Measurement of mean-square nuclear radii of Nd, Sm, and Gd by laser-excited fluorescence

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Isotope shifts in the optical spectra of the atoms Nd, Sm, and Gd have been measured. The method of measurement was based on the resonance fluorescence of these atoms. A tunable dye laser was used to excite them. The differences of the mean square charge radii of the nuclei of these elements with neutron number from 82 to 96 have been calculated from the measured values of the isotope shifts. It is shown that in the optical spectra of Nd, Sm, and Gd there are transitions for which the calculations for pure $ns^2 \rightarrow nsnp$ transitions are valid. The dependence of the mean square radii of neighboring even-even nuclei on the neutron number is discussed.

INTRODUCTION

The development of tunable lasers has raised to a new level the investigation of the optical spectra of atoms as a source of information about the properties of nuclei. The high power and spectral resolution of these lasers together with the possibility of tuning in a wide range of frequencies of the laser light have made it possible to carry out high-precision measurements of the isotope shifts and hyperfine structure using extremely few investigated atoms. Determination of the isotope shifts in atomic spectra is a very sensitive measure of the change of the mean square charge radii of nuclei when the number of neutrons in them is increased or decreased. These changes can be related to the deformation of the nucleus and its surface layer, and to various single-particle effects. At the same time, study of the hyperfine structure makes it possible to determine the values of the nuclear moments.

Measurements of the differences of the charge radii of neighboring isotopes ($\Delta\langle r^2\rangle$) are of greatest interest in the regions in which there are significant changes of the nuclear structure, for example, on going from spherical to deformed nuclei. One can discern here in the isotope shift in the region of $N = 90$ a distinct maximum due to a strong change in the nuclear shape.[1]

In this paper, we present the results of high-precision measurements of the isotope shifts, and the values of $\Delta\langle r^2\rangle$ deduced from them for the Nd, Sm, and Gd isotopes with neutron numbers in the interval $82 \leqslant N \leqslant 96$, i.e., in the region just noted, in which the transition from spherical to deformed nuclei occurs. Although for this region of nuclei there have been many measurements of the mean square radii, some of which were made by optical methods,[2,3] they are not yet sufficient for detailed conclusions about the changes in the sizes and shapes of the nuclei as functions of the number of neutrons. Most of these measurements were made using the classical interference technique, which does not have the resolution or, therefore, the accuracy of the laser methods. Measurements, using tunable lasers, of the differences of the mean square radii have been made in recent years only for the isotopes of Sm.[4,5] In addition, because of the complexity of the optical spectra of these elements a sufficiently correct procedure for extracting the $\Delta\langle r^2\rangle$ values from the measured differences of the resonance frequencies does not yet exist.

EXPERIMENTAL METHOD

The experimental method that we used in our experiments has been described in detail in Refs. 6 and 7 and is based on the following.

1. The formation by means of a system of collimators of a parallel beam of the investigated atoms in a crucible heated by an electric current.

2. The resonance excitation of the atoms by laser light with tunable frequency ("Spactra-Physics" ring dye laser).

3. Detection of the laser-excited resonance fluorescence by a photomultiplier (FÉU-79) operated in the single photon counting regime and connected to a multichannel time analyzer synchronized with the laser frequency.

The laser beam intersects the atomic beam at an angle of 90°, in an interaction region measuring 6 mm^3. We list the basic parameters of the experiment.[6,7]

1. Line width 30 MHz of the resonance fluorescence.

2. Power up to 800 mW of the laser radiation in the line.

3. Divergence 53 mrad of the atomic beam in the region of intersection with the laser beam.

4. The efficiency (the ratio of the number of detected pulses at the peak of the given line to the number of atoms leaving the crucible) depends naturally on the properties of the investigated atoms and the employed optical transition as well as on the geometrical factors. For example, the efficiency measured in the case of Eu atoms for the transition $4f^7 6s^2 \rightarrow 4f^7 6s6p$ with wavelength 5765.20 Å is $5 \cdot 10^{-7}$ and includes the collimation ratio $5 \cdot 10^{-4}$, photon counting efficiency $5 \cdot 10^{-3}$, and fraction $2 \cdot 10^{-1}$ of atoms in the given quantum state. This last quantity is determined by the number of components of the fine and hyperfine splittings and by the probability of transition from the excited state to the initial state. For the investigated elements Nd, Sm, and Gd, this quantity is of the same order as for Eu.

5. The minimal detectable flux of investigated atoms from the crucible at temperature 1350 K and power 100 mW of the laser radiation was $5 \cdot 10^8$ sec^{-1}. It was determined by

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TABLE I. Characteristics of investigated transitions.

λ, Å	Transition	Configuration (percentage present)	Level, cm^{-1}
		$_{60}$Nd	
5729.29	$^5I_6 \rightarrow {}^5H_5$	$4f^46s^2 \rightarrow 4f^35d6s^2$ (?)	2367 → 19 816
5813.90	?	$4f^46s^2 \rightarrow$?	? ?
		$_{62}$Sm	
5746.50	$^7F_2 \rightarrow {}^7F_3$	$4f^66s^2 \rightarrow 4f^66s6p$ (?)	812 → 18 209
5779.26	$^7F_3 \rightarrow {}^5F_2$	$4f^66s^2 \rightarrow 4f^66s6p$ (35) $4f^55d6s^2$ (65)	1490 → 18 788
		$_{64}$Gd	
5746.36	$^9D_4 \rightarrow {}^9D_4$	$4f^75d6s^2 \rightarrow 4f^75d6s6p$ (50)	533 → 17 931
5751.88	$^9D_2 \rightarrow {}^9F_2$	$4f^75d6s^2 \rightarrow 4f^75d6s6p$ (62)	0 → 17 381
5791.38	$^9D_4 \rightarrow {}^9D_3$	$4f^75d6s^2 \rightarrow 4f^75d6s6p$ (82)	533 → 17 795
5802.92	$^9D_4 \rightarrow$?	$4f^75d6s^2 \rightarrow$?	533 → 17 761

the level of the background from the optical emission of the crucible and the scattered laser radiation.

The measurements were usually made in the temperature range 1000–1400 K and at a power of the laser radiation of ~1 mW. To obtain accurate values of the differences of the resonance frequencies, the linearity of the frequency scale during scanning was tested. This was done by feeding into the analyzer a series of frequency marks synchronously with the optical spectra. The source for these was a confocal Fabry-Perot etalon with constant 1500 MHz.

EXPERIMENTAL RESULTS

Table I gives the characteristics of the optical transition in Nd, Sm, and Gd for which measurements of the isotope shifts have been made (terms, configurations, and energies of the initial and final states).[8] Examples of measured spectra for each of the elements are given in Fig. 1. In all the experiments, samples in the form of pure metals with natural isotopic composition of the elements were used. The spectra exhibit peaks corresponding to all even-even isotopes, and components of the hyperfine structure of the odd isotopes. To identify these components, measurements were also made with a sample enriched with ^{155}Gd to 90.7% (the corresponding spectrum is shown in the insert in Fig. 1). The centroid of the components of the hyperfine structure was determined by numerical integration. It can be seen from Fig. 1b that the hyperfine splitting of the line with wavelength 5746.5 Å in Sm is very small. It does not exceed 100 MHz, and the components of the hyperfine structure are not resolved. Therefore, the isotope shifts for the even and odd Sm isotopes were determined from the positions of the centroids of the corresponding peaks.

The measured isotope shifts for all the investigated optical lines are given in Table II. The error in their measurement was usually 2–15 MHz. Only in the case of ^{157}Gd were the positions of the components of the hyperfine structure determined with a somewhat greater error, and this reflected on the accuracy of the isotope shifts between this isotope and the neighboring even-even isotopes.

The compatibility of the obtained values of the isotope shifts was tested by means of King's graph.[2,9] In this graph, the scales of the axes the values of the modified isotope shifts for the compared pair of optical transitions:

$$\Delta\nu_{\text{mod}}^{A,A'} = \Delta\nu_{\text{exp}}^{A,A'} \frac{AA'(A_0'-A_0)}{A_0A_0'(A'-A)}, \quad (1)$$

where A, A' and A_0, A_0' are, respectively, the mass numbers of the measured and reference isotopes. These values of the isotope shifts must lie on a straight line, and this is indeed observed. In the case of the Gd isotopes, for which the isotope shifts were measured for four transitions, a more de-

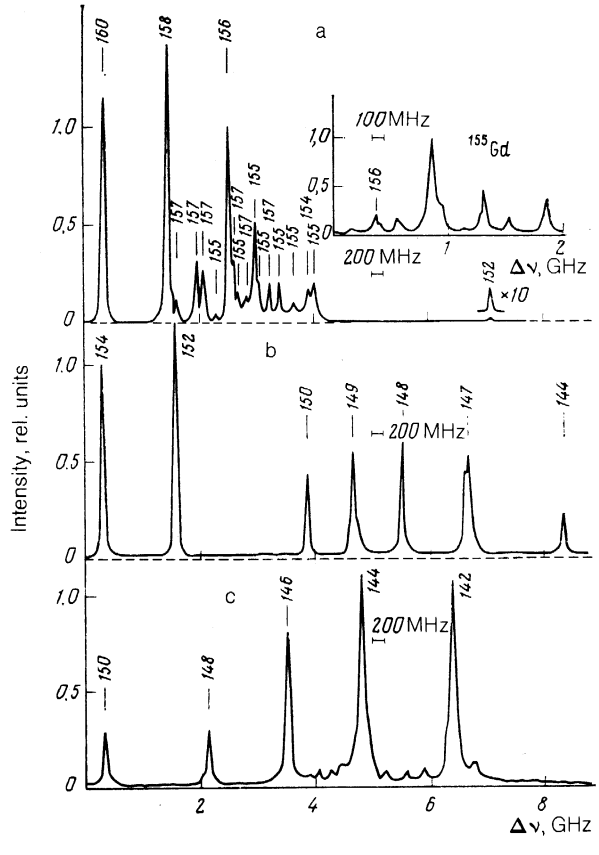

FIG. 1. Dependence of the number of resonantly scattered photons on the frequency of the laser radiation: a) Gd, $\lambda = 5791.38$ Å, transition $4f^75d\,6s^2\,{}^9D_4^0 \rightarrow 4f^75d\,6s6p\,{}^9D_3$; b) Sm, $\lambda = 5746.50$ Å, transition $4f^66s^2\,{}^7F_2 \rightarrow 4f^66s6p\,{}^7F_3^0$; c) Nd, $\lambda = 5813.90$ Å.

TABLE II. Isotope shifts in Nd, Sm, and Gd for different optical transitions ($\Delta\nu_{ex} = \nu^{A'} - \nu^{A}$).

A	A'	$\Delta\nu_{exp}^{A,A'}$, MHz			
		$_{60}$Nd		$_{62}$Sm	
		5729.29 Å	5813.90 Å	5846.50 Å	5779.26 Å
142	144	574(3)	−1457(5)	–	–
144	146	504(5)	−1339(4)	–	–
144	148	–	–	−2804(10)	1624(8)
146	148	619(4)	−1501(5)	–	–
148	150	1040(9)	−2056(8)	−1656(7)	1030(15)
150	152	–	–	−2307(6)	1662(12)
152	154	–	–	−1235(4)	706(10)
147	148	–	–	−831(7)	523(1)
148	149	–	–	−512(5)	231(1)

A	A'	$\Delta\nu_{corr}$, MHz			
		$_{64}$Gd			
		5746.36 Å	5781.88 Å	5791.38 Å	5802.92 Å
152	154	−3262(15)	−3290(9)	−3197(10)	−3329(10)
154	156	−1440(9)	−1438(6)	−1402(6)	−1486(6)
156	158	−1092(9)	−1082(5)	−1059(5)	−1126(5)
158	160	−1124(9)	−1115(5)	−1095(5)	−1158(5)
155	156	−725(6)	−740(5)	−722(5)	−764(4)
156	157	−221(12)	−204(9)	−241(16)	−227(9)

tailed analysis of the experimental results was made. A program based on the least-squares method was used to compare all possible combinations of pairs of optical transitions. For each pair of transitions, the isotope shifts lying on a straight line (smoothed isotope shifts) were calculated. It can be seen from Fig. 2 that the difference between the measured and smoothed values of the isotope shifts for the even-even isotopes does not exceed the limit of the errors, but for the odd isotopes the difference is larger. Further, from the smoothed values of the isotope shifts the so-called corrected isotope shifts[10] were calculated as the weighted means of all the smoothed values. These corrected isotope shifts are given in Table II and were used subsequently for all calculations in the case of the Gd isotopes.

DETERMINATION OF THE MEAN SQUARE CHARGE RADII

The theory of isotope shifts has been fairly well developed and has been described in a number of studies.[2,3] In accordance with this theory, the isotope shift is determined by two factors:

$$\Delta\nu_{i,\,exp}^{A,A'} = \Delta\nu_{i,\,fs}^{A,A'} + \Delta\nu_{i,\,ms}^{A,A'}, \qquad (2)$$

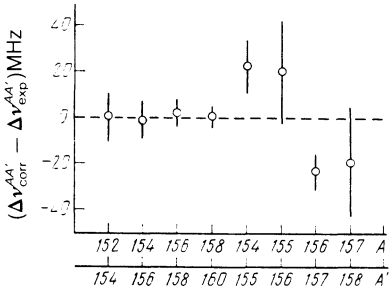

FIG. 2. Difference between the corrected and measured values of $\Delta\nu$ for different pairs of Gd isotopes, $\lambda = 5751.88$ Å.

where the subscripts fs and ms denote, respectively, the field shift and mass shift,

$$\Delta\nu_{i,\,fs}^{A,A'} = F_i\lambda^{A,A'}, \qquad (3)$$

$$F_i = E_i f(Z); \qquad (3')$$

here, E_i and $f(Z)$ are, respectively, the electron and nuclear factors, which are discussed below. The quantity $\lambda^{A,A'}$, includes the change in the parameters of the radial charge distribution of the nucleus:

$$\lambda^{A,A'} = \Delta\langle r^2\rangle^{A,A'} + \frac{C_2}{C_1}\Delta\langle r^4\rangle + \frac{C_3}{C_1}\Delta\langle r^6\rangle, \qquad (4)$$

where the C_i reflect the contribution of the moments of different orders. This contribution is almost independent of the principal quantum number of the electron shell,[11] and therefore it is the same for the optical transitions and the K x rays. In the latter case, the contribution of the moments of high orders ($n \geqslant 4$) is small[3,11]; usually, it is less than the error in the determination of the values of $\lambda^{A,A'}$ from the optical transitions. Therefore, to sufficient accuracy we can assume that

$$\Delta\nu_{i,\,fs}^{A,A'} = E_i f(Z)\Delta\langle r^2\rangle^{A,A'}. \qquad (5)$$

The mass shift includes the normal mass shift (nms) and the specific mass shift (sms):

$$\Delta\nu_{i,\,ms}^{A,A'} = \frac{A'-A}{AA'}(M_{i,\,nms} + M_{i,\,sms}) \qquad (6)$$

$$M_{i,\,nms} = 5.487\cdot 10^{-4}\nu_i\,[\text{MHz}]. \qquad (7)$$

For the transitions we investigated and for $A' - A = 2$ the normal mass shift is 22–25 MHz. Thus, the procedure for determining the differences of the mean square charge radii from the measured isotope shifts reduces to finding the constant M_{sms} and to calculation of the factors E_i and $f(Z)$. We discuss below, the procedure of this analysis.

1. Specific mass shift

The specific mass shift is due to the influence of the correlated motion of the electrons on the recoil energy of the nucleus. Although, as can be seen from the expression (6), this shift rapidly decreases with increasing mass number of the nucleus, in a number of cases its value can be significant (tens of times greater than the normal mass shift). The calculations of the specific shift are in only qualitative agreement with the experimental data. For $ns^2 \rightarrow nsnp$ transitions,[2]

$$\Delta\nu_{i,\,\mathrm{sms}}^{A,A'} = (0 \pm 0.5)\,\Delta\nu_{i,\,\mathrm{nms}}^{A,A'} . \qquad (8)$$

Usually, the specific mass shifts are determined experimentally by means of the King's graph considered above—the modified isotope shifts for the investigated optical transition are plotted along one of the axes and along the other the modified shifts for a transition with known value of the specific mass shift. However, in the case of the transitions that we investigated, such an approach can be used only for Sm; for the specific mass shift for the line 5746.50 Å can be determined from the value of the specific mass shift for the line 5779.26 Å, which was obtained with sufficient accuracy in Ref. 4. For Gd, the specific mass shifts are unknown, and for Nd they are too inaccurate. Therefore, a different approach was used, namely, the ordinate in King's graph was taken to be the modified isotope shift for the K x rays and/or the x rays of the mesic atoms, for which the mass-shift corrections can be calculated with good accuracy. The abscissa intercept of King's line gives the mass shift of the investigated optical line. For the K x rays we used data on the isotope shifts (ΔE_x) from the review of Ref. 12, and for the spectra of the mesic atoms the isotope shifts ΔE_μ were taken from Refs. 13–15 for Nd, Sm, and Gd, respectively.

It should be noted that the use of the isotope shifts from the spectra of the mesic atoms cannot be regarded as sufficiently well justified in the general case, since they reflect somewhat different parameters of the nuclear charge distribution. Criteria for the applicability of these isotope shifts are discussed in Refs. 16 and 17.

The independent determinations of the specific mass shifts (by the least-squares method) from the data on the K x rays and from the spectra of the mesic atoms lead to appreciable differences in their values (see the dashed lines in Figs. 3a and 3b). Usually, these differences are within the errors, but the errors are themselves large. Therefore, it would appear to be more correct to determine the specific mass shifts from the King's graph, since it includes data on the K x rays and the mesic atoms. This was done by the least-square method with allowance for the errors in the values of ΔE_x and ΔE_μ and subject to the additional condition that the two straight lines have a common abscissa intercept (Figs. 3a and 3b). Then the errors in the values of the specific mass shifts determined in this manner are decreased. The specific mass shifts, and also the field shifts needed to determine $\Delta\langle r^2\rangle$, are given in Table III.

2. Electron factor E_i

This factor determines the change in the total nonrelativistic density of the electron charge at the point of the nucleus, $\Delta|\psi(0)|^2$, for the considered optical transition (i):

$$E_i = (\pi a_0^2/Z)\,\Delta|\psi(0)|_i^2, \qquad (9)$$

where Z is the atomic number of the element, and a_0 is the Bohr radius. In the special case of the $ns^2 \rightarrow nsnp$ transitions, the simple theory of Ref. 2 gives the expression

$$\Delta|\psi(0)|^2_{ns^2\rightarrow nsnp} = -\gamma\,|\psi(0)|_{ns}^2, \qquad (10)$$

where $|\psi(0)|^2_{ns}$ is the density of the electron charge at the point of the nucleus for the configuration of the corresponding singly ionized atom. It can be calculated by means of the Goudsmit-Fermi-Segrè formula[18]:

$$|\psi(0)|_{ns}^2 = \frac{ZZ_a^2}{\pi a_0^3 n^{*3}}\left(1 - \frac{d\sigma}{dn}\right), \qquad (11)$$

where Z_a is the effective charge of the atomic core (in our case $Z_a = 2$), n and n^* are respectively the principal and effective quantum numbers, and σ is the quantum defect. The values of n^* and $d\sigma/dn$ for the elements we investigated can be calculated by using the data given in Ref. 19. These calculations gave the following values of $|\psi(0)|^2_{6s}$ in units of $Z/\pi a_0^3$: 0.453 (for Gd), 0.382 (for Sm), and 0.365 (for Nd).

Hartree-Fock calculations in both the nonrelativistic[20] and relativistic[21] approximations showed that the quantity γ in the expression (10) is almost constant in the region of the

TABLE III. Parameters needed to calculate $\Delta\langle r^2\rangle$.

Isotope	λ, Å	$\Delta\nu_{\mathrm{sms}}^{A,A'}$, MHz	$\Delta\nu_{6s}^{A,A'}$, MHz	$f(Z)$ GHz/Fm^2	E_i exp.	E_i cal.
^{148,150}Nd	5729.29	−500(60)	1510(60)	17.70	0.215(17)	*
	5813.90	30(70)	−2110(70)	–	−0.299(22)	0.266
^{144,148}Sm	5746.50	−110(200)	−2750(200)	19.41	−0.273(20)	−0.279
	–	−80(50)	−2780(50) **	–	–	–
	5779.26	−950(200)	−2520(200)	–	0.244(20)	*
^{158,160}Gd	5746.36	−70(40)	−1080(40)	23.24	−3.308(22)	−0.326
	5751.88	−30(40)	−1110(40)	–	−0.318(23)	−0.326
	5791.38	−70(40)	−1050(40)	–	−0.296(23)	*
	5802.92	−60(40)	−1120(40)	–	−0.319(24)	−0.326

*Not calculated because of the large and unknown configuration mixing.
**The value of $\Delta\nu_{\mathrm{sms}}^{A,A'}$ was determined from optical transitions.[4]

rare earth elements for transitions in which the number of f electrons does not change and is 0.72–0.73. The values of the electron factor ($E_{i,\mathrm{cal}}$) obtained as a result of the calculations of $|\psi(0)|^2_{ns}$ and γ are given in Table III.

In these calculations it was assumed that the levels between which the optical transition takes place belong to pure electron configurations, and that the contribution of the p electrons can be ignored. However, these assumptions are often not justified, particularly for elements with complicated optical spectra, and this seriously limits the accuracy of the calculations of $|\psi(0)|^2$. As can be seen from Table I, for the majority of the optical transitions that we investigated there is strong configuration mixing.

To estimate the degree of reliability of these calculations of E_i, it is necessary to compare them with the analogous quantities obtained from experimental data. The latter can be found from the slopes of the lines in the King's graphs discussed above (Fig. 3). Indeed, if equal values of $\Delta\langle r^2\rangle$ are determined from the isotope shifts for the K x rays and the optical transitions, the slope of King's line determines the ratio of their factors F_i from the expressions (2)–(4). Since the factors F_i can be calculated for the K x rays fairly accurately, it is possible to obtain from their ratio the values of F_i and therefore the factors E_i for the optical transitions ($E_{i,\mathrm{exp}}$). These values of $E_{i,\mathrm{exp}}$ are given in Table III. It can be seen that there is a comparatively good agreement between the calculated and experimental values of E_i (the difference is less than 10%) for all optical transitions in Gd and for the transition with $\lambda = 5746.50$ Å in Sm.

In the same way it is possible to determine the values of $E_{i,\mathrm{exp}}$ using the isotope shifts from the spectra of the mesic atoms, provided the criteria of validity noted above are taken into account.[16,17] For all the investigated transitions, the difference between the values of $E_{i,\mathrm{exp}}$ obtained from the spectra of the K x rays and the mesic atoms does not exceed a few percent.

3. Nuclear factor $f(Z)$

The factor $f(Z)$ includes corrections to the electron wave function for relativistic effects and for the finite size of the nucleus. The expression for it has the form

$$f(Z)=\frac{5}{2}\bar{A}^{1/3}C^{A,A'}\left(\frac{R_{\mathrm{eq}}}{r_0\bar{A}^{1/3}}\right)^{2\sigma-2}[r_0^2(A'-A)]^{-1}, \tag{12}$$

where

$$\bar{A}=(A+A')/2, \quad r_0=1{,}2\ \mathrm{Fm}, \quad R^2_{\mathrm{eq}}={}^5/_3\langle r^2\rangle,$$

$$\sigma=(1-\alpha^2Z^2)^{1/2}.$$

α is the fine structure constant, and $C^{A,A'}$ is the theoretical constant of the isotope shift for a uniformly charged sphere of radius $R=r_0A^{1/3}$. The numerical values of $C^{A,A'}$ were taken from Ref. 22 and subjected to the relativistic correction $(n/N)^3$ (N is the "effective" relativistic principal quantum number) given in Ref. 23. The values of $\langle r^2\rangle^{1/2}$ for Nd, Sm, and Gd were obtained from experiments on elastic scattering of electrons.[24] The values of $f(Z)$ calculated by means of the expression (12) are given in Table III.

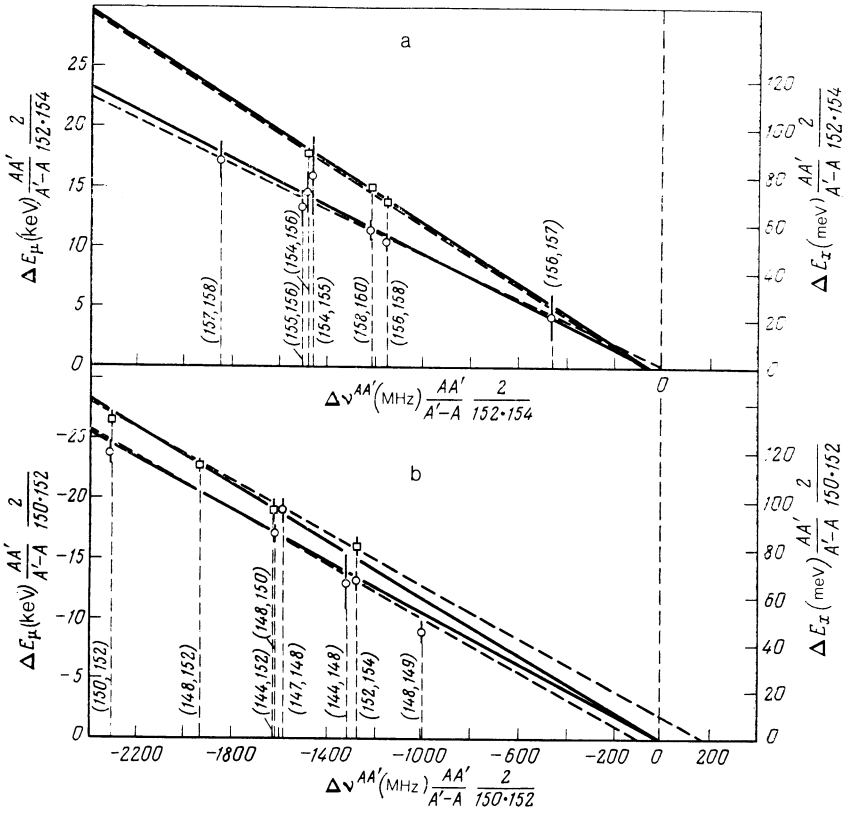

FIG. 3. King's graphs: a) Gd, $\lambda = 5746.36$ Å; b) Sm, $\lambda = 5746.50$ Å. The modified values of the optical isotope shifts are plotted along the x axis, and along the y axis the isotope shifts for the mesic atoms (open squares) (left-hand scale) and for the K x rays (open circles) (right-hand scale).

TABLE IV. Relative, λ_{rel}, and absolute $\Delta\langle r^2\rangle$, differences of the mean square radii of the isotopes of neodymium, samarium and gadolinium.

Nd				Sm		Gd			
A	A'	λ_{rel}	$\Delta\langle r^2\rangle^{A,A'}$, Fm^2	λ_{rel}	$\Delta\langle r^2\rangle^{A,A'}$, Fm^2	A	A'	λ_{rel}	$\Delta\langle r^2\rangle^{A,A'}$, Fm^2
142	144	0.718(9)	0.322(19)	–	–	152	154	2.97(4)	0.43(2)
144	146	0.662(10)	0.297(18)	–	–	154	156	1.291(6)	0.188(10)
144	148	–	–	2.00	0.51(3)	156	158	0.969(4)	0.143(8)
146	148	0.734(8)	0.329(19)	–	–	158	160	1.00	0.146(8)
148	150	1.00	0.45(3)	1.183(3)	0.304(16)	155	156	0.662(4)	0.097(5)
150	152	–	–	1.651(5)	0.42(2)	156	157	0.187(8)	0.027(2)
152	154	–	–	0.880(2)	0.226(12)				
147	148	–	–	0.593(3)	0.152(8)				
148	149	–	–	0.364(3)	0.093(5)				

DIFFERENCES OF THE MEAN SQUARE CHARGE RADII $\Delta\langle r^2\rangle$

From the experimentally measured isotope shifts (Table II) and the calculated parameters (Table III) we obtained the relative and absolute values of the differences $\Delta\langle r^2\rangle$ of the mean square radii by means of the expression (5); they are given in Table IV. The relative values (λ_{rel}) are the ratios of the field shifts ($\Delta\nu_{fs}^{A,A'}$) for the corresponding pairs of isotopes. In this case, the comparatively large error in the values of $\Delta\nu_{sms}$ and, therefore, $\Delta\nu_{fs}$ have little influence on the accuracy of λ_{rel}. Therefore, the error of the relative values of the differences of the charge radii, averaged over all the investigated optical transitions, does not exceed 2–3%.

To obtain the absolute values of $\Delta\langle r^2\rangle$ we used in addition to $\Delta\nu_{fs}$ the calculated values of the parameters E_i and $f(Z)$. Naturally, it is necessary to choose optical transitions for which the Goudsmit-Fermi-Segrè approximation holds or for which there is good agreement between the experimental and calculated E_i. This is the case for the transitions with wavelengths 5746.36, 5751.88, and 5802.92 Å for Gd, and with 5746.50 Å for Sm. In the case of Nd, the differences between $E_{i,\,exp}$ and $E_{i,\,cal}$ exceed the limits of the errors for both the investigated transitions. However, for the transition with $\lambda = 5813.90$ Å the difference is only 12%, and the value of $\Delta\nu_{sms}$ does not contradict the expression (8) for the $ns^2 \rightarrow nsnp$ transitions. The data for this transition were therefore used to determine $\Delta\langle r^2\rangle$.

The values of $\Delta\langle r^2\rangle$ obtained in this manner are given in Table IV. The error in the values of $\Delta\langle r^2\rangle$ includes the inaccuracy in the calculation of the factor E_i (estimated at 5%) and the experimental error in the field shift $\Delta\nu_{fs}^{A,A'}$ (in it, the main contribution is made by the specific mass shift) and is 6–8%.

It is of interest to compare our values of $\Delta\langle r^2\rangle$ with the data determined elsewhere by other methods (measurements of the spectra of the K x rays and of the mesic atoms). As we have already noted, the main contribution to our $\Delta\langle r^2\rangle$ values are not made by the error in the measurement of $\Delta\nu$ but by the uncertainties of the calculation, these being practically the same for all pairs of compared isotopes. These uncertainties influence the accuracy of the $\Delta\langle r^2\rangle$ values obtained by the other methods, though to a lesser degree. Therefore, instead of comparing $\Delta\langle r^2\rangle$ for each pair of isotopes, it is more illustrative to consider the averaged differences of the values of $\Delta\langle r^2\rangle$ for our investigation and the other studies:

$$\delta = \frac{1}{n}\sum_{i=1}^{n}\frac{\Delta\langle r^2\rangle_{i,a} - \Delta\langle r^2\rangle_{i,x}}{\Delta\langle r^2\rangle_{i,a}}$$

(the subscript a labels our data, and x the other). These quantities δ, and also the mean relative errors ε of the measurements for each of the methods, are given in Table V. It can be seen that in all cases the differences do not exceed the combined errors. This enables us to conclude that in the optical spectra of the elements that we investigated—Nd, Sm, and Gd—there are transitions for which the calculations of $\Delta\langle r^2\rangle$ for pure $ns^2 \rightarrow nsnp$ transitions are valid. In the case of Sm and, apparently, Nd these must be transitions with a small admixture of other configurations. These obviously

TABLE V. Comparison of obtained values of $\Delta\langle r^2\rangle$ with the results of other studies (references given in square brackets).

Element	Method						
	ours	K x rays		mesic atoms		optical transitions	
	ε_a, %	δ, %	ε_x, %	δ, %	ε_x, %	δ, %	ε_x, %
Nd	5.9	+14.5	9 [12]	+12.2	7 [13]	+12.8	4.5 [2]
Sm	9.8	+1.3	7 [12]	−5.0	5 [14]	+4.5	5.5 [2] 5.2 [4]
Gd	5.7	−7.0	17 [12]	−4.5	10 [15]	+8.1	23 [2]

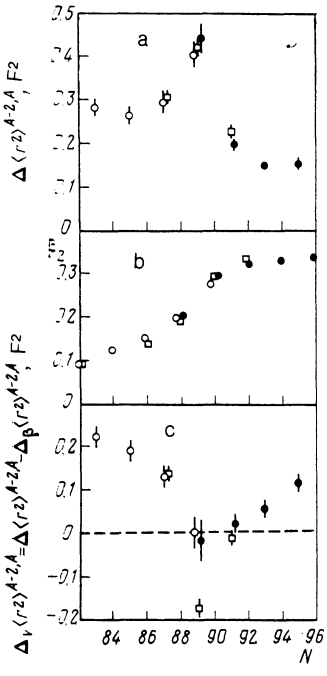

FIG. 4. Dependence of the difference of the mean square radii and the deformation parameter (β_2) on the number of neutrons in the nucleus: the open circles for Nd, the open squares for Sm, and the black circles for Gd.

include the transitions we used with $\lambda = 5813.90$ Å in Nd and $\lambda = 5746.50$ Å in Sm. In the case of Gd, the calculations are valid for transitions in which the number of f electrons does not change. For these, there may be large admixtures of other configurations.

DISCUSSION OF THE RESULTS

The good agreement between the values of $\Delta\langle r^2\rangle$ obtained by the different methods enables us to follow in detail the change of the mean square radii in this region of nuclear mass numbers ($N = 82$–96).

In Fig. 4 we have plotted the difference of the mean square radii of two neighboring even-even nuclei as a function of the number of neutrons in them. Two features can be noted.

1. The almost identical values of $\Delta\langle r^2\rangle$ for the nuclei of the different elements with the same number of neutrons.

2. The well-known jump in the values of $\Delta\langle r^2\rangle$ between the neutron numbers 88 and 90, where the shape of the nucleus changes from spherical to spheroidal.

Although the change in the mean square charge radius of the nucleus is due to numerous details of the nuclear structure, we can in a first approximation identify two main components. One of them, $\Delta\langle r^2\rangle_\nu$, is associated with the increase in the volume of the nucleus on the addition of neutrons, while the other, $\Delta\langle r^2\rangle_\beta$, is due to the change in the deformation of the nucleus:

$$\Delta\langle r^2\rangle = \Delta\langle r^2\rangle_\nu + \Delta\langle r^2\rangle_\beta, \tag{13}$$

$$\Delta\langle r^2\rangle_\nu = \langle r_0^2\rangle \frac{2\Delta A}{A}\chi, \tag{14}$$

$$\Delta\langle r^2\rangle_\beta = \langle r_0^2\rangle \frac{5}{4\pi}\sum \Delta(\beta_i^2), \tag{15}$$

where χ is the compressibility factor of nuclear matter, and β_i are the deformation parameters of the various orders (quadrupole, octupole, etc.). At the same time, it is also necessary to take into account both the static deformation and the dynamic deformation due to the vibrations of the nuclear surface.

In the investigated region of nuclei, the values of the deformation parameters are fairly well known (they have been obtained from the lifetimes of levels, Coulomb excitation, inelastic scattering, and the x-ray spectra of the mesic atoms). Although the absolute values of the β_i obtained from the different sources differ somewhat from each other (they depend on the chosen model and the parameters), the values of their differences ($\Delta\beta_i$) for neigboring even-even isotopes are not as sensitive to the method by which they are obtained and exhibit much smaller differences. In the region $86 \leqslant N \leqslant 92$, it is the quadrupole deformation parameter that changes most strongly (from 0.15 to 0.33). These values of β_2 obtained from the reduced probabilities in electric quadrupole transitions[25] are shown in Fig. 4b. The changes in the other deformation parameters (β_3, β_4) are much less ($\Delta\beta \leqslant 0.05$).

Using the known values of β_2 and the expressions (13)–(15), we can identify the change in the mean square radius associated with the increase in the volume of the nucleus on the addition of a pair of neutrons ($\Delta\langle r^2\rangle_\nu = \Delta\langle r^2\rangle_{\text{exp}} - \Delta\langle r^2\rangle_\beta$). It can be seen from Fig. 4c that the dependence of this quantity on the number of neutrons in the nucleus is by far not the one which follows from simple arguments (volume of the nucleus proportional to the number of neutrons). It is obvious that there are other factors as well that influence the mean square radius, for example, the thickness of the surface layer of the nucleus.

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[1]P. Brix and H. Kopfermann, Z. Phys. **126**, 344 (1949).
[2]K. Heilig and A. Steudel. At. Data Nucl. Data Tables **14**, 613 (1974).
[3]K. Heilig, Hyperfine Inter. 24–26 349, (1985).
[4]H. Brand, B. Seibert, and A. Steudel, Z. Phys. A **296**, 281 (1980).
[5]G. D. Alkhazov, É. E. Berlovich, H. Wagner, *et al.*, Preprint No. 1006 [in Russian], Leningrad Institute of Nuclear Physics (1984).
[6]Yu. P. Gangrskiĭ, K. P. Marinova, B. N. Markov, *et al.*, Izv. Akad. Nauk SSSR, Ser. Fiz. **49**, 2261 (1985).
[7]Yu. P. Gangrsky, I. Han Gyong, K. P. Marinova, *et al.*, Preprint E6-86-233 [in English], JINR, Dubna (1986).
[8]W. C. Martin, R. Zalubas, and L. Hagan, "Atomic energy levels. The rare-earth elements," Nat. Stand. Rev. Data Ser. (NSRDS)-NBS 60 U.S. GPO, Washington, D. C. (1978).
[9]W. H. King, J. Opt. Soc. Am. **53**, 638 (1963).
[10]P. Aufmuth, H. P. Clieves, K. Heilig, *et al.*, Z. Phys. A **285**, 357 (1978).
[11]E. C. Seltzer, Phys. Rev. **188**, 1916 (1969).
[12]F. Boehm and P. L. Lee, At. Data Nucl. Data Tables **14**, 605 (1974).
[13]E. R. Macagno, S. Bernow, S. G. Cheng, *et al.*, Phys. Rev. C **1**, 1202 (1970).
[14]R. J. Powers, P. Bareau, B. Bihoreau, *et al.*, Phys. Rev. A **316**, 295 (1979).
[15]D. Laubacher, Y. Tanaka, R. M. Steffen, *et al.*, Phys. Rev. C **27**, 1772 (1983).
[16]R. Engfer, H. Schneuwly, J. L. Vuilleumier, *et al.*, At. Data Nucl. Data Tables **14**, 509 (1974).

[17]D. L. Clark, M. E. Cage, D. A. Lewis, and G. W. Greenlees, Phys. Rev. A **20**, 239 (1979).
[18]H. G. Kuhn *Atomic Spectra* (Longmans, London, 1969).
[19]J. Sugar and J. Reader, J. Opt. Soc. Am. **55**, 1286 (1965).
[20]M. Wilson J. Phys. B **5**, 218 (1972).
[21]M. A. Coulthard, J. Phys. B **6**, 23 (1973).
[22]F. A. Babushkin, Zh. Eksp. Teor. Fiz. **44**, 1661 (1963) [Sov. Phys. JETP **17**, 1118 (1963)].
[23]D. Zimmermann, Z. Phys. A **315**, 123 (1984); A **321**, 23 (1985).
[24]E. Wesolowski, J. Phys. C **10**, 321 (1984).
[25]P. H. Stelson and L. Grodzins, Nucl. Data Tables **A1**, 21 (1965).

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