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EXPERIMENTAL OBSERVATION OF THE ISOTOPIC SHIFT OF THE $K_{\alpha 1}$ *X-RAY LINE OF MOLYBDENUM*

O. I. SUMBAEV and A. F. MEZENTSEV

A. F. Ioffe Physico-technical Institute, Academy of Sciences, U.S.S.R.

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The volume isotopic shift of the $K_{\alpha 1}$ x-ray line of the molybdenum isotopes has been observed by a method of successive introduction of the samples to be compared into the field of view of a Cauchois diffraction spectrometer used previously for the observation of the chemical shift of the K-series x-ray lines of comparatively heavy elements.[17,19] The differences in the energies of this line for $Mo^{92}O_3-Mo^{100}O_3$, $Mo^{94}O_3-Mo^{100}O_3$, and $Mo^{92}O_3-Mo^{94}O_3$ turned out to be respectively 0.030 ± 0.005 eV, 0.027 ± 0.008 eV, and 0.001 ± 0.005 eV. The relative sensitivity of the method is thus 0.00003%. The obtained results are in good agreement with theoretical predictions[21]:

$$\Delta E(92-100)_{\text{exp}} / \Delta E(92-100)_{\text{theoret}} = 1.15 \pm 0.19.$$

A comparison of the experimental errors with the theoretical estimates of the effect for a number of other elements[22] indicates that the isotopic shift of x-ray lines can be used as a new method of investigating nuclear properties. Its main advantage over the optical isotopic shift is apparently the possibility of a more exact separation of the nuclear part of the effect because of the simpler and more exact account of the mutual influence of the electron shells of the atom.

INTRODUCTION

THE isotopic shift of the optical levels of an atom, more accurately the so-called volume isotopic shift of the optical levels, was first observed in 1919.[1] Somewhat later followed the theoretical description of this effect.[2-4] In the years that followed the isotopic shift served as one of the methods of investigating such nuclear properties as the compressibility of nuclear matter, the change in the equilibrium shape of nuclei, and shell effects. The present state of the experimental and theoretical aspects of this method has been presented in several review papers (see, for example, [5-7]).

The existence of an analogous effect in x-ray spectra was theoretically predicted at practically the same time,[8] and a series of unsuccessful attempts was undertaken to observe this effect experimentally (commonly in conjunction with a search of the line broadening of x-ray lines due to hyperfine structure).[9-12] An upper estimate of the magnitude of the effect of 0.2 eV was obtained in the latest of these works, consisting of the measurement of the position of the $K_{\alpha 1}$ line of the separated $Mo^{92,95,100}$ isotopes and its broadening for the odd Mo^{95} isotope. Subsequent theoretical consideration of the isotopic shift of the $K_{\alpha 1}$ line of Mo^{92-100} indicated that the sought effect cannot exceed 0.08 eV.[13]

A preliminary report of the first successful experiment of observing the isotopic shift of the x-ray line was published by Brockmeier, Boehm, and Hatch.[14] Utilizing the sharp increase of the effect for the heaviest nuclei (an extrapolation of the results of Wertheim and Igo[13] led to a magnitude of the effect of the order of 2 eV for the $K_{\alpha 1}$ line in $U^{233}-U^{238}$), the authors of this work used a two-meter DuMond focusing gamma spectrometer[15] whose relative precision of energy measurements amounted in the 100 keV range to about 1 eV. The measuring procedure was in general the usual procedure for this type of diffraction apparatus, consisting of measurements of distances between the right and left symmetrical positions of the line, first with the U^{233} source, and then with U^{238}. The result was:

$$E(K_{\alpha 1}^{233}) - E(K_{\alpha 1}^{238}) = +1.3 \pm 0.5 \text{ eV}.$$

In 1963–1964 we worked out a method for measuring small energy shifts of x-ray or gamma

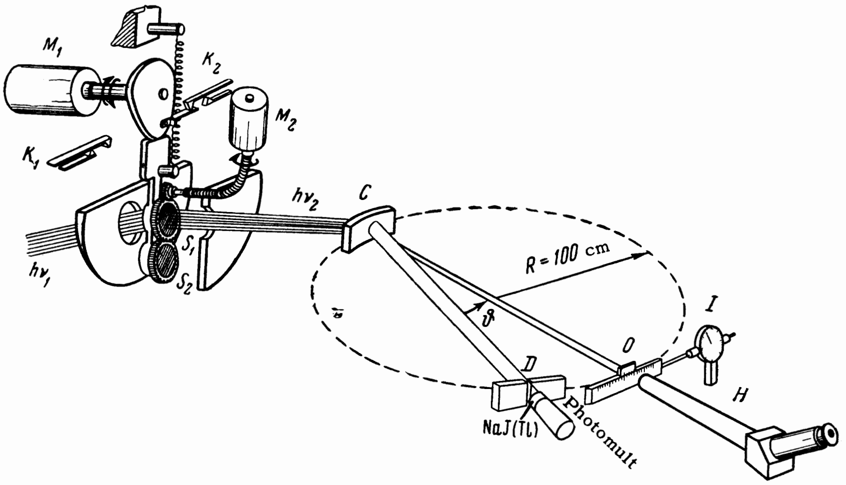

FIG. 1. Schematic diagram of the setup.

lines based on a successive introduction of the sources to be compared into the field of view of a focusing Cauchois gamma spectrometer with scintillation recording.[16,17] We planned a program of investigation of a series of effects in the range $\Delta E/E \approx 10^{-5}$—$10^{-7}$, such as the chemical shift of the K-series x-ray lines of relatively heavy elements, the isotopic, and possibly the isomeric shift.[17]

Before searching for the isotopic shift of x-ray lines, it was decided to clarify whether the generally accepted viewpoint that for elements with $Z \geq 29$ there is no chemical shift of the K_α x-ray lines[18] which could interfere with the search for the isotopic shift is correct. It turned out that these misgivings were justified. For Mo—MoO_3 and Sn—SnO_2 we observed a chemical shift amounting to 0.2 eV,[17-19] and exceeding the sought effect (see below) by more than a factor of six.

In these experiments we confirmed that the method is suitable for measurements of small shifts down to 0.01 eV (for lines with an energy ~ 20 keV). Thus there appeared a real possibility of observing the isotopic shift for most elements (with $Z \gtrsim 40$).

As the first object of investigation we chose the $K_{\alpha 1}$ line of the isotopes $Mo^{92,94,100}$.

DESCRIPTION OF THE METHOD

A schematic diagram of the setup is shown in Fig. 1. The primary beam $h\nu_1$ (20 mA, 200 kV) emitted by an x-ray tube with a molybdenum target falls on the source S and excites the characteristic fluorescence radiation $h\nu_2$. The voltage on the high-voltage transformer of the x-ray unit was stabilized by a ferroresonant stabilizer, and the tube current by an electronic stabilizer with plate-cathode feedback filament. The beam was diffracted after passing through a diaphragm by a 1-mm thick curved quartz crystal C [diffraction by the (13$\bar{4}$0) planes], converged into a narrow (~ 0.1 mm) focus at a distance of 2 m from the crystal, and detected past the receiving slit D with a scintillation counter with NaI(Tl) crystal.

During the measurement of the line the detector was stationary. The crystal was rotated with the aid of a two-meter level CO, the displacement of which was measured by means of the indicator I with a precision of up to 2 μ or less precisely with a microscope H.

The samples were thin-walled (0.02 mm), saucers 13-mm in diameter and ~ 0.5 mm high, pressed from aluminum foil and filled with molybdenum oxide. The working portion of the sources was bounded by a fixed diaphragm 11 mm in diameter, and was therefore perfectly identical for both compared samples. The change of samples was carried out with the aid of a cam gear driven by the motor M_1.

The measurement procedure and processing of the results were described in our previous paper.[17] In essence, the compared samples are successively introduced into the field of view of the instrument and the counting channels which accumulate the pulses from the scintillation counter are simultaneously switched. The curved crystal remains stationary. After several switches and exposures (in these experiments we employed a cycle of four one-minute intervals), ensuring the accumulation of the required statistics from each source, the instrument is moved to the next position in ϑ (usually by 2″), and the following pair of points is measured. Thus the whole double profile of the line

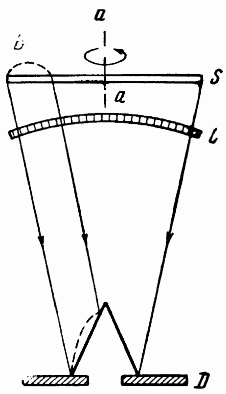

FIG. 2. Illustration of the method used to remove the effect of aberration.

is successively measured. The calculation of the line shifts is carried out analytically on the basis of the measured differences in the number of pulses in the channels of the first and second sources, and obtained from a plot of the widths and heights of the lines according to the relations[17]:

$$x_0{}^i = \frac{2\Delta n_1{}^i X_1{}^*}{n_0} \quad \text{or} \quad x_0{}^i = \frac{2\Delta n_r{}^i X_r{}^*}{n_0}, \tag{1}$$

$$\bar{x}_0 = \frac{2}{n_0} \sum p^i \Delta n^i X^* \Big/ \sum p^i, \tag{2}$$

$$p^i_{1(r)} \sim 1/n^i X^{*2}, \tag{3}$$

$$\sigma_{\text{res}} = \pm \left[p^i (\bar{x}_0 - x_0{}^i)^2 / (n-1) \sum p^i \right]^{1/2}, \tag{4}$$

where x_0^i is the shift of the profile of one of the lines relative to the other, calculated from the difference in the number of pulses from the first and second source at the i-th value of the angle ϑ; $\bar{x}_0$ is the mean shift of the profile of one of the lines relative to the other; n_0 is the height of the profile (more accurately, the height of the point of intersection of the straight lines which are the extensions of the line slopes); $X^*_{l(r)}$ is the half-width of the line profile at a height $n_0/2$ (in the case of an asymmetric profile the values X^* for the left and right parts X^*_l and X^*_r can differ somewhat); n^i is the number of pulses accumulated at the i-th point from one of the sources, Δn^i is the difference in the number of pulses accumulated from the first and the second source at the i-th point; σ_{res} is the mean-square error of $\bar{x}_0$.

Such a pair of points line profiles, containing usually 14—18 working points (in the calculation we used only points lying on the straight-line slopes of the line profile) is referred to throughout as a series.

The counting rate during these measurements amounted to ~ 8000 pulses/min at the line maximum, the background was 900 min^{-1}, the line width at half the height was about 300 μ (~ 9 eV).

The position of the line is determined by the angle ϑ (Fig. 1) between the reflecting planes and the crystal-detector direction; to a first approximation, it does not depend on the position of the source. This in fact is the basis of the method used. However, the presence of apparatus aberration can lead to aberration shifts of the line. Indeed, let S on Fig. 2 be the source, C the curved crystal, and D the receiving slit of the detector with a triangular line image. We assume that the source brightness B varies because of nonuniformity of the structure or of the thickness of the source. In the presence of apparatus aberration different portions of the crystal, and consequently of the source, contribute to the intensity of various portions of the line profile. The nonuniformity of B leads under these circumstances to an asymmetric line-profile distortion which can be perceived in the processing as a shift of the profile. To remove, or in any case decrease appreciably the aberrational shifts, the sources were continuously rotated about axes perpendicular to their surfaces and passing through their centers (axis a—a in Fig. 2). This rotation, at 30 rpm, was produced by the motor M_2 (Fig. 1) through a flexible shaft and a system of gears.

An essential condition for the absence of aberrational shifts is a decrease of the brightness nonuniformity of the sources. In the transmission mode of source operation, the variation of the intensity of the fluorescence radiation emitted in the direction of the crystal (perpendicular to the surface of the sample in our geometry) with the source thickness had a maximum, such as shown in Fig. 3 for molybdenum-oxide samples (natural isotopic composition). On the ordinate axis we plotted the counting rate at the maximum of the $K_{\alpha 1}$ line, and along the abscissa the thickness of the samples in

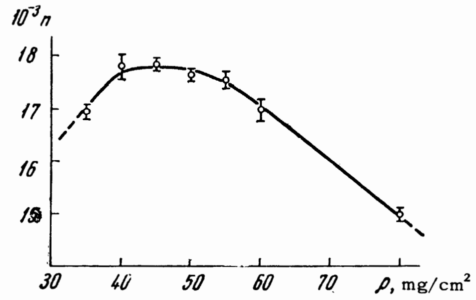

FIG. 3. Dependence of the intensity of the K_{α_1} molybdenum line on the thickness of the source.

Table I. Isotopic composition of the samples (per cent)

A	92	94	95	96	97	98	100
Mo^{92}	94.7	1.5	0.9	1.0	0.6	1.0	0,2
Mo^{94}	1.8	91.2	2.1	1.4	1.1	2.4	0.1
Mo^{100}	0.1	0.6	0.7	1.0	0.8	3.1	93.8

mg/cm^2 (of molybdenum). It is obviously convenient to choose a source thickness corresponding to the maximum of the intensity. This will not only ensure a maximum counting rate on the line but the unavoidable variations of the source density (thickness) will lead to minimum variations of the intensity emitted by the various portions of the sample, i.e., to minimum brightness variations over the source area.

EXPERIMENTAL RESULTS

The isotopic composition of the samples is given in Table I. In the first experiment (below we shall refer to it as preliminary, as opposed to the main experiment) the $Mo^{92}O_3$ and $Mo^{100}O_3$ sources were prepared from commercial quality oxides of the separated isotopes, obtained by thermal decomposition of molybdenum nitrate. The $Mo^{94}O_3$ was obtained by burning of metallic molybdenum in oxygen. The oxide was ground in an agate mortar and was deposited into the sample saucers by settling out from a suspension in alcohol with a small admixture of shellac.

Eight series of measurements were taken for each of the pairs $Mo^{92}O_3-Mo^{100}O_3$ and $Mo^{94}O_3-Mo^{100}O_3$, seven for $Mo^{92}O_3-Mo^{94}O_3$, and six control series for two samples of $Mo^{100}O_3$. In all series we accumulated $\approx 16 \times 10^3$ pulses at the maximum of the $K_{\alpha 1}$ line of each isotope. The average shifts turned out to be

$$E(K_{\alpha 1}{}^{92}) - E(K_{\alpha 1}{}^{100}) \equiv \Delta E(92-100) = 0.034 \pm 0.007 \text{ eV};$$

$$\Delta E(94-100) = 0.047 \pm 0.011 \text{ eV};$$

$$\Delta E(92-94) = -0.015 \pm 0.008 \text{ eV};$$

$$\Delta E(100_{\mathrm{I}} - 100_{\mathrm{II}}) = -0.001 \pm 0.017 \text{ eV}.$$

The errors given are external rms errors calculated from the deviations of the results of the individual series from the general average.

The results of the preliminary experiment are plotted on Fig. 4. The point 92* was obtained by summing the algebraic values of the shifts ΔE (94–100) and ΔE (92–94). It coincides within the limits of the errors with the directly measured difference ΔE (92–100) (point 92 on Fig. 4a). This agreement is evidence of the internal consistency of the results. The control value $\Delta E(100_{\mathrm{I}}-100_{\mathrm{II}}) = 0$ is also satisfied sufficiently well. The position of the point 94 is puzzling. The shift between the 94 and 100 isotopes turns out to be considerably larger than the shift between the 92 and 100 isotopes, and the shift ΔE (92–94) has a sign opposite to that expected.

Analyzing the situation, we have assumed that the reason for this anomalous position of point 94 is an admixture of a chemical shift due to the somewhat different chemical state of the $Mo^{94}O_3$ sample. Samples of commercial purity obtained from $Mo^{92}O_3$ and $Mo^{100}O_3$ by the "wet" method may contain an admixture of $MoO_3 \cdot 2H_2O$; the $Mo^{94}O_3$ sample obtained by burning molybdenum in oxygen cannot contain such an admixture. The crystal structures of MoO_3 and $MoO_3 \cdot 2H_2O$ are very similar. The oxygen ions lie on the vertices of an irregular octahedron in whose center lies the molybdenum.[20] The presence of water in the layers between the octahedra in $MoO_3 \cdot 2H_2O$ leads to an appreciable decrease in the average oxygen-molybdenum distances in $MoO_3 \cdot 2H_2O$ compared with MoO_3 (2.03 Å in MoO_3 as compared to 1.87 Å in $MoO_3 \cdot 2H_2O$). The electron density in the region of the Mo ion should therefore be smaller in MoO_3 than in $MoO_3 \cdot 2H_2O$, and the x-ray lines of MoO_3 should be shifted towards the harder side relative to $MoO_3 \cdot 2H_2O$. Anhydrous molybdenum oxide can be obtained by distillation of commercial-type oxide in a stream of oxygen at ~ 700°C.[20] We prepared a source from commercial-type molybdenum oxide purified in this way (natural isotopic composition), and measured the chemical shift of the $K_{\alpha 1}$ line of this sample relative to the sample of the unpurified commercial-type oxide. The measurements yielded the following result:

$$E(K_{\alpha 1}{}^{\mathrm{pur}}) - E(K_{\alpha 1}{}^{\mathrm{com}}) = +0.021 \pm 0.016 \text{ eV}$$

The shift has the expected sign. The introduction

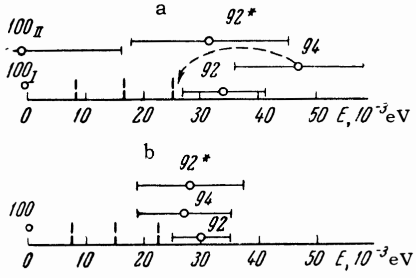

FIG. 4. Results of the preliminary (a) and main (b) experiments.

of such a correction (see the dashed arrow in Fig. 4a) shifts the point 94 into the region where it should be located.

The actual presence of the effect of the isotopic shift between $Mo^{92}O_3$ and $Mo^{100}O_3$ follows sufficiently convincingly from the results of this experiment. Nonetheless, wishing to confirm this result again, excluding at the same time the effect of chemical shifts, we repeated the experiment. The sources for the main experiment were prepared from the same molybdenum oxides as in the preliminary experiment, but purified by sublimation in a stream of oxygen at a temperature of ~ 700°C. As a result of the sublimation we obtained the characteristic colorless needle-shaped crystals of the anhydrous MoO_3, completely identical for all the three isotopes. The weights of MoO_3 were chosen such as to obtain in the samples the same number of molybdenum atoms per cm^2 as in a sample of natural oxide, corresponding to the maximum on the curve of Fig. 3. After being ground, the oxide powder was carefully and repeatedly mixed with 100 mg of teflon powder. The resulting mixture was poured into a press form and was pressed under a pressure of ~ 4000 atm into pellets 13 mm in diameter. The sublimation and pressing procedures were strictly identical for the samples of all three isotopes. In order to exclude the possibility that the results of the experiment be affected by possible chemical changes in the samples in the course of their irradiation with x rays, the third sample not belonging to the given pair was also placed in the beam so that the dose to which all three samples were exposed remained equal.

During the preliminary experiment attention was drawn to the possible presence of another effect which would distort the results. The distance between succeeding points amounted to $20\,\mu$ on the indicator scale ($\sim 2''$ in ϑ). The cycle was such that the accumulation of statistics at the investigated point commenced 30 sec after transferring the instrument into a new position. There appeared thus a danger of retarded movement of the crystal, occurring during the time when the statistics were being accumulated, due to the elastic aftereffects in the mechanical parts which moved the crystal. Since the measured effect corresponded to a relative shift of the lines by $\approx 1\,\mu$ (dispersion of 0.0291 eV/μ), even very small retarded displacements could lead to a considerable distortion of the results. In order to exclude the influence of this effect on the final result of the measurements, the positions of the sources were changed in the sample holder after we measured half the number of series planned for each pair. Thus, if during the first half of the series the elastic aftereffect led to an increase in the effect, being added on to it, then in the second half of the series the elastic aftereffect was subtracted out from the effect.

The results of the individual series of measurements for $\Delta E(92-100)$, $\Delta E(94-100)$, and $\Delta E(92-94)$ are cited in Table II. The mean-square errors of the averages are calculated from the errors of the series. The external mean-square errors found from the deviations of the individual series from the average are indicated in parentheses.

The graphical results of the main experiment are shown in Fig. 4b. The point 92*, calculated from the sum of $\Delta E(94-100)$ and $\Delta E(92-94)$, as in the preliminary experiment, is close to the directly measured point 92. The position of the point 94 can now be reconciled within the limits of the errors with the theoretically expected position. (The dashed lines divide the effect of the addition of eight neutrons into equal intervals, each corresponding to the addition of two neutrons.) The data plotted in Fig. 4b are corrected for the admixture of other isotopes to the main isotope constituting the sample (see Table I). The correction was made under the assumption that the shift due to each added neutron is constant $\delta\Delta E/\delta A = \text{const}$. In any case the corrections are not large and considerably smaller than the statistical errors.

The corrected results of the main experiment are accepted as the final values of the isotopic shifts of the $K_{\alpha 1}$ line of $Mo^{92,94,100}$:

$$\Delta E(92-100) = 0.030 \pm 0.005\,(0.004)\ \text{eV};$$

$$\Delta E(94-100) = 0.027 \pm 0.006\,(0.008)\ \text{eV};$$

$$\Delta E(92-94) = 0.001 \pm 0.005\,(0.005)\ \text{eV}.$$

Table II. Isotopic shifts of the $K_{\alpha 1}$ line of molybdenum

No. of series	$\Delta E(92-100)$, 10^{-3} eV	$\Delta E(94-100)$, 10^{-3} eV	$\Delta E(92-94)$, 10^{-3} eV
1	33 ± 19	38 ± 14	5 ± 10
2	38 ± 14	44 ± 14	-8 ± 15
3	26 ± 10	55 ± 15	23 ± 19
4	44 ± 15	20 ± 23	8 ± 14
5	10 ± 11	-5 ± 16	-9 ± 18
6	7 ± 11	19 ± 19	7 ± 13
7	61 ± 23	-3 ± 18	-22 ± 17
8	33 ± 12	39 ± 15	1 ± 16
9	18 ± 17		
10	28 ± 21		
11	26 ± 23		
12	27 ± 22		
Average	29 ± 5 (4)	26 ± 6 (8)	1 ± 5 (5)

DISCUSSION OF THE RESULTS

The theoretical value of the effect for the $K_{\alpha 1}$ line of Mo^{92-100} has been calculated by Wertheim and Igo.[13] In the case closest to reality, that of a uniformly charged nucleus, assuming that the increment of the radius of the nuclear charge upon the addition of neutrons is described by the simplest relation:

$$R = 1.2 \cdot 10^{-13} A^{1/3} \text{ cm}, \qquad \frac{dR}{R} = \frac{1}{3}\frac{dA}{A}, \tag{5}$$

Wertheim and Igo obtained for the volume isotopic shift

$$\Delta E(92 - 100)_{\text{vol}} = +0.0536 \text{ eV}.$$

Babushkin refined the calculation and found with the same assumptions[21]:

$$\Delta E(92 - 100)_{\text{vol}} = +0.0343 \text{ eV}.$$

Before we compare our results with these values we must include in them the mass isotopic shift. Assuming that the specific mass shift for the x-ray transition $2p_{3/2} \rightarrow 1s_{1/2}$ is negligibly small, we find the mass shift to be

$$\Delta E(92 - 100)_{\text{m}} = -0.0082 \text{ eV}.$$

The resulting theoretical value of the total isotopic shift thus turns out to be

$$\Delta E(92 - 100) = +0.0261 \text{ eV}.$$

The experimental result is in very good agreement with the theory:

$$C \equiv \Delta E(92 - 100)_{\text{exp}} / \Delta E(92 - 100)_{\text{theoret}} = 1.15 \pm 0.19.$$

The error incurred in our apparatus in an investigation of line shifts with different energies can be estimated from the relation

$$\delta E / E = kE. \tag{6}$$

Substituting E = 17.5 keV and $\delta E = 5 \times 10^{-3}$ eV, we find on the basis of experiment for the $K_{\alpha 1}$ line of molybdenum

$$\delta E / E = 1.63 \cdot 10^{-8} E \text{ (keV)}.$$

The expected errors are compared in Table III with the theoretical volume isotopic shift calculated by Babushkin under the assumption of a uniformly charged incompressible nucleus.[22] The comparison shows that the method will apparently make it possible to carry out systematic measurements of the isotopic shifts for numerous elements (isotopes). The measurement of the isotopic shifts of x-ray lines can thus be regarded as a new method for the study of nuclear properties. With respect to the possibilities and the nature of the information that it offers, such a method will be very close to the optical isotopic shift. The three principal problems solved in the analysis of optical isotopic shifts are:

1. The study of the relative magnitudes of the

Table III. Comparison of the theoretical values of the volume isotopic shift for the $K_{\alpha 1}$ line of various elements with extrapolated experimental error

Element	$A_1 - A_2$	Effect [22], 10^{-3} eV	Error, 10^{-3} eV	Element	$A_1 - A_2$	Effect [22], 10^{-3} eV	Error, 10^{-3} эв eV
$_{37}$Rb	85—87	4.8	±3	$_{63}$Eu	151—153	64.7	±28
$_{42}$Mo	92—94	8.6	±5	$_{64}$Gd	154—156	70.7	±30
	94—96	8.6			155—157	70.4	
	96—98	8.6			156—158	70.3	
	98—100	8.6			158—160	69.9	
$_{44}$Ru	96—98	10.8	±6	$_{70}$Yb	171—173	119	±45
	98—100	10.7			172—174	119	
	100—102	10.6			174—176	118	
	102—104	10.5		$_{72}$Hf	178—180	140	±51
$_{46}$Pd	106—108	13.0	±7	$_{74}$W	182—184	152	±57
	108—110	12.9			184—186	151	
$_{47}$Ag	107—109	14.5	±8	$_{75}$Re	185—187	181	±61
$_{48}$Cd	106—108	16.2	±8	$_{76}$Os	186—188	199	±65
	108—110	16.1			188—190	198	
	110—112	16.0			190—192	197	
	112—114	15.9		$_{77}$Ir	191—193	215	±69
$_{50}$Sn	114—116	19.6	±10	$_{78}$Pt	194—196	233	±73
	116—118	19.5		$_{80}$Hg	196—198	279	±82
	118—120	19.3			198—200	277	
	120—122	19.1			200—202	276	
	122—124	19.0			202—204	275	
$_{56}$Ba	134—136	34.2	±17	$_{81}$Tl	203—205	300	±87
	136—138	34.0		$_{82}$Pb	204—206	328	±92
$_{60}$Nd	148—150	48.8	±23		206—208	326	
$_{62}$Sm	150—152	59.0	±26		208—210	325	
	152—154	58.7					

shifts with successive addition of neutron pairs to the nucleus yields information on the successive filling of the nuclear shells. This problem became particularly real after recent work[23] in which a theoretical expression was obtained for the effect with explicit account of the shell on which the added pair "settles."

2. The relative shifts make it possible to reach conclusions on the changes in the shape of the nucleus. In particular, the abrupt loss of spherical symmetry by the nucleus at $N = 88$ is beautifully exhibited in the anomalously large value of the corresponding isotopic shift (see, for example, [7]).

3. Finally, comparison of the absolute values of the experimental and theoretically calculated isotopic shifts for the complete aggregate of data enables one to obtain $E'' = R^2\partial^2E/\partial R^2$ which characterizes the compressibility of nuclear matter.[5,24]

The relative precision of the measurements of the shifts is at present higher for the optical shift than for that in the x-ray region. Therefore, although in individual cases purely experimental circumstances may render the x-ray shift the more convenient method, in solving the first two problems the optical shift is generally preferable.

The situation changes when one is dealing with a comparison of absolute values. Before one can obtain from the experimentally observed shift the value which characterizes the change in the charge distribution in the nucleus (the charge radius of the nucleus) due to the addition of neutrons, one must take into account correctly the contribution of the electron shell, i.e., one must calculate the total change of electron density in the region of the nucleus for the optical or x-ray transition under consideration. For the optical shift this problem is very complicated. The contribution of the valence electron to the total electron density near the nucleus of heavy nuclei constitutes a negligible ($\sim 10^{-4}$) part of the contribution of the inner shells.[25] For this reason even small deformations of the latter accompanying an optical transition can in principle lead to density changes on the nucleus comparable to those due to the valence electron itself. It is important that the existence of noticeable deformations of the inner shells when the states of the valence electrons change follows directly from the chemical shift of the tin and molybdenum $K_{\alpha 1}$ lines which we have observed previously.[17,19] One is led to suspect that a certain discrepancy between the experimental and theoretical values of the optical isotopic shifts, explained by the compressibility and leading to the anomalously small value $E'' \approx 50$ MeV,[5-24] is due to an incorrect allowance for the deformation of the inner shells.1)

In the x-ray shift the K electron contributes the main part to the total density on the nucleus, the contributions of the remaining shells being in themselves relatively small. Account of small changes of these small contributions, caused by the deformation of the outer shells in the x-ray K transition, can apparently only yield very small corrections. Concrete calculations by Wertheim and Igo[13] and also by Babushkin[21], carried out under the assumption that the K electron participating in the transition is the only electron, yielded 0.0357 eV for the isotopic shift in Mo^{92-100}; identical calculations carried out with allowance for the presence of the remaining electron shells and their deformation yielded a value of 0.0343 eV, differing from the preceding value only by 4 per cent.2) The above circumstance renders the measurement of the x-ray isotopic shift a method which will apparently permit one to obtain the most reliable information on the absolute values of the changes of the charge radii of isotopes. The most direct application of this effect may turn out to be the investigation of the compressibility of nuclear matter.

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1)Theoretical calculations of the compressibility (of the type of calculations of Brueckner et al.[26] etc) lead to appreciably larger values $E'' \sim 170$–200 MeV.

2)The cited results from the work of Babushkin[21] were obtained without using the approximation $E \ll m_0c^2$ and are therefore more accurate.

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