

ISOTOPE SHIFT OF SPECTRAL LINES

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A formula for the isotope shift constant is derived by applying the Sommerfeld method of determining the wave functions with account of the finite size of the nucleus. The calculations are carried out for two nuclear charge distributions, a uniform and a trapezoidal one. By comparing experimental and theoretical values, two empirical parameters are introduced which characterize the nuclear compressibility: the regular compressibility coefficient $\eta = 0.7$ and the deformation compressibility coefficient $\xi = -5/8\pi$. These values lead to satisfactory agreement for most isotopes with known deformation parameters.

THE isotope shift of spectral lines occurs as a consequence of the difference in the level shifts of an electron in two isotopes of one and the same element due to the finite dimensions of the nucleus. The shift of the energy levels of the electron is very simply expressed in terms of the values of the wave function of the electron at the nuclear boundary:^[1]

$$\Delta E = \hbar c R^2 \{g_0(R) f_e(R) - g_e(R) f_0(R)\}, \quad (1)$$

where g_0 and f_0 are the relativistic Coulomb functions and g_e and f_e are the wave functions outside the nucleus with account of the finite nuclear dimensions.

It is seen from (1) that the calculation of the isotope shift reduces basically to the determination of the correct wave functions g_e and f_e . The numerous papers on the calculation of the isotope shift can therefore be divided into two groups according to the wave functions used.

The first group includes those papers which use the Breit-Rosenthal wave functions.^[2] These wave functions were found as solutions of the relativistic Dirac equations with $E = m_0c^2$. This assumption is indeed valid for the optical spectra. But if the calculations are extended to the isotope shift of x-radiation, it is hardly justified to use the Breit-Rosenthal wave functions, especially not for heavy elements.

The second group includes the papers which use an integral representation for the wave functions.^[3] In the last paper of Fradkin,^[4] where this method is used, a rather simple expression for the isotope shift is derived.

However, there exists a third method of determining the wave functions g_e and f_e which is free from the assumption of Breit and Rosenthal. The formulas obtained below can be used also for the calculation of the isotope shift of x-radiation, i.e.,

they are more general. The idea of this method is due to Sommerfeld.^[5] With the help of this method the author has computed^[6] the wave functions g_e and f_e :

$$\left\{ \frac{f_e}{g_e} \right\} = \pm C e^{-\rho/2} \rho^{\gamma-1} \sqrt{1 \mp \varepsilon} \left\{ \frac{\Delta n}{\rho^{2\gamma}} A_{\pm} \pm \frac{\Delta n}{\rho^{2\gamma-1}} B_{\pm} - D_{\pm} \right\}, \quad (2)$$

where

$$A_{\pm} = \frac{\Gamma(2\gamma)(2|\kappa|)!(n-|\kappa|)!}{(n+|\kappa|)!} [(N-\kappa) \pm (n'+2\gamma)],$$

$$B_{\pm} = \frac{\Gamma(2\gamma)(2|\kappa|)!(n-|\kappa|)!(n'+2\gamma)}{(n+|\kappa|)!(1-2\gamma)} \times [\pm(1-2\gamma-n') - (N-\kappa)],$$

$$C = \frac{\sqrt{\Gamma(2\gamma+n'+1)}}{\Gamma(2\gamma+1)\sqrt{4N(N-\kappa)n'}} \left(\frac{2Z}{Na_0}\right)^{1/2}, \quad a_0 = \frac{\hbar^2}{me^2},$$

$$D_{\pm} = (N-\kappa) \pm n', \quad n' = n - |\kappa|, \quad N = \sqrt{n^2 - 2n'(|\kappa| - \gamma)},$$

$$\kappa = \begin{cases} -(j+1/2) = -(l+1), & j = l+1/2, \\ +(j+1/2) = +l, & j = l-1/2, \end{cases}$$

$$\gamma = \sqrt{\kappa^2 - \alpha^2 Z^2}, \quad \rho = \frac{2Z}{Na_0} R, \quad \varepsilon = \frac{E}{m_0c^2},$$

$$\Delta n = \rho^{2\gamma} \frac{LD_{\pm} \mp \varepsilon_0 MD_{\pm}}{LA_{\pm} \mp \varepsilon_0 MA_{\pm}} \quad \text{for } j = l \pm 1/2, \quad \varepsilon_0 = \sqrt{\frac{1-\varepsilon}{1+\varepsilon}}. \quad (3)$$

Here for $ns_{1/2}$

$$L = \alpha Z (2/5 - 14/315 \alpha^2 Z^2), \quad M = 1 - 17/60 \alpha^2 Z^2.$$

The small difference between our L and M and the values given in ^[6] is due to the fact that we have included higher terms of the expansion in the present paper.

The wave functions (2) go over into the Sommerfeld wave functions if $\gamma \rightarrow 1$. Therefore the Sommerfeld wave functions are only correct for light elements.

Using the wave functions (2) in (1), we easily obtain an expression for the isotope shift constant for a uniform nuclear charge distribution:

$$C_{\text{unif}} = 4\gamma R_{\infty} \left(\frac{y}{N}\right)^{2\gamma} \frac{LD_- - \epsilon_0 MD_+}{LA_- - \epsilon_0 MA_+} \frac{2\Gamma(2\gamma + n)}{\Gamma(2\gamma + 1)(n + 1)!} \frac{\delta R}{R}; \quad (4)$$

here R_{∞} is the Rydberg constant, and $y = 2ZR/a_0$.

If we assume that the nuclear radius has the following dependence on A : $R = 1.2 \times 10^{-13} A^{1/3}$, we have

$$\delta R/R = \delta A/3A, \quad (5)$$

and the calculation of the isotope shift constant using (4) gives a result which is almost the same as that obtained by using the Breit-Rosenthal functions for light elements, but somewhat smaller ($\sim 6\%$) for heavy elements.

The results obtained are given in Table I. There is no necessity of including in this table the calculations based on the Breit-Rosenthal functions, which are given in the review article of Brix and Kopfermann.^[7] It suffices to quote from this review article the results for the lightest and the heaviest isotopes:

C_{unif} for $_{37}\text{Rb}$ (85-87):	30.1Ref.[7],	29.75 Eq.(4)
C_{unif} for $_{82}\text{Pb}$ (204-203):	524Ref.[7],	429 Eq.(4)

This result was to be expected, since the Breit-Rosenthal assumption is valid only for the light elements.

However, in these as in other calculations, the isotope shift constant comes out about twice as large as the experimental value. This disagreement is diminished somewhat if the results of Elton^[11] are used, who showed on the basis of an analysis of electron scattering on nuclei that the dependence of the nuclear radius on A is more complicated than (5):

$$R = (1.115 \cdot A^{1/3} + 2.151 \cdot A^{-1/3} - 1.742 \cdot A^{-1}) \cdot 10^{-13}. \quad (6)$$

Using this formula, we obtain

$$\frac{\delta R}{R} = \frac{\delta A}{3A} \frac{1 - 1.929A^{-2/3} + 4.687A^{-4/3}}{1 + 1.929A^{-2/3} - 1.562A^{-4/3}} \equiv \frac{\delta A}{3A} \zeta. \quad (7)$$

The factor ζ takes values between 0.90 and 0.833, the first value referring to $_{82}\text{Pb}$, the second to $_{37}\text{Rb}$. In this way the calculated values of the isotope shift constant are somewhat decreased, but not enough to reach complete agreement between the experimental and theoretical data (see Table I).

This problem can be viewed in two different ways. On one hand, it was proposed by Meligy^[8,12] that good agreement between the experimental and theoretical data can be achieved if the non-uniformity of the nuclear charge distribution is taken into account.

Meligy restricted himself in his calculations to the simplest type of charge distribution, viz., the trapezoidal one:

$$\rho = \begin{cases} \rho_0, & 0 \leq r \leq d-t \\ \rho_0(d+t-r)/2t, & d-t \leq r \leq d+t \\ 0, & d+t \leq r \end{cases} \quad (8)$$

here d is the distance at which the charge density has dropped to half its central value, and $2t$ is the surface thickness of the nucleus. Meligy obtained the value

$$C_{\text{trap}}/C_{\text{unif}} \approx 0.5.$$

If this were indeed so, we would obtain excellent agreement between the experimental and theoretical values of the isotope shift constant, even if (5) is assumed valid for R . However, it was pointed out recently by Bodmer^[13] that the paper of Meligy contains some mistakes and that this ratio should be of the order ~ 0.87 . In view of this controversy about the effect of the nuclear charge distribution on the value of the isotope shift constant, the author has carried out a new calculation with a trapezoidal distribution.

The method of calculation was the same as the one used by Meligy. The difference consists in the use of different wave functions. The result obtained is

$$C_{\text{trap}} = 4\gamma R_{\infty} \left(\frac{y}{N}\right)^{2\gamma} \frac{2\Gamma(2\gamma + n)}{\Gamma(2\gamma + 1)(n + 1)!} \frac{L_{\text{trap}}D_- - \epsilon_0 MD_+}{L_{\text{trap}}A_- - \epsilon_0 MA_+} \frac{\delta R_{\text{trap}}}{R_{\text{trap}}} \\ \times \left\{ 1 + \frac{\alpha Z}{10\gamma} \frac{t}{R_{\text{trap}}} \left(1 - \frac{t}{R_{\text{trap}}}\right)^{-2} \right. \\ \left. \times \frac{\epsilon_0 M (A_+ D_- - A_- D_+)}{(L_{\text{trap}} A_- - \epsilon_0 M A_+) (L_{\text{trap}} D_- - \epsilon_0 M D_+)} \right\}, \quad (9)$$

where

$$R_{\text{trap}} = d + t, \quad L_{\text{trap}} = \alpha Z \left(\frac{2}{5} + t/5d - \frac{14}{315} \alpha^2 Z^2\right).$$

The parameters d and t were taken to be^[14]

$$d = 1.07 \cdot 10^{-13} A^{1/3}, \quad t = 1.49 \cdot 10^{-13} = \text{const.} \quad (10)$$

The values of C_{trap} calculated by this formula are given in Table I. It is seen from these data that the ratio $C_{\text{trap}}/C_{\text{unif}}$ varies within the limits 0.86 for Pb and 0.83 for Rb if $R = 1.2 \times 10^{-13} A^{1/3}$, and is almost equal to unity if R is assumed to be given by (6). These results confirm the conclusions of Bodmer, although we have used different wave functions. Thus the nonuniformity of the nuclear charge distribution leads to a very insignificant decrease of the value of the isotope shift constant (~ 1 to 3%).

According to the second point of view, the entire difference between the experimental and theoretical values is ascribed to the nuclear compressibility

Table I. Isotope shifts

Element	$A_1 - A_2$	$C_{\text{theor}}, 10^{-3} \text{ cm}^{-1}$						$C_{\text{exp}}, 10^{-3} \text{ cm}^{-1}$
		Cunif. R from(Eq.5)	Cunif. R from(Eq.6)	Ctrap	Cunif $\eta=0.7$	Cdef $\xi=0$	Cdef $\xi=-\beta/\delta\pi$	
³⁷ Rb	85—87	29.7	25.6	24.8	17.9	14.0	16.0	8±12 [7]
³⁸ Sr	84—86	32.2	27.7	26.8	19.4			~0 [7]
	86—88	31.9	27.5	26.6	19.2			~0 [7]
⁴⁴ Ru	96—98	48.0	41.4	40.1	29.0	52.3	39.7	} 34±9 [7] (average)
	98—100	47.6	41.0	39.7	28.7	37.9	32.9	
	100—102	47.2	40.7	39.4	28.5	43.7	35.5	
	102—104	46.9	40.4	39.1	28.3	46.1	36.5	
⁴⁶ Pd	106—108	53.2	45.8	44.3	32.1	41.1	36.2	42±9 [7]
	108—110	52.7	45.5	43.9	31.8	42.6	36.7	36±5 [7]
⁴⁷ Ag	107—109	57.3	49.3	47.7	34.5	41.3	37.6	38±6 [7]
⁴⁸ Cd	106—108	61.5	53.0	51.4	37.1	43.6	40.0	} 32±4 [7]
	108—110	61.1	52.6	51.0	36.8	32.5	34.9	
	110—112	60.6	52.3	50.6	36.6	40.1	38.1	
	112—114	60.2	51.9	50.2	36.3	40.0	38.0	
⁵⁰ Sn	114—116	59.9	51.6	49.9	36.1	37.0	36.5	} 40±10 [7]
	112—114	69.1	59.7	57.7	41.8	44.0	42.8	
	114—116	68.7	59.3	57.3	41.5	39.9	40.8	
	116—118	68.2	58.9	56.9	41.2	40.9	41.1	
	118—120	67.7	58.5	56.5	40.9	40.8	40.9	
⁵⁴ Xe	120—122	67.3	58.2	56.2	40.7	40.5	40.7	15±10 [7]
	122—124	66.9	57.8	55.8	40.4	40.4	40.4	15±10 [7]
	132—134	85.6	73.9	71.5	51.7			18±4 [8]
	134—136	85.1	73.5	71.1	51.5			25±6 [7]
⁵⁶ Ba	134—136	97.5	84.4	81.6	59.1			44±9 [32]
	136—138	96.9	83.8	81.1	58.6			67±13 [32]
⁵⁸ Ce	136—138	112	96.6	93.8	67.6			6±1 [8]
	138—140	111	96.0	93.2	67.0			6±1 [8]
⁶⁰ Nd	140—142	110	95.5	92.7	66.8	85.6	74.9	147±30 [7]
	142—144	125.8	108.8	105.7	76.2			187±35 [7]
	144—146	125.1	108.1	105.1	75.7	78.3	76.8	
	146—148	124.4	107.6	104.4	75.3	144.5	104.4	202±38 [8]
⁶² Sm	148—150	123.7	107.0	103.8	74.9	302.5	170.6	286±54 [8]
	144—146	143.1	123.7	120.4	86.6			190±40 [7]
	146—148	142.3	123.1	119.7	86.2			190±40 [7]
	148—150	141.5	122.4	119.0	85.7	155.2	114.5	217±46 [8]
⁶⁴ Eu	150—152	140.7	121.7	118.3	85.2	423.0	225.0	317±67 [8]
	152—154	140.0	121.1	117.6	84.8	222.0	141.6	170±35 [4]
	151—153	149.4	129.2	125.7	90.4	664.6	326.2	450±50 [7]
	152—154	160.6	139.0	135.5	97.3			375±60 [8]
⁶⁴ Gd	154—156	159.9	138.3	134.7	96.8	259.4	163.0	162±26 [9]
	155—157	159.3	137.9	134.3	96.5	107.6	101.4	109±18 [9]
	156—158	159.0	137.6	133.8	96.3	182.5	131.4	121±19 [9]
	158—160	158.2	136.9	133.2	95.8	146.9	116.7	125±20 [9]
⁷⁰ Yb	170—172	231.0	200.2	196.0	140.1	174.1	153	110±10 [7]
	171—173	230.5	199.7	195.4	139.8	81.2	117.3	112±12 [4]
	172—174	229.9	199.2	194.9	139.4	130.4	135.5	99±8 [7]
	174—176	228.8	198.2	193.9	138.7	102.9	124.7	94±8 [7]
⁷² Hf	178—180	260.5	225.8	224.8	158	97.3	135.2	114±13 [7]
	180—182	297.3	257.9	253.1	180.5			80±20 [8]
⁷⁴ W	182—184	295.9	256.7	252.2	179.7	104.7	152.3	132±23 [10]
	184—186	294.3	255.8	250.8	179.0	164.2	173.6	117±20 [10]
	185—187	314.8	274.4	268.7	192.1	30.4	133.7	157±40 [7]
	186—188	336.4	292.1	287.5	204.5	77.9	159.4	176±27 [8]
⁷⁶ Os	188—190	334.9	290.9	286.0	203.6	134	178.8	150±23 [8]
	190—192	333.2	289.2	284.5	202.5	-71.5	105	130±20 [8]
⁷⁷ Ir	191—193	356.7	310.2	305.2	217.1	44.7	156.7	130±30 [7]
⁷⁸ Pt	194—196	379.7	330.1	323.9	231.1	27.7	160.9	135±25 [7]
	196—198	378.3	328.6	322.6	230.0	337.3	267	
⁸⁰ Hg	196—198	434.3	377.8	373.3	264.5			
	198—200	432.4	375.9	371.5	263.1	191.8	239.3	243±27 [8]
	200—202	430.6	374.4	369.7	262.1			270±30 [8]
	202—204	428.3	372.8	367.6	261.0			267±30 [8]
⁸¹ Tl	203—205	459.2	399.6	395.0	279.7			280±40 [7]
⁸² Pb	204—206	491.7	427.9	423.7	299.5			280±31 [8]
	206—208	489.6	425.8	421.5	298.0			315±35 [8]
	208—210	487.3	424.1	419.5	296.9			548±61 [8]

effect. If we adopt this viewpoint and introduce the regular compressibility coefficient^[15] $\eta = 0.7$, we obtain indeed, as is seen from Table I, satisfactory agreement between the experimental and theoretical values for spherical nuclei.

It stands to reason that further deviations due to the deformability of the nucleus will be observed. To obtain a formula for the isotope shift constant with account of the nuclear deformability it is necessary to multiply (4) by the factor

Table II. Deformation parameters

Nucleus	Q_0	β	Nucleus	Q_0	β	Nucleus	Q_0	β
Rb ⁸⁵	0.924 [16]	0.110	Sn ¹²²	1.23 [19]	0.0888	Yb ¹⁷³	7.77 [28]	0.305
Rb ⁸⁷	0.70 [16]	0.083	Sn ¹²⁴	1.19 [19]	0.0852	Yb ¹⁷⁴	7.72 [26]	0.302
Ru ⁹⁶	1.60 [17]	0.147	Ce ¹⁴⁰	1.65 [20]	0.0947	Yb ¹⁷⁶	7.60 [26]	0.296
Ru ⁹⁸	2.18 [17]	0.196	Ce ¹⁴²	2.03 [20]	0.115	Hf ¹⁷⁸	6.85 [29]	0.260
Ru ¹⁰⁰	2.40 [17]	0.212	Nd ¹⁴⁴	1.52 [21]	0.0835	Hf ¹⁸⁰	6.61 [29]	0.250
Ru ¹⁰²	2.71 [17]	0.235	Nd ¹⁴⁶	1.58 [21]	0.0863	W ¹⁸²	6.34 [29]	0.233
Ru ¹⁰⁴	3.05 [17]	0.259	Nd ¹⁴⁸	2.67 [18]	0.141	W ¹⁸⁴	6.04 [29]	0.222
Pd ¹⁰⁶	2.55 [17]	0.208	Nd ¹⁵⁰	4.99 [18]	0.253	W ¹⁸⁶	5.99 [29]	0.219
Pd ¹⁰⁸	2.73 [17]	0.220	Sm ¹⁴⁸	2.24 [18]	0.116	Re ¹⁸⁵	6.28 [17]	0.226
Pd ¹¹⁰	2.94 [17]	0.234	Sm ¹⁵⁰	3.14 [18]	0.158	Re ¹⁸⁷	5.60 [17]	0.202
Ag ¹⁰⁷	2.65 [18]	0.211	Sm ¹⁵²	5.96 [22]	0.286	Os ¹⁸⁶	6.57 [30]	0.233
Ag ¹⁰⁹	2.79 [18]	0.219	Sm ¹⁵⁴	6.85 [18]	0.323	Os ¹⁸⁸	6.10 [30]	0.216
Cd ¹⁰⁶	2.17 [17]	0.173	Eu ¹⁵¹	2.33 [23]	0.117	Os ¹⁹⁰	5.83 [30]	0.205
Cd ¹⁰⁸	2.32 [17]	0.182	Eu ¹⁵³	6.94 [23]	0.323	Os ¹⁹²	4.54 [17]	0.161
Cd ¹¹⁰	2.25 [17]	0.175	Gd ¹⁵⁴	5.82 [24]	0.271	Ir ¹⁹¹	4.39 [8]	0.155
Cd ¹¹²	2.33 [17]	0.179	Gd ¹⁵⁵	6.48 [24]	0.297	Ir ¹⁹³	3.40 [8]	0.121
Cd ¹¹⁴	2.42 [17]	0.184	Gd ¹⁵⁶	6.83 [25]	0.311	Pt ¹⁹⁴	4.19 [31]	0.145
Cd ¹¹⁶	2.46 [17]	0.186	Gd ¹⁵⁷	6.57 [24]	0.299	Pt ¹⁹⁶	2.97 [31]	0.104
Sn ¹¹²	1.34 [19]	0.102	Gd ¹⁵⁸	7.34 [24]	0.329	Pt ¹⁹⁸	3.68 [31]	0.127
Sn ¹¹⁴	1.42 [19]	0.106	Gd ¹⁶⁰	7.65 [26]	0.339	Hg ¹⁹⁸	3.36 [8]	0.113
Sn ¹¹⁶	1.38 [19]	0.102	Yb ¹⁷⁰	7.54 [27]	0.299	Hg ²⁰⁰	2.92 [8]	0.0984
Sn ¹¹⁸	1.38 [19]	0.101	Yb ¹⁷¹	7.98 [28]	0.314			
Sn ¹²⁰	1.31 [19]	0.0952	Yb ¹⁷²	7.72 [26]	0.304			

$$f_{\text{def}} = \frac{\delta \langle R_{\text{def}}^{2\gamma} \rangle_{\text{av}}}{\delta \langle R_{\text{def}}^{2\gamma} \rangle_{\text{av}}} = 1 + \frac{2\gamma(2\gamma+3)}{8\pi} \left(\frac{\beta_1 + \beta_2}{2} \right)^2 + \frac{1}{2\gamma} \frac{R}{\delta R} \frac{2\gamma(2\gamma+3)}{8\pi} (\beta_2^2 - \beta_1^2), \quad (11)$$

Then

$$C_{\text{def}} = C_{\text{unif}} f_{\text{def}}. \quad (12)$$

The deformation parameters for axially symmetric prolate deformed nuclei were determined by the well-known formula^[4]

$$Q_0 = (3/\sqrt{5\pi}) ZR^{2\beta} (1 + 0.36\beta), \quad (13)$$

where R was assumed to be given by (6). The values of Q_0 and β are given in Table II.

It is seen from a comparison of the experimental and theoretical values for deformed nuclei that agreement can be obtained only if, in analogy to the case of spherical nuclei, the deformation compressibility coefficient^[4]

$$\xi = -5/8\pi \quad (14)$$

is introduced and f_{def} is set equal to

$$f_{\text{def}} = 1 + \frac{2\gamma(2\gamma+3) - 5}{8\pi} \left(\frac{\beta_1 + \beta_2}{2} \right)^2 + \frac{1}{2\gamma} \frac{R}{\delta R} \frac{2\gamma(2\gamma+3) - 5}{8\pi} (\beta_2^2 - \beta_1^2). \quad (15)$$

The results of the calculation using (15) are also given in Table I.

In conclusion, we note that a comparison between the experimental and theoretical values of Table I shows good agreement for spherical and weakly deformed nuclei. Complete agreement for strongly deformed nuclei (Sm, Eu) could not be obtained. It

is quite probable that this is explained by the approximate character of the calculations taking into account only terms of order β^2 , whereas higher powers of the deformation parameter must be included for large deformations. Moreover, it is possible that the deviations for some pairs of isotopes (Ce, Nd, W) arise from incorrect values of the deformation parameter β .

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