### STUDY OF THE CHARGE RADII OF THE STABLE LEAD ISOTOPES

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### Abstract

Isotope shifts have been measured of the  $K\alpha_1$  X-ray lines emitted after photo ionization of  $^{204}\text{Pb}$ ,  $^{206}\text{Pb}$ ,  $^{207}\text{Pb}$  and  $^{208}\text{Pb}$  samples. The results are compared with theoretical values for  $\delta < r^2 >$  calculated with a microscopic model. The X-ray shift data are also compared with optical data and the nuclear parameters  $\lambda$  derived from electron scattering results.

### 1. Introduction

In the recent years progress in nuclear theory has led to more detailed predictions of the properties of the states of atomic nuclei, especially in the vicinity of double magic nuclei like  $^{208}\text{Pb}$ . Among other EO properties the mean square charge radius  $<\!r^2>$  has been the subject of a theoretical study¹). The determination of K X-ray isotope shifts provides data on the change  $\delta<\!r^2>_{\text{A}\!\rightarrow\text{A}\!+\!\delta\text{A}}$ , where  $\delta\text{A}$  is the number of neutrons added to the isotope with mass number A.

Such experiments have been performed by Boehm and his coworkers<sup>2</sup>) and by Sumbaev and his group<sup>3</sup>) in various interesting regions of the heavy nuclei. The first data on the <sup>208</sup>Pb-<sup>206</sup>Pb shift were obtained by Chesler and Boehm<sup>4</sup>). Later, Lee and Boehm<sup>5</sup>) studied the shifts of all stable lead isotopes.

Prior to the first X-ray isotope shift studies, optical Pb-isotope shift measurements were carried out by Manning and his coworkers $^6$ ), Brix and coworkers $^7$ ) and Steudel $^8$ ). Through a comparison of the optical data with the K X-ray shift data the optical shifts can be normalized and then used for the determination of <r $^2>$  values of nuclei, for which X-ray shifts are impractical, but optical shifts have been obtained as e.g.  $^{210}$ Pb  $^7$ ) and  $^{202}$ ,  $^{203}$ Pb  $^9$ ).

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Information about the nuclear charge radius can also be obtained from (e,e)-data and from muonic data. The <r $^2>$  values from muonic data alone are model dependent apart from the uncertainty that arises from the large nuclear polarization shifts of the muonic transitions.

As the effects of nuclear polarization on the electronic K X-ray lines are negligible and since the measured energy shifts do only weakly depend on the higher moments of the nuclear charge distribution, we felt it worth the effort to carry out an independent measurement of the electronic K X-ray shifts of the stable lead isotopes with the goal to minimize the total errors and thus to provide a reasonably precise set of data that allows to make better use of the existing optical data and those that are presently taken<sup>10</sup>).

## 2. Theoretical prediction

The mean square charge radius of a nuclear state  $|\Psi\rangle$  is

$$\langle r^2 \rangle = \frac{1}{Z} \langle \Psi | \Sigma r_i^2 | \Psi \rangle = \frac{1}{Z} \int \rho(r) r^2 d^3 r$$
 (1)

The sum includes all protons;  $\rho(r)$  is the charge density.

For the computation of the isotope shift  $\delta < r^2 >_{207 \rightarrow 208}$  first RPA calculations were carried out of the particle-hole states in  $^{208}$ Pb with a properly chosen empirical interaction  $^1$ ), and then for the neighbouring nuclei particles or holes were coupled to the  $^{208}$ Pb states. The correct density dependence of the Migdal force

$$F^{ph}(r,r') = C\delta(r-r')\{f(r)+f'(r)\tau \cdot \tau' + \sigma \cdot \sigma'(g+g'\tau \cdot \tau')\}$$
(2)

was found to be crucial<sup>1)</sup> for reproducing the monopole properties in the lead region. The expected isotope shift is then

$$\delta < r^2 >_{207 \rightarrow 208} = 0.061 \text{ fm}^2 = < r^2 >_{208} - < r^2 >_{207} .$$

For  $\delta < r^2 >_{206 \rightarrow 208}$  the ground state wave function of the two-hole system had to be calculated. Using a phenomenological interaction, Klemt obtained the following wave function<sup>1</sup>) for the <sup>206</sup>Pb ground state:

$$\Psi = -0.116 \text{ Oh}_{\frac{1}{2}}^{-2} -0.171 \text{ 1f}_{\frac{7}{2}}^{-2} -0.426 \text{ 1f}_{\frac{5}{2}}^{-2}$$
$$-0.322 \text{ 2p}_{\frac{3}{2}}^{-2} -0.816 \text{ 2p}_{\frac{1}{2}}^{-2} +0.262 \text{ 0i}_{\frac{1}{2}}^{-2}_{\frac{9}{2}}$$
and  $\delta < r^2 >_{2.06 \to 2.08} = 0.1149 \text{ fm}^2$ .

In previously reported shell model calculations<sup>11)</sup> of the isotopes <sup>206,204</sup>Pb the same six neutron hole states in 208Pb were included in the model space. In this model space the effective Hamilton operator is specified by six single-hole energies, taken from the observed spectrum of 207Pb, and by a two-body residual interaction. The two-body matrix elements are based on those constructed<sup>12</sup>) by Kuo and Herling for this model space from the Hamada-Johnston nucleon-nucleon potential. That exact Kuo-Herling interaction was modified to the extent that the core polarization (or hole particle bubble) contribution was multiplied by a factor of 0.75 to improve agreement between the calculated and observed spectrum of 208Pb 11). All Pauli-allowed states of all configurations of holes in this model space were included in the calculation. The resultant spectra for 204,206,207Pb are in good agreement with observed spectra. The shell model wave functions were then used to evaluate  $\langle r^2 \rangle$ . The input to the calculation of <r2> are the calculated effective matrix elements  $\langle j||r^2||j\rangle$ . These were taken directly from Ref. 1.

The predicted isotope shifts are then  $\delta < r^2 >_{2\,0\,6} \rightarrow_{2\,0\,8} = 0.1163 \text{ fm}^2 \text{ in very close correspondence with the value obtained with the use of Klemt's wave function, and } \delta < r^2 >_{2\,0\,4} \rightarrow_{2\,0\,8} = 0.227 \text{ fm}^2.$ 

# 3. The isotope shift of K X-rays

The isotope shift is directly measurable as energy shift of electronic K X-rays if other shifts are absent or corrected for. Chemical shifts  $^{13}$  can be avoided if the isotopically enriched samples exist in identical chemical forms. Shake-off  $^{14}$  and atomic structure shifts  $^{15}$  are eliminated if all samples are excited in the same manner through photo-ionization.

Under such experimental conditions the measured isotope shift

$$\delta E_{\text{exp }A \rightarrow A + \delta A} = E(KX)_A - E(KX)_{A + \delta A}$$
 (3)

corresponds with the shift of the energy of the  $\ln_{1/2}$  level when going from A to A+ $\delta$ A. It is given by the

Coulomb shift  $\delta E_{\text{COUl}}$ ,  $A \rightarrow A + \delta A$ , which is the volume effect, the dominant term for heavy nuclei, and by the mass shift  $\delta E_{\text{mass},A \rightarrow A + \delta A}$ . The nuclear polarization has been estimated to be a very small correction for electronic X-rays, and it is negligible in comparison with the other effects. Therefore the experimental shift is

$$\delta E_{\text{exp}} = \delta E_{\text{coul}} + \delta E_{\text{mass}}$$
 (4)

The mass shift of heavy nuclei is to good approximation $^4$ )

$$\delta E_{\text{mass,A}\rightarrow A+\delta A} \sim -\frac{2}{3} \frac{\delta A}{1836A^2} \cdot E_{KX,A}$$
 (5)

Inserting for the energy of the K X-ray line of the isotope A the value e.g. of the  $K\alpha_1$  line of Pb:  $E\alpha_1$  = 75.0 keV, one readily obtains

$$\delta E_{\text{mass.A}\rightarrow\text{A+1}} \sim -0.7 \text{ meV}$$
,

which implies that the total mass shift is less than 1% of the measured<sup>5</sup>) shifts and less than 7% of the experimental uncertainties of the previous data<sup>5</sup>).

In the treatment of our measured data we neglect the nuclear polarization, but we correct for the mass shift in order to obtain

$$\delta E_{\text{coul}} = \delta E_{\text{exp}} - \delta E_{\text{mass}}$$
 (6)

The Coulomb shift reflects the change of the nuclear size and shape. It has been studied in detail by Seltzer<sup>16</sup>) who obtained the electron wave functions from self consistent field calculations starting with the Dirac equation and using the Slater free-electron exchange approximation. The calculation of the nuclear Coulomb potential was based on a Fermi charge distribution.

For the Coulomb shift Seltzer obtained a rapidly converging expansion

$$\delta E_{coul} = \sum_{n=1}^{\Sigma} C_n \delta \langle r^{2n} \rangle$$
 (7)

with

$$\delta < r^{2n} > = \int \delta \rho(r) r^{2n} d^3 r / (\rho(r) d^3 r . \tag{8}$$

With the numerical values  $^{16}$ ) of the expansion coefficients for  $_{82}{\rm Pb}$ , the  ${\rm ls}_{1/2}$  level is shifted by

$$\frac{\delta E_{\text{coul}}}{\text{meV}} = 1880 \frac{\delta < r^2 >}{\text{fm}^2} - 2.11 \frac{\delta < r^4 >}{\text{fm}^4} + 5.59 \cdot 10^{-3} \frac{\delta < r^6 >}{\text{fm}^6} + \dots$$
(9)

Another way of writing eq. (9) is

$$\delta E_{\text{coul}} = \lambda \cdot 1880 \, \frac{\text{meV}}{\text{fm}^2} \tag{10}$$

where  $\lambda$  is called: nuclear parameter

$$\lambda = \delta < r^2 > + \frac{C_2}{C_1} \delta < r^4 > + \frac{C_3}{C_1} \delta < r^6 > + \dots$$
 (11)

This parameter is what both the atomic optical and the X-ray isotope shifts measure.

As the shift of the  $2p_{\frac{3}{2}}$  level is negligibly small<sup>5</sup>) one has

$$\delta E_{\text{exp}} = \delta E_{\text{K}\alpha},$$
 (12)

since the  $K\alpha_1$  line is the  $2p_{3/2} \rightarrow 1s_{1/2}$  transition. Together with eq. (6) we now have

$$\delta E_{\text{coul}} = \delta E_{\text{K}\alpha_1} - \delta E_{\text{mass}}$$
 (13)

and with eq. (10)

$$\lambda = (\delta E_{K\alpha_1} - \delta E_{mass}) fm^2 / 1880 \text{ meV} . \tag{14}$$

### 4. Experimental method

The isotope shifts are extremely small and they can only be measured with reasonable accuracy under conditions of high resolution. This requirement implies the use of diffraction spectrometers. Curved crystal spectrometers are chosen because of their better luminosity. The CalTech group<sup>2</sup>, <sup>4</sup>, <sup>5</sup>) has used with one exception<sup>17</sup>) - the Cauchois geometry<sup>18</sup>). This geometry has been exclusively applied by the Leningrad group<sup>3</sup>, <sup>19</sup>). A DuMond spectrometer<sup>20</sup>) was employed only by Van Eijk and his group<sup>21</sup>, <sup>22</sup>).

The intensity at a Cauchois spectrometer is as high as that at a DuMond spectrometer. The Cauchois spectrometer utilizes a narrow detector which minimizes the background, and the DuMond spectrometer allows the use of very small sources which is of advantage in studies on enriched isotopes.

The expected isotope shifts are of the order of  $10^{-6}$  of the X-ray energies and only  $\sim 10^{-3}$  of the natural line width  $\Gamma_{\text{nat}}$ . For the shift measurement only the difference is determined of the energies of corresponding K X-rays emitted from different sources containing different isotopes. Such a measurement strategy avoids not only the necessity of absolute measurements on a level of accuracy of  $10^{-7} > \Delta E/E$ , it also eliminates instrumental errors to a large extent. The accuracy being limited only by the counting statistics requires the experiment to be carried out under optimized conditions<sup>23</sup>). This implies the minimization of  $\Gamma/\sqrt{N}$  where  $\Gamma$  is the total width of the X-ray reflection and N is the total number of counts under that peak.

As the CalTech group utilized  $Pb(NO_3)_2$  and PbS samples",  $^5$ ) we decided to carry out our measurement on metallic Pb samples at the source positions of

the CERN DuMond spectrometer<sup>24</sup>) which had served already for extensive X-ray shift studies<sup>25</sup>). The Pb K X-rays were induced through photo-ionization by means of a 50 Ci 169Yb source at a distance of  $\sim$  0.5 mm from the two metallic Pb samples, each 4 mm high, 2 mm deep and 0.12 mm wide in the direction x of the dispersion of the spectrometer. The two sources were located with their centers 3 mm above and below the symmetry plane of the spectrometer (upper and lower source). The symmetry plane was determined experimentally through the requirement that the measured shift becomes zero for identical upper and lower sources. In addition, the vertical abberation error was eliminated by carrying out each measurement also with the upper and lower sources interchanged. The sources were aligned as well as possible on top of each other so that their reflections coincided very well. The residual displacement  $\partial x = 200$ , with 2 being the focal length of the spectrometer, could be allowed for through recording each reflection at positive  $(\Theta_R^+)$  and negative  $(\Theta_R^-)$ Bragg angle, because an energy shift affects the distance  $|\Theta_B^+|$  +  $|\Theta_B^-|$  of these reflections whereas a source displacement  $\partial x$  shifts both of these reflections by  $\partial\Theta$  keeping their distance unchanged.

Because of the small widths of the sources these contributed only  $\sim$  30 eV to the total experimental width, which was  $\Gamma\sim$  100 eV, because of the high quality of our curved crystal. Thus in the present experiment we have achieved  $\Gamma\sim$  1.3  $\Gamma_{nat}$  (Pb,  $K\alpha_1)^{-26}$ ).

## 5. Experimental results

Differences were measured of the energies of the  $K\alpha_1$  lines for all possible combinations of isotopically enriched (96.6%  $^{208}\text{Pb}$ , 92.4%  $^{207}\text{Pb}$ , 90.4%  $^{206}\text{Pb}$  and 70.9%  $^{204}\text{Pb}$ ) pairs of samples. Control measurements were also carried out on the  $^{206}\text{Pb}-^{208}\text{Pb}$  pair, where the shifts were measured of the  $K\alpha_2$  and  $K\beta_1$  lines and on the  $^{207}\text{Pb}-^{208}\text{Pb}$  pair, where the  $K\alpha_1$  shift was measured again at the end of all of the measurement series. The first control run was a check of the absence of a chemical shift, the latter was for the control of the absence of changes with time. The results of these tests agree in full with the  $^{206}\text{Pb}-^{208}\text{Pb}$   $K\alpha_1$  shift and the previous  $^{207}\text{Pb}-^{208}\text{Pb}$   $K\alpha_1$  shift, respectively.

The six directly measured shifts contain three basic differences:  $\delta < r^2 >_{207 \rightarrow 208}$ ,  $\delta < r^2 >_{206 \rightarrow 208}$  and  $\delta < r^2 >_{204 \rightarrow 208}$ . The six shifts were therefore subject to a level fit and then corrected for the isotopic composition of the samples. The obtained fitted

shifts of isotopically pure samples were corrected for mass shift according to eq. (6) resulting in Coulomb shifts that are listed in Table 1 together with the results of the CalTech group<sup>5</sup>).

Table 1: Coulomb shifts of the  $\mathrm{K}\alpha_1$  line of lead isotopes

Isotope	Isotope 2	Coulomb shift (meV) of $E\alpha_1(1)$ - $E\alpha_1(2)$			
"		This work	Ref. 5		
204	208	414 ± 17			
204	207	264 ± 17			
204	206	186 ± 20	200 ± 38		
206	208	228 ± 14	186 ± 18		
206	207	78 ± 14	50 ± 20		
207	208	150 ± 14	136 ± 25		
I		L			

While our data for the <sup>207</sup>Pb-<sup>208</sup>Pb and <sup>204</sup>Pb-<sup>206</sup>Pb pairs are consistent with the CalTech data, we find slightly larger shifts for the <sup>206</sup>Pb-<sup>208</sup>Pb and <sup>206</sup>Pb-<sup>207</sup>Pb pairs. The higher accuracy of the present data is due to the significantly better resolution that has been achieved in our measurement.

## 6. Comparison with optical data

For an optical transition i of the isotopes A and A+ $\delta$ A the atomic optical isotope shift is given in analogy to eq. (4) by

$$\delta E^{i} = \delta E^{i}_{coul} + \delta E^{i}_{mass} . \tag{15}$$

To a very good approximation  $\delta E_{\mbox{coul}}^{\mbox{i}}$  can be written as

$$\delta E_{\text{coul}}^{i} = \lambda \cdot C_{1}^{i} . \tag{16}$$

The similarity to eq. (10) is obvious. This is due to the fact that the ratios  $\mathrm{C_n/C_1}$  in eq. (11) are largely independent<sup>16</sup>) of the principal quantum numbers. However, whereas  $\mathrm{C_1}$  can be easily calculated<sup>16</sup>) for an X-ray transition, this is not the case for the optical transition. The mass shift is now expressed as

$$\delta E_{\text{mass}}^{i} = M^{i} \frac{\delta A}{A(A + \delta A)}$$
 (17)

so that eq. (15) reads

$$\delta E^{\dagger} = \lambda C_1^{\dagger} + M^{\dagger} \frac{\delta A}{A(A + \delta A)}$$
 (18)

The unknown parameters  $C_1^i$  and  $M^i$  can be determined with the use of the measured X-ray isotope shifts from which together with Seltzer's coefficients<sup>16</sup>) the nuclear parameter  $\lambda$  can be found according to eq. (10), provided that  $\delta E^i$  values have been measured for at least two pairs of isotopes. Then, with the aid of  $C_1^i$  and  $M^i$  the nuclear parameters can be obtained

also for those pairs of isotopes which cannot be investigated by X-ray shift studies but where optical measurements are feasible (see Fig. 1). As our X-ray shift data and the optical shift data $^8$ ) for the 405.8 nm line comprise three pairs of isotopes we can determine  $C_1$  and M through a fit:

$$C_1^{405\cdot8} = 0.45 \pm 0.15 \text{ fm}^{-2} \text{ cm}^{-1}$$

$$M^{405 \cdot 8} = 570 \pm 380 \text{ cm}^{-1}$$
.

These parameters imply that the total mass shift is about  $30 \pm 20$  percent of the observed optical shifts.

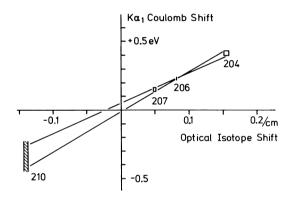


Fig. 1: Plot of the Coulomb shifts obtained from our measured  $K\alpha_1$  X-ray shifts allowing for mass shift according to eq. (13) versus the atomic optical shifts of the 405.8 nm line for the isotopes  $^{2.04}\text{Pb}$ ,  $^{2.06}\text{Pb}$  and  $^{2.07}\text{Pb}$  relative to  $^{2.08}\text{Pb}$ . The figure also contains the atomic optical shift of the 405.8 nm line of  $^{2.10}\text{Pb}$  relative to  $^{2.08}\text{Pb}$ . The figure clearly shows the small influence of the mass shift of the optical line and the larger increase of <r $^2>$  when going from  $^{2.08}\text{Pb}$  to  $^{2.10}\text{Pb}$  than when approaching  $^{2.08}\text{Pb}$  from  $^{2.06}\text{Pb}$ .

## 7. Comparison with other data and discussion

Detailed information about the isotopic changes of the nuclear Coulomb field can be obtained from muonic X-ray shift data that are rather large. The extraction of <r $^2>$  values from muonic data alone is model dependent, however. In a systematic study of the muonic X-ray shifts of the even isotopes of Sm good agreement was obtained with electronic X-ray and optical data $^{27}$ ). For the Hg isotopes, though, a major uncertainty arises $^{28}$ ) from the insufficient knowledge of the nuclear polarization. For the isotopes of Pb the nuclear polarization has been calculated in the RPA $^{29}$ ) with the conclusion that the

uncertainty of the calculation should be around 10%. Through a Fourier Bessel analysis of the elastic electron scattering data of 208Pb a model independent charge density distribution  $\rho(r)$  was derived which was used for the calculation of muonic binding energies<sup>30</sup>). From the difference between these and the measured energies, adjusted for all corrections, experimental nuclear polarization effects were determined. These appear to disagree<sup>30</sup>) with the prediction. Also the problem<sup>29</sup>) of the simultaneous explanation of the 1s nuclear polarization shift and the 2p splitting suggests that the nuclear polarization in muonic X-ray spectra of lead isotopes is not yet fully understood. When this problem has been solved, muonic and X-ray shift data can be combined in order to obtain more information about the shape of the nuclear charge density distribution and its isotopic variation e.g. for a Fermi distribution where variations of the half-density radius and skin thickness can be extracted.

Based on the analysis by Euteneuer who used (e,e)-data from Mainz and Stanford and muonic data by Kessler and coworkers<sup>31</sup>), Fricke, Miska and Rychel<sup>32</sup>) have carried out a simple preliminary determination of the nuclear parameters for the stable Pb isotopes and found the values listed in column 4 of Table 2.

Table 2: Nuclear parameters and changes of  $\langle r^2 \rangle$  in fm<sup>2</sup> for various isotopes of Pb

Nuclear parameter	Present result		Ref. 5		Ref. 32		δ <r²>theor</r²>
	ł		0.072			1	
λ <sub>206→208</sub>	0.121	0.008	0.099	0.010	0.128	0.004	0.116
λ <sub>204</sub> →208	0.220	0.009	0.205	0.022	0.236	0.007	0.226

The results of our X-ray isotope shift measurement are tabulated in column 2 and our theoretical predictions are given in the last column. The values in column 2 and column 3 can be compared directly. Those of column 4 are model dependent. The predicted  $\delta \! < \! r^2 \! >$  values differ from the  $\lambda$  values as given in eq. (11). The magnitude of this difference depends on the higher moment differences  $\delta < r^{2n} >$ . For a homogeneous charge distribution one has 33)

$$\delta < r^{2n} > = \frac{2n}{2n+3} R_0^{2n} \frac{\delta A}{A}$$
 (19)

with  $R_0 = 1.2 \text{ A}^{\frac{1}{3}}$  fm. With this model one obtains

$$C_1 \delta < r^2 >_{206 \rightarrow 208} = 367 \text{ meV}$$

$$C_2 \delta < r^4 >_{206 \to 208} = -29.5 \text{ meV}$$

and

$$C_3 \delta < r^6 >_{206 \to 208} = 4.6 \text{ meV}$$
,

which suggests that the approximation  $\lambda \sim \delta < r^2 >$ should be good to  $\sim$  7%. Keeping this uncertainty of the contributions of the higher moments of  $\rho(r)$  in mind, the experimental results can be compared with theory

The reduction of  $\langle r^2 \rangle$  when going from <sup>208</sup>Pb to  $^{207}\text{Pb}$  is due to the effect of the  $\text{p}_{1/2}$  neutron hole  $(Z\delta r^2 = 5.018 \text{ fm}^2)$  on the monopole mode of the Z=82 proton core for which r2 (proton, Woods Saxon) = 36.074 fm<sup>2</sup>. From these numbers one could deduce an effective neutron charge of 0.14. For the 206Pb-<sup>208</sup>Pb pair the measured X-ray shifts and the theoretical value are less than twice the respective 207Pb-<sup>208</sup>Pb shift values. This can be understood because also the  $f_{5/2}$ ,  $p_{3/2}$ ,  $f_{7/2}$ ,  $h_{9/2}$  and  $i_{1.3/2}$  two neutron holes contribute to the  $^{2.06}$ Pb ground state  $^{1}$ ) with smaller effective neutron charges than the p<sub>1/2</sub> neutron hole. A state-independent effective neutron charge does therefore not appear to be a good approximation.

For a more detailed comparison it would be valuable to have both experimental and theoretical results on at least the quantity  $\delta < r^4 >$ , because its effect is now comparable to the experimental errors.

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