# STATE DEPENDENT VOLUME ISOTOPE SHIFT ANALYSIS OF THE LOW LYING STATES OF Ba I and Ba II.

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Relativistic multi-configuration Dirac–Fock wavefunctions, coupled to good angular momentum $J$, have been calculated for low lying states of Ba I and Ba II. The resulting electronic factors show good agreement with data derived from recent high-resolution laser spectroscopy experiments and results from a comparison of muonic and optical data.

The electron density at the nucleus is a very important quantity in a number of measurements (isomer-shifts, optical-, X-ray, and muonic-isotope shifts), where electronic and nuclear operators describe the interaction [1]. In order to extract the nuclear charge distribution or the difference of this quantity between different isotopes from this kind of measurements, the knowledge of the electronic density at the nucleus is absolutely necessary. For a long time several groups have tried to extract nuclear charge distribution parameters from consistent exploration of muonic and optical spectroscopy data using ab-initio or semi-empirical estimations of the electron density at the nucleus. This is in principle possible as both experiments measure different integral quantities caused by differences of the nuclear and electronic charge densities.

In a theoretical treatment of the isotope shift (IS) it is important to include the effect of relativity and configuration interaction. In nearly all publications each of these effects have been analyzed separately, only. Ref. [2] is a good example of the discussion of relativity whereas ref. [3] is an extended but non-relativistic discussion of the influence of configuration interaction in the isotope-shift. Grant [4] has published an exploratory work of the influence of both effects on the volume isotope-shift. We present here the results of ab-initio relativistic state-dependent multi-configuration Dirac–Fock (MCDF) calculations for the charge density at the nucleus for low lying states of Ba I and Ba II. The calculations are performed with the MCDF-computer program of Desclaux [5]. In this type of calculations relativity as well as certain types of configuration mixing between open shell electrons are included directly in the selfconsistent procedure. This is absolutely necessary since both effects are large and affect each other.

The isotopic shift in an atomic transition $i$ is given as (neglecting higher order tems in the nuclear charge distribution) [1,2]:

$$\delta_{\nu_i}^{AA'} = F_i\, \delta \langle r^2 \rangle^{AA'} + M_i (m_{A'} - m_A)/m_A m_{A'} ,$$

where the first term represents the volume shift and the second term the mass shift. $F_i$ is the electronic factor of the volume shift and $\delta \langle r^2 \rangle^{AA'}$ the difference of nuclear mean-square charge radii. In this work we will calculate the volume-isotope shift, only.

Since the electronic charge density varies strongly over the nuclear volume we use an extended nucleus with a two-parameter Fermi-nuclear charge distribution [6] and 21 mesh-points inside the nuclear radius. The electronic charge densities presented in this paper are always values at the center of the nucleus. The five lowest levels with positive parity and the 16 lowest levels with negative parity, all originating from the atomic levels $6s_{1/2}$, $6p_{1/2}$, $6p_{3/2}$, $5d_{3/2}$, and $5d_{5/2}$,

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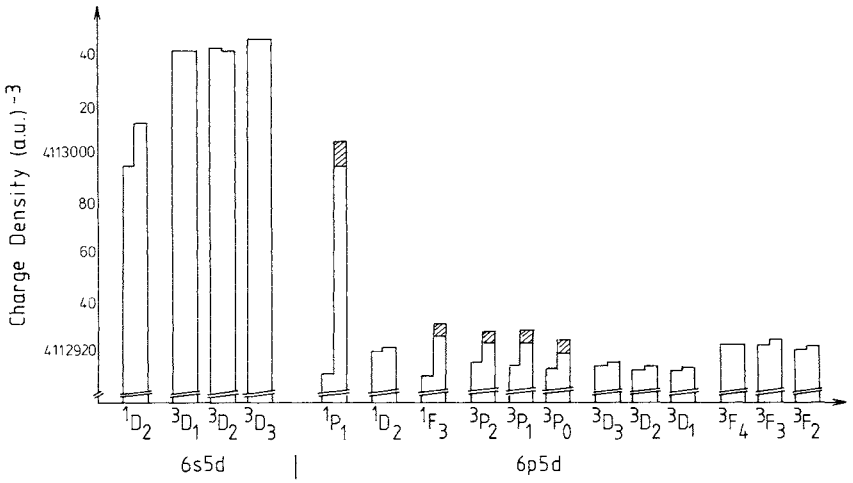


Fig. 1. Electron densities at the nucleus for Ba I in the configuration 6s5d and 6p5d calculated with an MCDF program. For each state two different values are shown in the columns. The values to the left have been calculated for each state by mixing within the 6s5d and 6p5d configurations, respectively. The values to the right are obtained by mixing of the configurations resulting from the wavefunctions 6s, 6p, 5d. The shaded area shows the uncertainty due to convergence problems.

coupled to good angular momentum $J$, were calculated. In addition, calculations were performed for the $6s_{1/2}$, $6p_{1/2}$, and $6p_{3/2}$ states of Ba II. These results are compared with experimental volume shifts of each combination of optical transitions in the low lying spectrum of Ba I derived from recent high-resolution laser spectroscopy measurements [2,7]. In addition electronic factors are also available from muonic data [8] which are used for a further check of the calculations.

The electronic charge density $4\pi\rho$ at the nucleus for the lowest states of positive parity (6s5d configuration) is given in fig. 1. For each state the value obtained for coupling within the main configuration is given to the left of each column. To the right in each column in fig. 1 we give the electronic charge density obtained by mixing of different configurations with the same parity, i.e. ($6s^2$, 6s5d, $6p^2$, $5d^2$) and (6s6p, 6p5d). There is a significant change by including full mixing (i.e. taking into account all possible states resulting from the levels discussed here), particularly for the singlet states. For example the $^1P_1$ states designated as 6s6p and 6p5d changed by −75 and +90 (in units used in fig. 1) when the admixtures of these configurations were taken into account.

In the analysis of isotope shifts a King plot procedure is normally performed. The isotope shifts in one transition $i$ are plotted as a function of the corresponding isotope shifts in another transition $j$, known as the reference. The slope of the resulting line is given as the ratio between the changes in electron density at the nucleus $\Delta\rho_i/\Delta\rho_j$ or in terms of the factors $F_i/F_j$ for the two transitions $i$ and $j$. Following this type of analysis of the high-resolution laser spectroscopy measurements between the 6s5d and 6p5d configurations

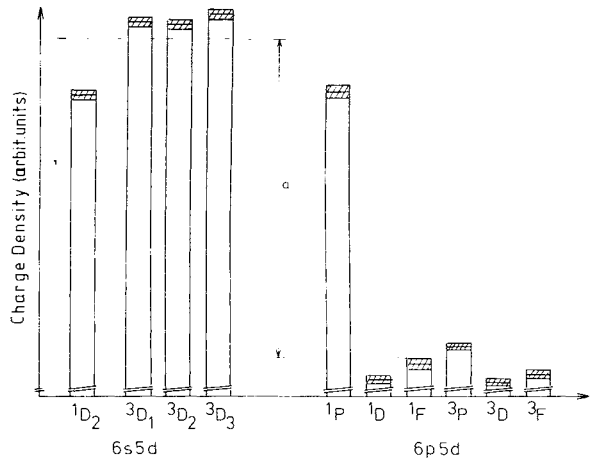


Fig. 2. Experimental electron densities at the nucleus as evaluated from experimental high-resolution laser spectroscopy measurements. The shaded area shows the experimental uncertainty.

Table 1
Comparison of experimental and theoretical slopes of lines in a King plot analysis with the $6s^2\ ^1S_0-6s6p\ ^3P_1$ transition at 7911 Å as the reference.

| Spectra | Transition | Wavelength (Å) | Single config. DF | MCDF | Exper. ref. [7] |
|---|---|---|---|---|---|
| Ba I $6s^2-6s6p$ | $^1S_0-^1P_1$ | 5535 | 0.586 | 1.175 | 1.176(9) |
| Ba I | $^1D_2-^1P_1$ | 5826 | 0.653 | 0.11(5) | −0.008(13) |
| 6s5d–6p5d | $^1D_2-^3D_3$ | 7359 | 0.628 | 0.828 | 0.836(12) |
| | $^1D_2-^3D_1$ | 7812 | 0.646 | 0.848 | 0.827(36) |
| | $^3D_3-^1D_2$ | 7417 | 0.983 | 1.064 | 1.053(11) |
| | $^3D_3-^3F_3$ | 7488 | 0.965 | 1.027 | 1.043(11) |
| | $^3D_1-^1D_2$ | 7120 | 0.947 | 1.025 | 1.029(17) |
| Ba II 6s–6p | $^2S_{1/2}-^2P_{1/2}$ | 4934 | 1.357 | 1.471 | 1.471(44) |

the relative difference in the charge density at the nucleus for the states in the two configurations was derived [7]. The result is shown in fig. 2. The distance "$a$" in the figure corresponds to the average change in the electron density $\Delta\rho$ (6s5d–6p5d) given in arbitrary units. The deviation from the average for each state is clearly observed. We notice how the charge densities, evaluated from the MCDF wavefunctions displayed in fig. 1, resemble the experimental structure in fig. 2. We also observe the significant improvement achieved when the full MCDF wavefunctions are used compared with DF-calculations within one configuration (fig. 1). The comparison between the theoretical and experimental results are explicitly given in table 1. The values are presented as ratios $F_i/F_j$ (i.e. slopes in a King plot analysis) of different transitions, with the $6s^2\ ^1S_0 - 6s6p\ ^3P_1$ transition as the reference. The experimental values are fully reproduced within the error by the MCDF-calculations. This is even true for transitions to the strongly disturbed $^1P_1$ states.

However, to be able to extract the change in the mean-square radius $\delta\langle r^2\rangle$ from the measured isotope shifts, known values of the specific mass shift and the electronic factor $F_i$, not the ratio $F_i/F_j$, of the volume shifts are required. In table 2 $F_i$ values for some transsitions, calculated with different ab-initio methods, are given.

The values in the fourth column have been calculated

Table 2
Comparison of different theoretical ab initio calculations of the changes in electron density at the nucleus for some transitions.

| Spectra | Transition | Wavelength (Å) | $F_i$ (GHz/fm$^2$) | | | |
|---|---|---|---|---|---|---|
| | | | Average DF ref. [7] | Single config. DF | MCDF | Exper. ref. [8] |
| Ba I $6s^2-6s6p$ | $^1S_0-^3P_1$ | 7911 | −2.34 | −2.67 | −2.55 | −2.59(22) [a] |
| | $^1S_0-^1P_1$ | 5535 | | −1.56 | −2.99 | −3.04(26) |
| Ba II 6s–6p | $^2S_{1/2}-^2P_{1/2}$ | 4934 | −3.48 | −3.76 | | −3.80(33) [a] |

[a] These values have been calculated from the value for the 5535 Å transition [3.04(26)] using the experimental slopes in table 1.

from Dirac–Fock (DF) wavefunctions [7] evaluated for the average energy of the terms achieved in the particular configuration [9]. These values have been evaluated assuming a uniform charge distribution with the nuclear radius $R_A = 1.2A^{1/3}$ fm. The single DF and MCDF values presented in column 5 and 6 have been calculated with a nuclear Fermi charge distribution. The values are given as the electronic factor corrected for the change of the electronic charge density over the nuclear volume. In connection with recent muonic X-ray measurements a King plot analysis was performed [8]. In a plot of the optical isotope shift in the 5535 Å transition versus the muonic $\delta\langle r^2\rangle$ values the electronic factor $F_{5535}$ was estimated to be in good agreement with our result evaluated with the MCDF wavefunctions (table 2). The experimentally derived electronic factors for the 4934 Å and 7911 Å transitions – using $F_{5535}$ and the experimental slopes in table 1 – are also included in table 2. Some years ago Fischer et al. [10] estimated the factor $F_{4934} = -4.98$ GHz/fm$^2$ by use of magnetic hyperfine structure data. It should, however, be noted that the magnetic hyperfine operator and the fieldshift operator are relativistically quite different [11] and it is necessary to evaluate the electronic factor in the correct way. In addition, polarization and correlation effects affect the two operators differently. In the evaluation of nuclear mean-square radii from measured isotope shifts in the $6s^2\,{}^1S_0 \rightarrow 6s6p\,{}^1P_1$ transition the semi-empirical value by Fischer et al. [10] has been used as the reference in the calculation of the electronic factor [12,13]. Just recently another value $F_{5535} = -3.99\,(65)$ GHz/fm$^2$ of the electronic factor in the 5535 Å transition has been achieved from muonic data [14]. This value, with a rather big uncertainty, is in good agreement with the old semi-empirical value of Fischer et al. [10]. More consistent results of electronic factors derived from muonic experiments are therefore necessary for a further check.

From the comparison performed in this work it is evident that ratios of the change in electron density at the nucleus between different transitions as well as the absolute magnitude of the density can be well described by state dependent MCDF calculations. For this type of calculation it is not sufficient to use quantities evaluated for the average configuration and by including only the state dependency within one configuration but also the mixing of different configurations. The MCDF approach seems to work well for the low lying states, while calculations for higher lying states will probably be more difficult due to a strong mixing of many states. A more extensive analysis of the volume isotope shifts as well as the fine structure, the magnetic dipole- and electric quadrupole-interaction will be given in a forthcoming paper.

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