to the large absolute values of the external interaction constants, the contribution of surface phenomena in the nucleus proves to be quite substantial, in spite of the fact that the boundary region of the nucleus is  $A^{1/3}$  times smaller than the interior. The strong interaction of the added neutrons with the rest of the nucleons causes the mean-square radius of the nuclear charge to depend on the state  $\lambda$  in which the added neutrons exist.

The relations for  $\delta \langle r^2 \rangle$  derived in the theory of finite Fermi systems permitting calculations for concrete nuclei are very cumbersome and require machine calculation. Hence we shall not present them. We shall point out that V. P. Kraĭnov and M. A. Mikulinskiĭ<sup>[21]</sup> have recently solved the problem of calculating  $\delta \langle r^2 \rangle$  analytically in the quasiclassical approximation. As a result, they derived a simple formula for the variation in the mean-square radius of the charge distribution as neutrons are added to the nucleus into a state with orbital angular momentum  $l$ :

$$\frac{\delta \langle r^2 \rangle}{R^2} = \frac{\delta A}{2Z} \left( a + b \frac{l^2}{l_0^2} \right), \quad (2.12)$$

where

$$a = \frac{0.2 + 1.3A^{-1/3}}{2.7 - 8.8A^{-1/3}},$$

$$b = -\frac{0.5 + 0.5A^{-1/3}}{2.7 - 8.8A^{-1/3}}, \quad l_0 = 1.7A^{1/3}.$$

This approximate formula gives somewhat poorer results than the exact calculations do. Nevertheless, it very graphically illustrates the implications of the theory of finite Fermi systems with regard to the anomalies in the isotope shift upon filling neutron levels. Thus, the jumps in the isotope shift at the magic neutron numbers 50, 82, and 126 are easily explained by addition of the pair of neutrons to a new level of smaller orbital angular momentum  $l$ . Here the mean-square proton radius increases considerably according to (2.12), since the neutrons of smaller angular momentum  $l$  interact effectively with a large number of nuclear quasiparticles, and this causes a substantial redistribution of the charge in the nucleus.

Calculations of the isotope shift in the Fermi-fluid theory have been carried out in the region of spherical nuclei. An attempt is being made at present to apply the theory to deformed nuclei.

### III. COMPARISON OF THE THEORY OF THE ISOTOPE SHIFT WITH EXPERIMENT

Eq. (2.1) implies that the ratio  $\delta E/\Phi$  must be a characteristic constant for each pair of isotopes, independently of the electronic state of the atom. When this ratio is determined by experiment, it is called the experimental isotope-shift constant for the two isotopes of the element  $Z$ , and is determined by the formula

$$C_{\text{exp}} = \frac{\delta T_s}{\pi a_0^3 \psi_s^2(0)/Z}, \quad (3.1)$$

where  $\delta T_s$  is the volume isotope shift in the  $s$ -electron term, and  $\psi_s(0)$  is the value of the non-relativistic wave function of the  $s$  electron at the site of the nucleus. The quantity  $\delta T_s$  is found from the experimentally-measured isotope shifts in the lines by eliminating the mass shift and taking into account the shielding effect of the electrons in the unfilled shells.<sup>[2]</sup>

The value of  $\psi_s^2(0)$  is usually estimated by the approximate Fermi-Segre formula

$$\psi_s^2(0) = \frac{ZZ_a^2}{\pi a_0^3 n_a^2} \left( 1 - \frac{d\sigma}{dn} \right),$$

where  $Z_a$  is the effective charge acting on the outer part of the orbital of the  $s$  electron ( $Z_a = 1$  for a neutral atom,  $Z_a = 2$  for a singly-ionized atom, etc.);  $n_a = n - \sigma$  is the effective quantum number, which is found by equating the energy of the  $s$ -electron term with the quantity  $R_\infty Z_a^2/n_a^2$ ;  $n$  is the principal quantum number; and  $\sigma$  is the quantum defect.

Neglect of the shielding effect of the electron core gives rise to a certain indefiniteness in the value of the experimental isotope-shift constant found in this way. One takes this indefiniteness into account by applying the coefficient  $\beta' \approx 1$  to  $C_{\text{exp}}$ .\* One usually assumes that the quantity  $\beta' C_{\text{exp}}$  gives an estimate of the experimental constant  $C_{\text{exp}}$  to an accuracy of about 20%.

The values of  $\beta' C_{\text{exp}}$  are suitable for testing the theory of the isotope shift, since they permit one directly to compare the experimental data with the theoretical isotope-shift constants calculated by Eq. (2.1) or (2.8) for some particular model of the nucleus.

\*We write the coefficient  $\beta$  with a prime to distinguish it from the nuclear deformation parameter.

We shall compare the experimental isotope-shift data with the results given by the model of a compressible deformed nucleus and the model taking the shell structure of the nucleus into account.

The deformation parameters  $\beta$  required to calculate the theoretical isotope-shift constants are usually found<sup>[12,16]</sup> from the probabilities of  $E^2$  electric quadrupole transitions from the ground state of the nucleus with spin  $J = 0$  to the first excited state with spin  $J = 2$ :

$$B(E2.0 \rightarrow 2) = a^2 \beta^2 (1 + 0.36\beta)^2. \quad (3.2)$$

where  $a = (3/4\pi)eZR^2$ ,  $e$  is the charge of the electron, and  $R = 1.2 A^{1/3} \times 10^{-13}$  cm. This relation holds both for deformed and spherical even-even nuclei. The quantity  $\beta^2 = \langle \beta^2 \rangle$  is the mean square of the varying deformation in the ground state of the nucleus.

In a deformed nucleus, the surface can undergo longitudinal  $\beta$ -oscillations with respect to the equilibrium shape characterized by the static deformation parameter  $\beta_0$ . Here the mean-square value of the parameter  $\beta$  proves to be<sup>[23]</sup>

$$\langle \beta^2 \rangle = \beta_0^2 \left( 1 + \frac{\mu^2}{2} \right),$$

where  $\mu$  is the non-adiabaticity parameter introduced by A. S. Davydov and A. A. Chaban,<sup>[24]</sup> which characterizes the relation of the zero-point  $\beta$ -oscillations of the surface to the rotation of the nucleus. If  $\mu = 0$ , then the rotation of the nucleus and the longitudinal oscillations occur independently, and  $\langle \beta^2 \rangle = \beta_0^2$ .

For spherical nuclei ( $\beta_0 = 0$ ), the mean value  $\langle \beta^2 \rangle$  in the ground state of the nucleus is<sup>[23]</sup>

$$\langle \beta^2 \rangle = \frac{5}{2} \beta_{00}^2,$$

where  $\beta_{00}$  is the amplitude of the zero-point  $\beta$ -oscillations of the surface of the nucleus with respect to its equilibrium spherical shape.

Equation (3.2) permits one to use the probabilities of  $E2$  transitions found by Coulomb excitation of nuclei to calculate  $\beta = \sqrt{\langle \beta^2 \rangle}$ . However, the problem of the nature of the values of  $\beta$  thus obtained remains unclear. Analysis of the higher excited states of nuclei has shown<sup>[23]</sup> that the non-adiabaticity parameter is small for nuclei of neutron numbers  $88 < N < 120$  ( $\mu \leq 0.2$ ). That is, static deformation plays the major role in these nuclei, and the calculated values of  $\beta$  practically coincide with the parameter  $\beta_0$  characterizing the deformation of the equilibrium shape of the nucleus. As we approach the nuclei having a magic number of neutrons, the parameter  $\mu$  rapidly increases (especially near  $N = 82$ ). In the region  $50 \leq N \leq 82$ , the non-adiabaticity is also quite significant ( $\mu \geq 0.5$ ). This means that the contribution of zero-point oscillations of the surface is substantial in these nuclei.

Thus, if the non-adiabaticity parameter  $\mu$  substantially differs from zero, the values of  $\beta$  calculated from transition probabilities cannot serve as an unambiguous characteristic of the equilibrium shape of the nucleus. In this case, the values of  $\beta$  characterize the dynamic deformation of the nucleus, which can exist even in spherical nuclei. Analogously to static deformation, it must be manifested in the isotope shift.

Table XI gives the reduced probabilities of electric quadrupole transitions of nuclei from the ground state

Table XI. Reduced probabilities of  $E2$  transitions (in barns<sup>2</sup>) and deformation parameters of even-even nuclei

Nucleus	$N$	$B(E2)$ $e^2$	Reference	$\beta$	Nucleus	$N$	$B(E2)$ $e^2$	Reference	$\beta$
Sr	84	0.34	26	0.216	Ce	140	0.27	26	0.094
	86	0.19	26	0.161		142	0.41	26	0.114
	88	0.20	26	0.163	Nd	142	—	—	0
Zr	90	—	—	0		144	0.23	26	0.083
	92	0.094	26	0.105		146	0.25	26	0.085
	94	0.081	26	0.103		148	0.57	26	0.125
Mo	92	—	—	0		150	1.92	26	0.220
	94	0.270	25	0.164	Sm	144	0.39	26	0.103
	96	0.302	25	0.171		148	0.89	27	0.150
	98	0.270	25	0.160		150	1.32	27	0.179
100	0.614	25	0.232	152		3.40	27	0.276	
154	4.61	27	0.315						
Ru	96	0.254	25	0.150	Gd	154	3.43	27	0.267
	98	0.475	25	0.200		156	4.57	27	0.302
	100	0.572	25	0.214		158	5.44	27	0.325
	102	0.733	25	0.237		160	5.80	27	0.332
104	0.928	25	0.262	Dy	156	3.79	27	0.269	
Pd	104	0.547	25		0.196	158	4.67	27	0.293
	106	0.646	25		0.211	160	4.46	27	0.286
	108	0.742	25		0.222	162	5.11	27	0.303
	110	0.860	25	0.235	164	5.64	27	0.314	
Cd	106	0.470	25	0.174	Er	162	4.89	27	0.287
	108	0.535	25	0.183		164	5.04	27	0.290
	110	0.504	25	0.176		166	5.66	27	0.304
	112	0.542	25	0.179		168	5.72	27	0.303
	114	0.584	25	0.184	170	5.44	27	0.294	
	116	0.900	25	0.185	Yb	168	5.43	27	0.287
Sn	112	0.18	25	0.102		170	5.53	27	0.289
	114	0.20	25	0.107		172	5.89	27	0.295
	116	0.207	25	0.107		174	5.89	27	0.293
	118	0.228	25	0.112		176	5.78	27	0.288
	120	0.220	25	0.108	Hf	174	5.26	27	0.271
	122	0.252	25	0.115		176	5.27	27	0.269
124	0.213	25	0.105	178		4.66	27	0.253	
180	4.35	27	0.243						
Te	120	—	25	0.195	W	182	4.00	27	0.227
	122	—	4	0.175		184	3.62	27	0.215
	124	—	26	0.160		186	3.57	27	0.212
	126	0.532	25	0.153		Os	186	4.36	28
	128	0.412	25	0.135	188		3.7	28	0.208
130	0.340	25	0.122	190	3.38		28	0.199	
192	2.92	28	0.185	Hg	198	1.13	30	0.109	
Ba	130	0.73	29		0.163	200	0.85	30	0.085
	132	0.72	29		0.160	202	0.59	30	0.079
	134	0.75	26	0.161	Pt	194	1.7	12	0.138
	136	0.53	26	0.137		196	1.3	12	0.121
138	0.27	26	0.097	198		1.4	12	0.124	
Pb	206	0.14	12	—	208	—	12	0	

to the first excited state for most of the discussed elements, and also the corresponding values of the deformation parameter  $\beta$  as calculated by Eq. (3.2). The probabilities of the  $E2$  transitions were found from experiments on Coulomb excitation of nuclei, and we have taken them from the studies cited in the table. In most cases, the errors in the  $\beta$  values do not exceed 5–10%.

The left-hand side of Table XII gives the most reliable experimental data on relative isotope shifts and isotope-shift constants (with citation of the studies from which they are taken). Whenever the values of the relative shifts were not given in the cited studies, we have calculated them from the experimentally-

Table XII. Relative isotope shifts and isotope-shift constants (in  $10^{-3} \text{ cm}^{-1}$  units)

Isotope pair N <sub>1</sub> -N <sub>2</sub>	Experimental isotope-shift data		Theoretical isotope-shift constants			$\lambda$
	Relative isotope shift	$\beta'C_{\text{exp}}$	Eqs. (2,8) (2,11)	Fermi fluid		
				$C_{\text{vol}}$	$C_{\text{vol}}+C_{\text{def}}$	
<sup>38</sup> Sr	35	35				
46-48	< 0.2	< 7	10	12	2	1g <sub>9/2</sub>
48-50	< 0.2	< 7	20	12	12	1g <sub>9/2</sub>
50-52	1	47±5	—	20	—	2d <sub>5/2</sub>
<sup>40</sup> Zr	37	37				
50-52	1.47±0.03	37±3	28	27	33	2d <sub>5/2</sub>
52-54	1	25±3	22	27	27	2d <sub>5/2</sub>
54-56	0.68±0.03	17±2	—	17	—	1g <sub>7/2</sub>
<sup>40</sup> Mo		*				
50-52	2.03±0.15	41	44	19	37	1g <sub>7/2</sub>
52-54	1.37±0.15	28	28	19	21	1g <sub>7/2</sub>
54-56	1	20	23	19	17	1g <sub>7/2</sub>
56-58	2.08±0.20	43	46	19	40	1g <sub>7/2</sub>
<sup>44</sup> Ru	42	22				
52-54	1.24	42±10	45	22	37	1g <sub>7/2</sub>
54-56	1.09	37±9	35	22	27	1g <sub>7/2</sub>
56-58	1	34±8	38	22	31	1g <sub>7/2</sub>
58-60	1.10	37±9	40	22	33	1g <sub>7/2</sub>
<sup>46</sup> Pd	43	22				
56-58	0.79	28±4	—	39	—	2d <sub>5/2</sub>
58-60	1.21	44±6	41	39	46	2d <sub>5/2</sub>
60-62	1.08	39±5	38	39	44	2d <sub>5/2</sub>
62-64	1	36±5	39	26	32	1g <sub>7/2</sub>
<sup>48</sup> Cd	49	22				
58-60	1.07	34±4	42	31	35	1g <sub>7/2</sub>
60-62	1.00	32±4	35	31	28	1g <sub>7/2</sub>
62-64	1	32±4	38	31	32	1g <sub>7/2</sub>
64-66	0.93	30±4	39	25	27	1h <sub>11/2</sub>
66-68	0.65	20±3	37	25	25	1h <sub>11/2</sub>
<sup>50</sup> Sn	51	51				
62-64	1.40	44±10	45	37	39	1g <sub>7/2</sub>
64-66	1.39	44±10	43	37	37	1g <sub>7/2</sub>
66-68	1	31±10	44	34	35	1h <sub>11/2</sub>
68-70	1.04	32.5±10	41	28	27	1h <sub>11/2</sub>
70-72	0.86	27±10	44	22	24	1h <sub>11/2</sub>
72-74	0.37	11.5±10	39	18	15	1h <sub>11/2</sub>
<sup>52</sup> Te	52	—				
68-70	1.34	—	35	40	27	2d <sub>3/2</sub>
70-72	1.26	—	40	40	32	2d <sub>3/2</sub>
72-74	1	—	44	25	21	1h <sub>11/2</sub>
74-76	0.96	—	38	25	16	1h <sub>11/2</sub>
76-78	0.83	—	41	25	19	1h <sub>11/2</sub>
<sup>54</sup> Xe	22	22				
78-80	~0.7	~18	—	20	—	1h <sub>11/2</sub>
80-82	1	25±6	—	40	—	2d <sub>3/2</sub>
<sup>56</sup> Ba	55	55,4				
74-76	0.47	37	59	39	37	1h <sub>11/2</sub>
76-78	0.47	37	61	39	39	1h <sub>11/2</sub>
78-80	0.42	33	44	39	23	1h <sub>11/2</sub>
80-82	1	78	38	71	50	2d <sub>3/2</sub>
<sup>58</sup> Ce	56	22				
78-80	0.26	38±8	—	63	—	1h <sub>11/2</sub>
80-82	0.26	38±8	—	63	—	1h <sub>11/2</sub>
82-84	1	147±30	79	131	142	2f <sub>7/2</sub>
<sup>60</sup> Nd	60	22				
82-84	1	187±35	98	142	162	2f <sub>7/2</sub>
84-86	0.87±0.02	163±33	79	142	143	2f <sub>7/2</sub>
86-88	1.04±0.01	194±38	103	142	168	2f <sub>7/2</sub>
88-90	1.42±0.02	266±54	186	142	251	2f <sub>7/2</sub>
<sup>62</sup> Sm	63, 66	22				
82-86	2	380±80	225	310	354	2f <sub>7/2</sub>
86-88	1.21±0.01	230±38	127	155	192	2f <sub>7/2</sub>
88-90	1.81±0.02	344±75	284	155	330	2f <sub>7/2</sub>
90-92	0.87±0.01	165±36	193	75	179	1h <sub>9/2</sub>
<sup>64</sup> Gd	70	22				
88-90	3.00±0.07	375±70	—	165	—	2f <sub>7/2</sub>
90-92	1.30±0.09	163±37	195	80	176	1h <sub>9/2</sub>
92-94	0.97±0.08	121±30	170	80	151	1h <sub>9/2</sub>
94-96	1	125±30	121	80	103	1h <sub>9/2</sub>
<sup>66</sup> Dy	73, 82	—				
90-92	1.64±0.03	—	189	—	—	—
92-94	0.97±0.02	—	90	—	—	—
94-96	1.11±0.01	—	170	—	—	—
96-98	1	—	153	—	—	—
<sup>68</sup> Er	74, 82	—				
94-96	1.14±0.01	—	139	—	—	—
96-98	0.98±0.01	—	183	—	—	—
98-100	0.95±0.01	—	125	—	—	—
100-102	1	—	91	—	—	—
<sup>70</sup> Yb	79	22				
98-100	1.35±0.01	133±12	153	—	—	—
100-102	1.25±0.01	123±11	170	—	—	—
102-104	1	99±8	135	—	—	—
104-106	0.91±0.01	90±8	118	—	—	—

Table XII. (continued)

Isotope pair N <sub>1</sub> -N <sub>2</sub>	Experimental isotope-shift data		Theoretical isotope-shift constants			$\lambda$
	Relative isotope shift	$\beta'C_{\text{exp}}$	Eqs. (2,8) (2,11)	Fermi fluid		
				$C_{\text{vol}}$	$C_{\text{vol}}+C_{\text{def}}$	
<sup>72</sup> Hf	82	22				
102-104	0.89±0.01	101±13	154	—	—	—
104-106	0.85±0.01	97±12	89	—	—	—
106-108	1	114±13	117	—	—	—
<sup>74</sup> W	86	86				
106-108	0.79±0.07	92±24	—	—	—	—
108-110	1.13±0.02	133±25	132	—	—	—
110-112	1	117±20	172	—	—	—
<sup>76</sup> Os	90	91				
108-110	1.31±0.04	149±22	—	—	—	—
110-112	1.29±0.02	147±20	110	—	—	—
112-114	1.12±0.02	128±18	164	189	147	1i <sub>13/2</sub>
114-116	1	114±14	143	190	128	1i <sub>13/2</sub>
<sup>78</sup> Pt	22	22				
116-118	1	135±25	180	215	158	1i <sub>13/2</sub>
118-120	1.04	140±26	244	215	224	1i <sub>13/2</sub>
<sup>80</sup> Hg	96	97				
116-118	0.81±0.04	190±34	—	241	—	1i <sub>13/2</sub>
118-120	0.89±0.01	209±29	227	242	201	1i <sub>13/2</sub>
120-122	1	235±30	227	243	203	1i <sub>13/2</sub>
122-124	0.95±0.02	223±33	—	244	—	1i <sub>13/2</sub>
<sup>82</sup> Pb	22, 100	22				
122-124	0.90	284±32	—	277	—	1i <sub>13/2</sub>
124-126	1	315±35	288	288	273	1i <sub>13/2</sub>
126-128	1.74	548±60	—	494	—	2g <sub>9/2</sub>

\*See Table VI.

measured shifts between the components of the isotopes, deducting the mass shift whenever possible.

As we pointed out in Sec. 1, a number of elements have shown a difference between the relative shifts in lines shifted in positive and negative directions. At present there is no sufficiently grounded theoretical explanation for this phenomenon. Hence, in finding the relative isotope shift due to the volume effect, we have taken the mean of the relative shifts for lines having opposite directions of shift. To some degree, this has made it possible to diminish the influence on the relative shift of supplementary effects of whatever nature.

We have recalculated the experimental isotope-shift constants given in the table to agree with the new data on the relative shifts. As the starting value of  $\beta'C_{\text{exp}}$  for each element, we have taken its value for the pair of isotopes on which the relative shift was normalized (the experimental constants for these pairs of isotopes were found directly from the studies cited in the table).

Figure 10 shows the relation of the experimentally-measured relative isotope shifts to the number of neutrons in the nuclei (using the data of Table XII). We can easily see that the relative shift follows an analogous course for the isotopes of different elements containing the same number of neutrons in their nuclei.

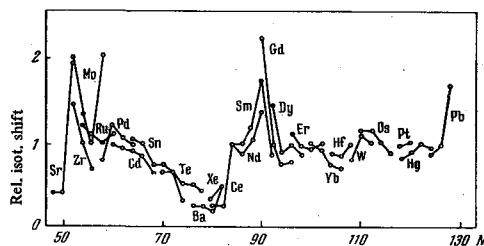


FIG. 10. Experimental values of the relative shifts.



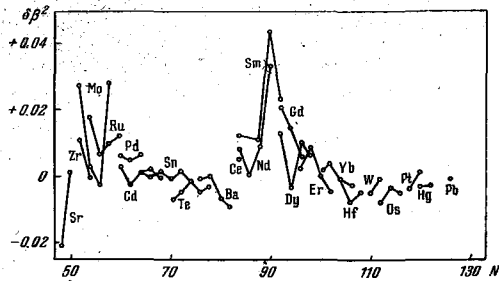


FIG. 11. Change in the deformation upon adding two neutrons to the nucleus (nuclear deformation parameters found by Coulomb-excitation experiments).

The relative isotope shift increases considerably when neutrons are added to nuclei having a magic number of neutrons (Sr, Zr, Mo, Ce, and Pb). A jump in the relative shift is also observed for the neutron pair 88–90, i.e., for the isotopes in which the nuclear deformation varies greatly (Nd and Sm). The subsequent slow variation in the nuclear deformation produces smaller relative shifts (Sm, Gd, and further on).

Figure 11 shows the nuclear deformations found from Coulomb-excitation experiments in the form of the difference of  $\beta^2$  values for adjacent even-even nuclei (the graph is based on Table XI). Comparison of Figs. 10 and 11 shows that the course of the relative shift agrees rather well with that of the nuclear deformation upon adding neutrons. Apart from the magic numbers of neutrons, variations in the nuclear deformations basically give a correct description of all the experimentally-found deviations from equidistance in the positions of the components due to even-even isotopes. This permits us to use with assurance Coulomb data on nuclear deformations to calculate theoretical isotope-shift constants.

In calculating theoretical isotope-shift constants, we must also know the axial-asymmetry parameter  $\gamma$  of the nuclei, which is found from data on the energies of excited states. A. S. Davydov<sup>[23]</sup> has shown that the parameter  $\gamma$  is small ( $\gamma \leq 13^\circ$ ) in the rare-earth region. Near  $N = 126$  and in the region  $50 \leq N \leq 88$ ,  $\gamma$  exceeds  $20^\circ$ . This shows that either such nuclei lack axial symmetry even in the ground state or that they show considerable zero-point  $\gamma$ -oscillations of the surface of an axially-symmetric nucleus. We see from Eqs. (2.5) and (2.11) that the effect of the parameter  $\gamma$  on the size of the theoretical isotope-shift constant should be insignificant, since the function  $0 \leq f(\gamma) \leq 1$  figures in the deformation constant in the form of a small additive term. For a number of elements (Gd, Os, etc.), whose nuclei have substantially different  $\gamma$  values, calculations have shown<sup>[16,31]</sup> that the contribution to the isotope shift from the variation in axial asymmetry of the nuclei does not exceed several percent. In line with this, we have assumed that  $\gamma = 0^\circ$  for all nuclei in calculating the theoretical isotope-shift constants. The values of the theoretical isotope-shift constants are given on the right-hand side of Table XII.

The fourth column of the table gives the isotope-shift constants calculated under the assumption of a compressible deformed nucleus by Eqs. (2.8) and (2.11)

with a compressibility coefficient  $\eta = 0.7$  and  $\xi = -5/8\pi$ . The values of the  $\beta$  parameters were taken from Table XI.

The next column of the table gives the isotope-shift constants taken from<sup>[8,9,20]</sup>\* These constants were calculated by Eq. (2.8), the variation in the mean-square proton radius being found from the theory of the Fermi fluid in finite systems.

The isotope-shift constants in the sixth column of Table XII were obtained by formally taking into account the nuclear deformations in the Fermi-fluid theory. We can picture this physically as follows. When the addition of neutrons to a nucleus does not alter the nuclear deformation, then the variation in the mean-square proton radius of a deformed nucleus can be calculated by the Fermi-fluid theory developed for spherical nuclei. However, when the nuclear deformation varies upon adding neutrons, then it can be taken into account in the usual way. In this case, the isotope-shift constants are found by Eqs. (2.8) and (2.11). The first term in (2.11) is calculated by the Fermi-fluid theory, and the second term from the nuclear deformation determined by Coulomb-excitation experiments. To illustrate the simultaneous effect on the isotope shift of nuclear deformation and shell structure, the isotope-shift constants have been calculated by the Fermi-fluid theory for several strongly-deformed nuclei (samarium and gadolinium).

The last column in Table XII gives the sequence of filling of the neutron levels used in calculating the isotope-shift constants by the Fermi-fluid theory. In many cases it is hard to select unequivocally the state  $\lambda$  into which the neutrons are added. This is because different  $j$  levels often compete in the filling of neutron levels. One can select the state  $\lambda$  here only by comparing the calculated and experimental values of the isotope-shift constants. The  $\lambda$  states indicated in the table, to which the neutrons are added in going from isotope to isotope, do not contradict the generally accepted pattern of filling of neutron levels.

Comparison of the theoretical and experimental isotope-shift constants shows that both of the discussed models give fairly good results. In Fig. 12, the theoretical isotope-shift constants are compared with the experimental constants for the corresponding pairs of isotopes. The circles denote calculations based on the model of a compressible deformed nucleus, and the crosses denote the results given by the Fermi-fluid theory taking account of the nuclear deformation. The two straight lines drawn on the graph indicate the limits of experimental error in  $\beta'C_{\text{exp}}$  (about  $\pm 20\%$ ). Most of the Fermi-fluid points lie within the limits of experimental accuracy, while the model of the compressible deformed nucleus gives a considerably greater spread of points. Thus, account of the shell structure of the nucleus gives considerably better agreement of the calculated isotope-shift constants with experiment.

If we compare the relative values of the isotope-shift constants for the isotopes of each element with the experimental values of the relative shifts, then we note that the Fermi-fluid theory also gives somewhat

\*V. A. Belyakov and M. A. Mikulinskii have made a supplementary calculation of the isotope-shift constant for some elements.

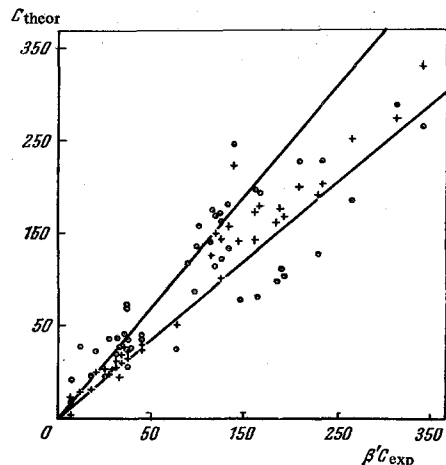


FIG. 12. Comparison of experimental isotope-shift constants with the theoretical values (in  $10^{-3} \text{ cm}^{-1}$ ), as calculated by the Fermi-fluid theory (+) and from the compressible-nucleus model (o) (with account taken of nuclear deformation).

better results. Thus, in the spectra of Cd, Sn, and Te, the relative positions of the components of the isotopes cannot be understood from the standpoint of the model of a compressible nucleus showing dynamic deformation of the surface. In the nuclei of the neighboring elements Mo and Ru, the neutrons fill the same level  $lg_{7/2}$ , and the non-equidistance in the positions of the components belonging to the even-even isotopes is correctly explained by surface oscillations of the nucleus. In the nuclei of Cd, Sn, and Te, the neutrons fill different levels, and this must be manifested in the relative isotope shift. It has previously been noted<sup>[7,32]</sup> that one must not only take into account the dynamic deformation of the nucleus in order to explain the relative positions of the components of the isotopes in the spectra of these elements, but also the shell structure. As we see from Table XII, an account of the states into which the neutrons are added in going from isotope to isotope permits one to give a fundamentally correct explanation of the relative shifts in these elements. One can get somewhat better agreement with experiment by taking into account the mixing of neighboring neutron levels. An analogous picture is observed in Er, W, and Pt, for which one single variation in the nuclear deformation does not permit one to explain the relative positions of the components of the isotopes.

Imprecision in the nuclear-deformation parameters can play a certain role in the discrepancy between theory and experiment. They are found from Coulomb-excitation experiments with a considerably greater error than that of the experimentally-measured relative isotope shifts.

In the region of transition from nuclei having a vibrational spectrum to rotational nuclei, the model of a compressible deformed nucleus gives values of the isotope-shift constants that are somewhat too low. Application of the Fermi-fluid theory developed for spherical nuclei to deformed nuclei gives better agreement of the calculations, both with the experimental constants and with the relative isotope shifts (see Table XII). The abrupt increase in the nuclear deformation of the nuclei upon adding neutrons to nuclei having

$N = 88$  explains the anomalously large isotope shift between the components of the isotopes  $\text{Nd}^{148}$  and  $\text{Nd}^{150}$ , and between  $\text{Sm}^{150}$  and  $\text{Sm}^{152}$ . The relatively gradual variation of the deformation in going to the following nuclei having  $N > 90$  can only partially explain the decline in the shift between the components of the isotopes  $\text{Sm}^{152}$  and  $\text{Sm}^{154}$ , and  $\text{Gd}^{154}$  and  $\text{Gd}^{156}$ . If we bear in mind the fact that the neutron pair to be added to a nucleus with  $N = 90$  enters a level having a large orbital angular momentum, the discrepancy with experiment practically vanishes. Thus, the peculiarities of filling of the neutron shells together with the deformation effect permit one quantitatively to explain the anomaly observed in the isotope shift for the neutron pair 88–90.

The Fermi-fluid theory also gives the correct order of magnitude for the jumps in the isotope shift for nuclei having the magic numbers of neutrons 50, 82, and 126, whereas the model of a compressible deformed nucleus is in no position to explain these jumps. A very important point is that the hypothesis of compressibility of the nuclear matter need not be adduced in the Fermi-fluid theory to estimate quantitatively the isotope shift, while an entire set of physical phenomena is treated from the standpoint of a system of strongly interacting quasiparticles having the same interaction constants.

All of this implies that the isotope shift in the spectra of the heavy elements can serve as a highly sensitive means of studying the charge distribution in the nuclei of isotopes, as characterized by the quantity  $\langle r^2 \rangle$ . We see from Eqs. (2,8) and (2,11) that the relative positions of the components of the isotopes in the spectrum of an element depend only on the differences in values of  $\langle r^2 \rangle$  of the nuclei of the isotopes. That is, it is determined by the variation in the deformation of the nuclei in going from isotope to isotope (neglecting the effect of the shell structure of the nucleus). The tables and graphs given above show that the variation in the deformation of the nuclei rather well explains the non-equidistance in the positions of the components of the isotopes. Hence, if we know the relative isotope shift and deformation parameters of two nuclei of an element, we can use the cited relations to calculate the deformation parameters of all the rest of the nuclei of this element. On the other hand, we can compare Eq. (2,8), which gives the theoretical isotope-shift constant for two isotopes of an element, with the experimental value of the constant  $\beta'C_{\text{exp}}$  for the same isotopes. Then, if we know the deformation parameter of the nucleus of one of these isotopes, we can calculate the value of  $\beta$  for the nucleus of the other isotope.

These two methods of calculating the deformation parameters of nuclei from isotope shifts can be applied to find the parameters  $\beta$  of nuclei whose percent content in the natural isotope mixtures does not suffice for performing Coulomb-excitation experiments on the nuclei. The calculated values of the deformation parameters of these rare nuclei serve as a basis for calculating the intrinsic quadrupole moments of the nuclei

$$Q_0 = \frac{3}{\sqrt{5\pi}} ZR^2\beta(1 + 0.36\beta)$$

and the reduced probabilities of electric quadrupole transitions from the ground state of the nucleus to the

first excited state (Eq. (3.2)).

We should note that the deformation parameters of certain rare isotopes (e.g., Dy<sup>156</sup> and Dy<sup>158</sup>) found in nuclear Coulomb-excitation experiments agree poorly, both with the general trend of variation of the deformation upon adding neutrons to the nucleus, and with the relative isotope shifts (see Table XI and Figs. 10 and 11). In this regard, the calculation of nuclear deformations from isotope-shift data performed for a number of heavy elements in<sup>[33]</sup> can give more plausible results. However, we must treat such calculations with some caution, since their fundamental relations (2.8) and (2.11) do not take account of the shell structure of the nuclei, which in some cases can appreciably affect the results of calculating the deformation parameters. Use of the Fermi-fluid theory to calculate  $\beta$  parameters of nuclei is somewhat premature, since this theory has been developed only for spherical nuclei thus far. The application that we have made of it to deformed nuclei is only illustrative in nature. We may hope that the further development of the theory of finite Fermi systems will encompass the extensive region of deformed nuclei, and will make it possible to perform more reliably all the calculations concerning the isotope shift in the spectra of the heavy elements.

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