

## State-dependent volume isotope shifts of low-lying states of group-IIa and -IIb elements

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Results of relativistic multiconfiguration Dirac-Fock calculations with an extended nucleus are used to analyze the volume isotope shifts of the resonance transitions in the group-IIa and -IIb elements as well as in Yb. This is done together with a review of the isotope shift theory, including a critical evaluation and comparison of the semiempirical calculation of volume isotope shifts commonly used today. Electronic factors  $F_i$ , proportional to differences of electronic densities over the nuclear volume, are discussed within various approximations and compared with experimental results.

### I. INTRODUCTION

For many elements systematic measurements of optical isotope shifts (IS) and hyperfine-structure splittings (hfs) in spectral lines for series of stable as well as radioactive isotopes have been carried out for a long time.<sup>1-4</sup> During the last years many of these old investigations have been extended considerably<sup>5-8</sup> due to the access to good narrow-band tunable-laser radiation in broad wavelength regions. In particular alkali-metal, alkaline-earth (group-IIa), and alkaline-earth-like atoms (group-IIb) have been studied with use of different laser spectroscopic techniques with ever-increasing accuracy.

Parallel to these experimental developments, theoretical methods and computer programs for treatment of atomic systems within the self-consistent-field (SCF), Hartree-Fock (HF),<sup>9,10</sup> Dirac-Fock (DF),<sup>11,12</sup> multiconfiguration Hartree-Fock (MCHF), and multiconfiguration Dirac-Fock (MCDF)<sup>9,13-16</sup> approaches have been developed. In addition many-body perturbation theory (MBPT) approaches<sup>17,18</sup> have been developed further, nonrelativistically as well as relativistically. This type of *ab initio* atomic calculation opens the possibility to test semiempirical methods<sup>1,2,19-22</sup> for the derivation of electronic properties such as breakdown of *LS* coupling, perturbation from different configurations, and different expectation values.

Examples of areas where semiempirical approaches have been used for a long time is the analysis of experimental hfs and IS data. The possibility of doing *ab initio* calculations eliminates the need of the semiempirical approach. As the semiempirical approach has been used very frequently with relatively good success the *ab initio* calculations can be used to test the validity of that approach. Further, detailed evaluations of the electronic part of the hfs and the volume IS will give accurate values for nuclear properties like nuclear radial moments and change of charge radii between isotopes, respectively. These calculated nuclear quantities can then be compared

with results of other fields like the study of muonic and electronic x-rays or electron scattering<sup>23-25</sup> as a check of the evaluation procedure. To summarize, hfs and IS are excellent properties for testing of and linking together *ab initio* semiempirical methods and electronic-nuclear properties.

Recently the electronic factors of the volume isotope shift were calculated in an *ab initio* way with the DF and the MCDF method for states in Ba I, Ba II,<sup>26</sup> and Au I.<sup>27</sup> In this paper we extend those calculations to all other elements in group IIa and group IIb of the periodic table as well as Yb. Usually IS for these elements have been measured in the resonance lines of the ionized system. Calculations have, therefore, been done for some low-lying states of the second spectrum as well.

The paper is organized as follows. Section II gives a short review of the MCDF method and isotope shift theory including a review of the semiempirical approaches. A presentation of the results of the calculations and comparison of experimental data is presented in Sec. III, with the conclusions in Sec. IV.

### II. THEORETICAL APPROACH

#### A. Multiconfiguration Dirac-Fock method

The MCDF method is well known.<sup>15,16</sup> Two computer programs by Desclaux<sup>13</sup> and by Grant *et al.*<sup>14</sup> now exist so that a brief description should be sufficient here.

In its most commonly used form the MCDF method starts from the following zeroth-order Hamiltonian

$$H = \sum_i H_D(i) + \sum_{\substack{i,j \\ i < j}} r_{ij}^{-1},$$

i.e., a sum of one-electron Dirac operators  $H_D$  plus the classical Coulomb repulsion between the electrons. The Dirac equation which has to be solved is

$$H |\Psi\rangle = E |\Psi\rangle.$$

The total wave function  $|\Psi\rangle$  for a given atom is expressed as a linear combination of configuration state functions (CSF)  $|\phi_i\rangle$ ,

$$|\Psi\rangle = \sum_i W_i |\phi_i\rangle, \quad (1)$$

where each of the CSF is a simultaneous eigenstate of the total angular momentum  $J^2$ , its projection  $J_z$ , and the parity operator. This is achieved by a linear combination of Slater determinants  $|\alpha_j\rangle$ :

$$|\phi_i\rangle = \sum_j \beta_{ij} |\alpha_j\rangle. \quad (2)$$

The Slater determinants  $|\alpha\rangle$  are constructed from  $N$  one-particle wave functions  $\varphi$  which in the relativistic case are spinors of rank four,

$$\varphi = \begin{pmatrix} \frac{P_{n\kappa}(r)}{r} \chi_{\kappa}^{m_j} \\ i \frac{Q_{n\kappa}(r)}{r} \chi_{-\kappa}^{m_j} \end{pmatrix}. \quad (3)$$

$P_{n\kappa}(r)/r$  and  $Q_{n\kappa}(r)/r$  are the large and small components of the radial wave functions. The angular part is a linear combination of spherical harmonics  $Y_l^m$  and the spin function  $S^{m_s}$  which is a spinor of rank 2:

$$\chi_{\kappa}^{m_j} = \sum_{\sigma} Y_l^{m_j - \sigma} S^{\sigma} \langle l, m_j - \sigma, \frac{1}{2}, \sigma | j, m_j \rangle.$$

Using the variational principle to achieve the minimum in the total energy of the whole atom both the mixing coefficients  $W_i$  and the radial components  $P(r)$  and  $Q(r)$  of the Dirac spinors are optimized in a self-consistent process. Since this variation is performed usually with respect to the radial part of the wave function only, this is a restricted Dirac-Fock procedure. This means that the radial wave functions are the same for all values of  $m_j$  for a given  $j$ .

In more sophisticated calculations, concerning especially good total energies, additional contributions like spin-spin, spin-other-orbit, and retardation are added in perturbation theory via the expectation value of the Breit operator.<sup>28-32</sup> The same is done with the contribution of the lowest-order vacuum polarization potential as well. The influence of vacuum fluctuation, which is the main part of the quantum electrodynamic (QED) correction in electronic atoms<sup>33,34</sup> up to now, is included in a heuristic manner only.<sup>31,32</sup> This approach implies that the SCF wave functions do not include the effect of the Breit and QED operators.

Of course, by far the largest influence on the wave function at the origin,  $r=0$ , comes from the extended nucleus. We use a Fermi-type charge distribution of the nuclear charge with the best-known half-density radii  $R_C$  and skin thickness  $t$ .<sup>35</sup>

## B. Isotope shift theory

The measured isotope shift in optical spectral lines or in electronic and muonic x rays is given as the sum of the volume (field) isotope shift and mass shift where the latter is composed of the normal mass shift (NMS) and specific mass shift (SMS),<sup>1-4,36</sup> i.e.,

$$\delta\nu^{A_1 A_2} = \delta\nu_{\text{FS}}^{A_1 A_2} + \delta\nu_{\text{MS}}^{A_1 A_2},$$

where  $A_1$  and  $A_2$  denote the mass number of two isotopes. For a certain transition the NMS can be evaluated exactly<sup>1,4</sup> while usually some semiempirical procedure is used in the estimation of the SMS.<sup>4</sup> Some *ab initio* calculations of the SMS have been done using the HF<sup>37</sup> and MCHF methods<sup>38,39</sup> as well as many-body calculations for light atoms.<sup>40,41</sup> Correcting the measured IS for the NMS and SMS gives the remaining part, i.e., the volume isotope shift or field shift.

Generally, the volume isotope shift  $\delta\nu_{\text{FS}}$  can be written as a sum of the four integrals

$$\begin{aligned} \delta\nu_{\text{FS}} = & -e^2 \iint d\mathbf{r} d\mathbf{r}' \frac{1}{|\mathbf{r}-\mathbf{r}'|} \\ & \times [\rho_{A_1}(\mathbf{r})\rho_{e_1}(\mathbf{r}') - \rho_{A_1}(\mathbf{r})\rho_{e_2}(\mathbf{r}') \\ & - \rho_{A_2}(\mathbf{r})\rho_{e_1}(\mathbf{r}') + \rho_{A_2}(\mathbf{r})\rho_{e_2}(\mathbf{r}')], \end{aligned} \quad (4)$$

where  $\rho_{A_i}$  is the nuclear charge distribution of the two isotopes  $A_1$  and  $A_2$ , and  $\rho_{e_i}$  the electronic charge distribution of two electronic states  $e_1$  and  $e_2$ . Usually a spherical nucleus is assumed which changes the denominator from  $|\mathbf{r}-\mathbf{r}'|$  to  $r_>$ , which means the larger of the two values  $|\mathbf{r}|$  and  $|\mathbf{r}'|$  during the integration. The electron density within the nucleus, i.e.,  $r < R$ , normally is expressed as a polynomial

$$\rho = a_0 + a_2 r^2 + a_4 r^4 + \dots \quad (5)$$

Each of the above integrals in Eq. (4) can be expressed with this ansatz as

$$\frac{2\pi}{3} Z e^2 a_0 \langle r^2 \rangle^A + \frac{\pi}{5} Z e^2 a_2 \langle r^4 \rangle^A + \frac{2\pi}{21} Z e^2 a_4 \langle r^6 \rangle^A + \dots$$

and if we take the difference of the four integrals in Eq. (4) we get

$$\begin{aligned} \delta\nu_{\text{FS}} = & \frac{2\pi}{3} Z e^2 (\Delta a_0 \Delta \langle r^2 \rangle^{A_1 A_2} + \frac{3}{10} \Delta a_2 \Delta \langle r^4 \rangle^{A_1 A_2} \\ & + \frac{1}{7} \Delta a_4 \Delta \langle r^6 \rangle^{A_1 A_2} + \dots) \end{aligned} \quad (6)$$

with  $\Delta a_0$  the difference of the electronic densities in the two configurations at  $r=0$ , usually written as  $\Delta |\Psi(0)|^2$ , and  $\Delta \langle r^\alpha \rangle^{A_1 A_2}$  the difference of the expectation values of  $r^\alpha$  over the nuclear charge distribution of the two nuclei. Expression (6) can be written in various ways:<sup>4,42-44</sup>

$$\begin{aligned}\delta v_{\text{FS}} &= C_1 \left[ \Delta \langle r^2 \rangle^{A_1 A_2} + \frac{C_2}{C_1} \Delta \langle r^4 \rangle^{A_1 A_2} + \frac{C_3}{C_1} \Delta \langle r^6 \rangle^{A_1 A_2} + \dots \right] \\ &= C_1 \lambda = C_1 \Delta \langle r^2 \rangle^{A_1 A_2} \left[ 1 + \frac{C_2}{C_1} \frac{\Delta \langle r^4 \rangle^{A_1 A_2}}{\Delta \langle r^2 \rangle^{A_1 A_2}} + \frac{C_3}{C_1} \frac{\Delta \langle r^6 \rangle^{A_1 A_2}}{\Delta \langle r^2 \rangle^{A_1 A_2}} + \dots \right] \\ &= F \Delta \langle r^2 \rangle^{A_1 A_2} K\end{aligned}\quad (7)$$

with  $\lambda$  the so-called nuclear parameter.  $F$  is the electronic factor which is proportional to the electron density difference in the two configurations at  $r=0$ ,

$$F = \frac{2\pi}{3} Z e^2 \Delta |\Psi(0)|^2. \quad (8)$$

The values  $C_2/C_1$  and  $C_3/C_1$ , etc. can be expressed in terms of  $\Delta a_2/\Delta a_0$  and  $\Delta a_4/\Delta a_0$ . The change of electron density at the nucleus between two configurations can be attributed nearly totally to the direct and indirect change of the  $s$  and  $p_{1/2}$  densities, which all more or less have a parabolic behavior at  $r=0$ . Thus the quotient  $\Delta a_2/\Delta a_0$  is very much independent of the specific configuration as long as the neutral atoms are concerned, so that it is equal to  $a_2/a_0$  (and analogous the other quotients); then

$$\frac{C_2}{C_1} = \frac{3}{10} \frac{\Delta a_2}{\Delta a_0} = \frac{3}{10} \frac{a_2}{a_0}$$

and

$$\frac{C_3}{C_1} = \frac{1}{7} \frac{\Delta a_4}{\Delta a_0} = \frac{1}{7} \frac{a_4}{a_0}.$$

This fact allows us to calculate the expression in the large parentheses on the second line of Eq. (7)—defined there as  $K$ —as a function of the atomic system and thus as function of  $Z$  if we use an estimation of the term  $\Delta \langle r^4 \rangle^{A_1 A_2} / \Delta \langle r^2 \rangle^{A_1 A_2}$ , etc. Using the liquid drop model with the nuclear radius  $R = r_0 A^{1/3}$  (usually  $r_0$  is taken as 1.2 fm) we arrive at the expression

$$K = K(A) = 1 + \frac{C_2}{C_1} \frac{10}{7} R^2 + \frac{C_3}{C_1} \frac{5}{3} R^4 + \dots \quad (9)$$

In the earlier analysis of the x-ray isotope shift by Seltzer<sup>42</sup> he calculated the  $C_2/C_1$  and  $C_3/C_1$  factors from the  $1s$  wave functions only. The relatively small differences between his values and ours (given in Sec. III), which are calculated from neutral atoms, show that indeed the main contribution comes from the  $1s^2$  shell.

To summarize the expression for the volume isotope shift we get

$$\delta v_{\text{FS}} = F \Delta \langle r^2 \rangle^{A_1 A_2} K. \quad (10)$$

The electronic factor  $F$  for the volume isotope shift as well as the correcting factor  $K$  are known if one has calculated the charge density at the nucleus plus the expansion of the charge density over the nuclear volume according to Eq. (5) using the *ab initio* wave functions. However, evaluations of the electronic factor are performed mostly in a semiempirical way<sup>1,2,4,45</sup> as reviewed in Sec. II C.

### C. Semiempirical estimate of $F_i$

In the semiempirical approach the change of the density at the nucleus  $\Delta |\Psi(0)|^2$  in Eq. (8) is obtained from various experimental quantities containing  $|\Psi(0)|^2$ . The first step in this approach is to factorize  $\Delta |\Psi(0)|^2$  into the product of the difference of the total nonrelativistic charge density for a point nucleus and a function  $f(Z)$ , which takes care of the relativistic corrections to  $F_i$  for a certain type of nuclear charge distribution, i.e.,

$$\begin{aligned}F_i &= \frac{2\pi}{3} Z e^2 \Delta |\Psi(0)|^2 = \frac{\pi a_0^3}{Z} \Delta |\Psi(0)|_{\text{nr}}^2 f(Z) \\ &= E_i f(Z),\end{aligned}\quad (11)$$

where we have followed the convention used by Heilig and Stuedel.<sup>4</sup>

Normally  $f(Z)$  is taken from a paper by Babushkin<sup>46</sup> in which he calculates the quantity  $C_{\text{unif}}^{AA'}$ , which is the main part of  $f(Z)$ , for a uniform and, trapezoidal nuclear charge distribution. (Recently Zimmermann<sup>47</sup> has pointed out that there is an error in the formulas of Babushkin<sup>46</sup> and that an additional normalization constant should be added.) In this way the problem is reduced to the estimate of the difference of the total nonrelativistic (nr) charge density for a point nucleus at  $r=0$ , i.e.,  $\Delta |\Psi(0)|_{\text{nr}}^2$ , which will now be related to the charge density of the  $ns$  valence electron.

If we consider a transition for an alkali-metal atom, the difference of the total charge density between the two electronic states  $ns \rightarrow np$  can be expressed as

$$\begin{aligned}\Delta |\Psi(0)|_{ns \rightarrow np}^2 &= |\Psi(0)|_{\text{core}}^2 + |\Psi_{ns}(0)|^2 \\ &\quad - |\Psi(0)|_{\text{core}'}^2 - |\Psi_{np}(0)|^2 \\ &\approx \beta |\Psi_{ns}(0)|^2\end{aligned}\quad (12)$$

with

$$\beta \approx \frac{|\Psi(0)|_{\text{core}}^2 + |\Psi_{ns}(0)|^2 - |\Psi(0)|_{\text{core}'}^2}{|\Psi_{ns}(0)|^2},$$

where in the last relation the contribution from the  $np$  electron is neglected. The screening factor  $\beta$ , which takes into account the screening of the core electrons when the valence electron is excited, normally is taken from HF or DF calculations. If no isotope-shift measurements in alkali-metal-like transitions are available Blaise and Stuedel<sup>48</sup> (quoted by Heilig and Stuedel<sup>4</sup>) have given the following empirical screening ratio to be used with  $n=6$  or 7.

$$\frac{F_i((\text{core} + ns^2) - (\text{core} + nsnp))}{F_j((\text{core} + ns) - (\text{core} + np))} \approx 0.65 \quad (13)$$

with an uncertainty of 10%.

Finally, in the evaluation procedure of  $F_i$  the nonrelativistic value of  $|\Psi_{ns}(0)|_{nr}^2$  for the outermost electron has to be estimated. This usually is done in an empirical way by the following schemes.

As described by Kopfermann<sup>1</sup> the fine-structure level scheme can be used to calculate  $|\Psi_{ns}(0)|_{nr}^2$  by the Goudsmit-Fermi-Segré formula

$$|\Psi_{ns}(0)|_{nr}^2 = \frac{1}{\pi a_0^3} \frac{ZZ_a^2}{n_a^3} \left[ 1 - \frac{d\sigma}{dn} \right] \quad (14)$$

with  $Z_a = 2$  for the alkaline-earth ions,  $n_a$  the effective quantum number,  $\sigma$  the quantum defect, and  $a_0$  the Bohr radius. Although Kopfermann<sup>1</sup> claims that the charge density calculated with Eq. (14) is a nonrelativistic charge density this is probably not too correct, as the whole equation is a very rough approximation only.

Further, if the magnetic dipole interaction constant  $a_{ns}$  is known in the ground state, the following relation has been given:<sup>12</sup>

$$a_{ns}^{\text{expt}} = \frac{16\pi}{3} \frac{\mu_0}{4\pi} \mu_B |\Psi_{ns}(0)|_{nr}^2 \frac{\mu_I}{I} F(ns_{1/2})(1-\delta)(1-\epsilon), \quad (15)$$

where  $\mu_B$  is the Bohr magneton,  $I$  the nuclear spin, and  $\mu_I$  the nuclear magnetic dipole moment.  $F(ns_{1/2})$  is a relativistic correction factor for the magnetic dipole interaction.<sup>1,20,12</sup> The factors  $(1-\delta)$  and  $(1-\epsilon)$  represent the Breit-Rosenthal<sup>49</sup> and Bohr-Weisskopf factors,<sup>50</sup> respectively. These take into account the effect of an extended nuclear charge distribution and the extended distribution of the magnetic moment. The nonrelativistic charge density thus is given by

$$|\Psi_{ns}(0)|_{nr}^2 = \frac{a_{ns}^{\text{expt}}}{\frac{16\pi}{3} \frac{\mu_0}{4\pi} \mu_B \frac{\mu_I}{I} F(ns_{1/2})(1-\delta)(1-\epsilon)}. \quad (16)$$

The  $F_i$  values are then obtained by introducing the  $|\Psi_{ns}(0)|_{nr}^2$  values of Eqs. (14) and (16) into Eq. (12) and then (11).

Instead of using an *ab initio* value for  $\Delta|\Psi(0)|^2$ , in both approaches a mixture of experimental, pure theoretical and semiempirical values is used. Here it is interesting to note that in a nonrelativistic treatment, both the magnetic dipole interaction and the volume isotope expression are assumed to be proportional to the nonrelativistic value of  $|\Psi_{ns}(0)|_{nr}^2$ .

However, this does not hold in a relativistic treatment due to the different tensorial structure of the two interactions as discussed for example by Bauche.<sup>51</sup> This question is also discussed in a recent paper by Blundell *et al.*<sup>52</sup>

A relativistic treatment gives the following contribution to the magnetic dipole interaction from the outermost  $ns$  electron in an alkali-metal atom:

$$4\pi |\Psi_{ns}^M(0)|_{\text{rel}}^2 = \frac{2}{\alpha a_0} \int_0^\infty \frac{P_{ns}(r)Q_{ns}(r) + P_{ns}(r)Q_{ns}(r)}{r^2} dr$$

$$= F(ns_{1/2})(1-\delta)4\pi |\Psi_{ns}(0)|_{nr}^2 \quad (17)$$

while the contribution from the outermost  $ns$  electron in the volume isotope shift analysis is

$$4\pi |\Psi_{ns}(0)|_{\text{rel}}^2 = \lim_{r \rightarrow 0} \left[ \frac{P_{ns}(r)^2 + Q_{ns}(r)^2}{r^2} \right]$$

$$= R_{ns} 4\pi |\Psi_{ns}(0)|_{nr}^2. \quad (18)$$

It should be noted that we have included the Breit-Rosenthal correction in Eq. (17) because our calculations of  $P$  and  $Q$  have been done for an extended nucleus.

### III. RESULTS AND DISCUSSIONS

#### A. Electronic structure

The group-IIa and -IIb elements are characterized by an outermost closed  $ns^2$  shell configuration in the ground state. Excitation of one of these electrons results in simple two-electron systems with quite different wave functions for the singlet  $^1P$  and triplet  $^3P$  states, already noticed by Hartree *et al.*<sup>53</sup> a long time ago in  $LS$ -dependent HF calculations. Some recent calculations for these systems can be found in Refs. 9 and 54. Schematic experimental level energy diagrams of the lowest states<sup>55,56</sup> for the first spectra of those elements are presented in Figs. 1(a) and 1(b) for the group-IIa and -IIb elements, respectively. In Fig. 1(b) we include the element Yb as well, which has an analogous ground state  $4f^{14}6s^2$ .

As a general trend, the levels for the even as well as the odd configurations of the IIa elements in Fig. 1(a) move together closer in heavier elements. For the very heavy element Ra the trend is reversed again, due to the influence of strong relativistic effects. This demonstrates that it is absolutely necessary to include relativity and configuration interaction on an equal basis and to evaluate wave functions with one-electron angular momenta  $j$ , coupled to good total  $J$  within the particular configurations. In our MCDF calculations those levels of positive and negative parity as well as good  $J$  were taken into account, which can be constructed from the  $ns_{1/2}$ ,  $np_{1/2}$ ,  $np_{3/2}$ ,  $(n-1)d_{3/2}$ , and  $(n-1)d_{5/2}$  one-electron wave functions.

The permitted resonance line corresponds to the transitions  $ns^2\ ^1S_0 \rightarrow nsnp\ ^1P_1$  in the single  $LS$ -configuration description. Due to the breakdown of  $LS$  coupling a mixture will take place between the  $^3P_1$  and  $^1P_1$  states, which means that measurements in the transition  $ns^2\ ^1S_0 \rightarrow nsnp\ ^3P_1$  may be done. The other  $^3P_0$  and  $^3P_2$  states are metastable and can be populated in a plasma discharge oven or in a radio frequency discharge.<sup>57-59</sup> Furthermore, populations in these metastable states open up the possibility of doing measurements in spectral lines connected with these states. This was done, for example, in Ca (Ref. 57) and Cd (Ref. 59) in transitions connected with the  $ns(n+1)s\ ^3S_1$  state. As can be seen in Fig. 1(a) the  $^1D$  and  $^3D$  states originating from the  $ns(n-1)d$  con-

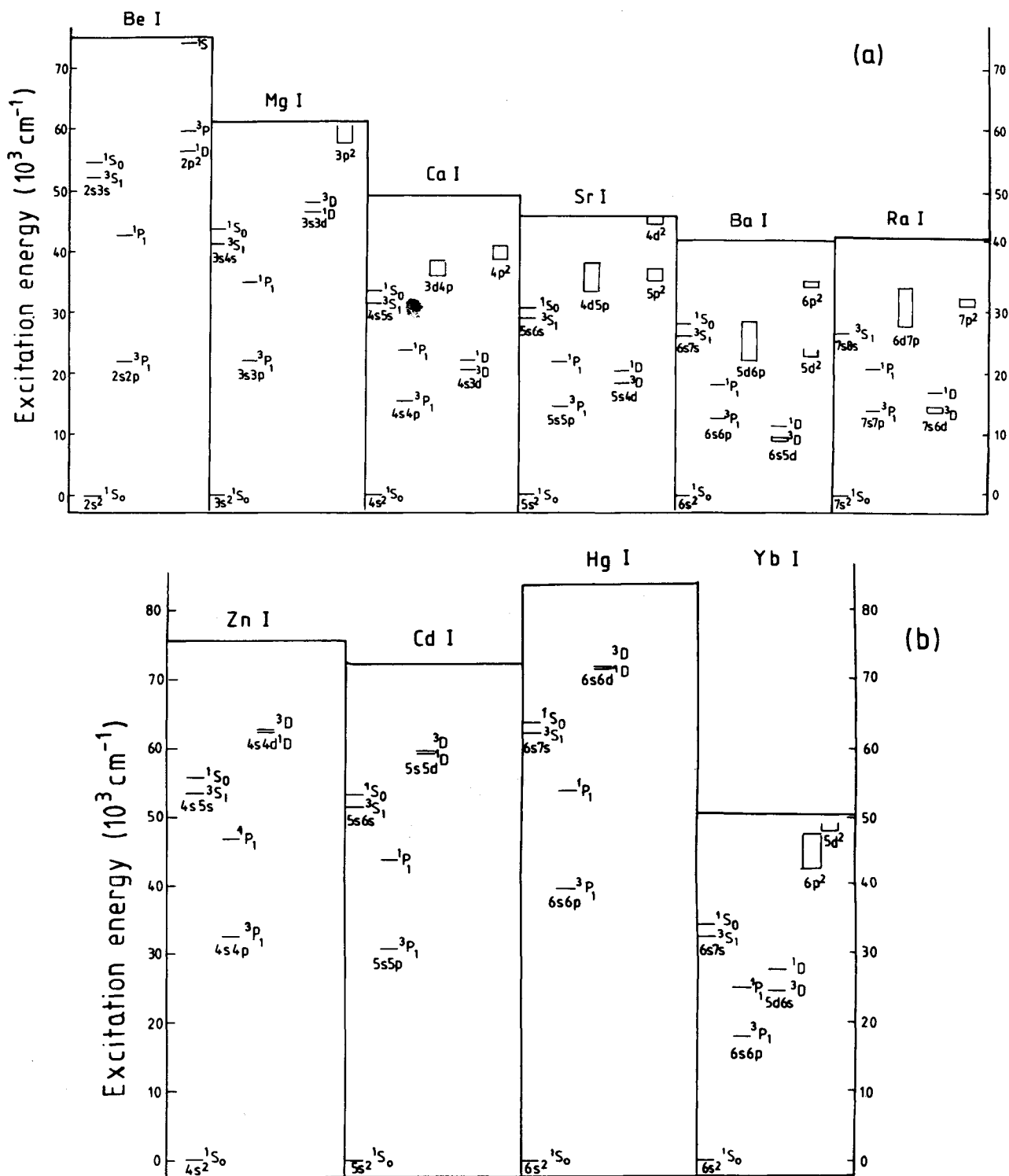


FIG. 1. (a) Schematic energy-level diagram of the lowest configurations of the neutral group-IIa elements. (b) Schematic energy-level diagram of the lowest configurations of the neutral group-IIb elements and Yb.

figuration are bound even stronger than the  $^1P$  and  $^3P$  states, which originate from the  $nsnp$  configuration, in heavier elements, e.g., Ba. This implies that these states are metastable for heavier elements and may act as a platform for investigations of transitions to the  $np(n-1)d$  configuration. Extensive measurements of this kind have been performed recently in Ba.<sup>60,61</sup>

The energy-level diagrams for the group-IIb elements, shown in Fig. 1(b), are quite different. The configuration  $nsnd$  is located for all elements at higher energy than the states in the  $nsnp$  configuration. The  $nsnd \ ^1D$  and  $^3D$  states are close in energy and reversed. The  $npnd$ ,  $np^2$ , and  $nd^2$  configurations have not been observed to be bound for these elements. The reason for this difference

in term structure of the group-IIa and -IIb elements is probably the presence of the occupied shell of  $(n-1)d$  electrons just below the two valence electrons in the group-IIb elements. The level scheme for Yb, however, reminds one more of the IIa elements.

The results for the single-configuration and multiconfiguration Dirac-Fock calculations for the group-IIa and -IIb elements are shown in Tables I and II. Table I summarizes the results for the ground-state calculations. Columns 1 and 2 give the element as well as the half-charge radius  $R_C$  and skin-thickness parameter  $t$  in the two-parameter Fermi charge distribution, which was used to describe the extended nucleus. In the remaining columns we present the contributions of the various configurations, in percent, which were taken into account to calculate the ground state. In addition, the electronic charge density at the nucleus (at  $r=0$ ) is shown in the last column.

For the IIa elements the configuration  $ns^2$  contributes between 90% and 93% to the mixing in the wave functions, whereas the  $np^2$  configuration takes over 6–8%. The higher  $(n-1)d^2$  configurations (only possible for the element Ca and heavier) contribute very little. As only the  $s$  and (much less) the  $p_{1/2}$  wave functions contribute to the charge density at  $r=0$  it is easy to understand that this quantity is larger for the single  $ns^2$  configuration calculations than in multiconfiguration calculations where the weight of the  $ns^2$  configuration is reduced. The additional configurations contribute very little or nothing with their  $np$  and  $(n-1)d$  electron wave functions. For the IIb elements the  $np^2$  and  $nd^2$  configurations are not even bound, so we present the single-configuration results only, although they could contribute nonnegligibly in the MCDF procedure. Analogous relativistic calculations have been published for the even-parity state of BaI by Rosén *et al.*,<sup>62</sup> and for nonrelativistic ones by McCavert

and Trefftz.<sup>63</sup> Their results are very comparable to ours.

Table II contains the analogous results for the  $^3P_1$  and  $^1P_1$  states of the IIa and IIb elements. For each state we present up to four rows denoted  $a-d$ . Under  $a$  we give the results of those calculations where only the  $nsnp_{1/2}$  and  $nsnp_{3/2}$  configuration state functions are taken into account. As the calculations are done in a  $jj$  basis the percentage mixing of the  $nsnp_{1/2}$  and  $nsnp_{3/2}$  configurations are given. For pure  $LS$  coupling the mixing of these two states for the  $^3P_1$  state is 66.7% and 33.3%, respectively, and vice versa for the  $^1P_1$  state. In  $b$  we present the results for those MCDF calculations taking into account all five odd configurations with  $J=1$ , which can be constructed from the  $ns$ ,  $np$ , and  $(n-1)d$  single-electron states. We see that  $pd$  states contribute quite considerably, although we notice that their influence is much larger on the  $^1P_1$  states than on the  $^3P_1$  states. For an easy comparison with the experimental analysis in  $d$  we state under  $c$  the weights of the two configurations  $nsnp_{1/2}$  and  $nsnp_{3/2}$  from the calculation  $b$ , renormalized, however, to 100%.

Analysis of the mixing between different states is performed normally by a least-squares fit of parametrized energy expressions, including electrostatic and magnetic interactions to the experimental energy levels as, for example, described by Condon and Shortley.<sup>19</sup> Usually, this type of analysis is performed in terms of pure  $LS$  states with total  $J$  as the good quantum number. The configuration-interaction contribution and the breakdown of  $LS$  coupling for heavier elements will then appear in the change of the mixing parameters as compared with the results for pure  $LS$  states. The intermediate wave functions are in this way expressed as linear combination  $|SLJ\rangle$  states within the investigated configuration.

In our case—the coupling of  $^1P_1$  and  $^3P_1$  states—one may write

TABLE I. DF and MCDF results for the  $^1S_0$  ground state of the IIa and IIb elements and Yb.

Element	Nuclear parameters		Electron state $ns^2 SLJ\rangle$	Electron $ns^2$	Configuration contributions (%)				$4\pi \Psi(0) _{\text{tot}}^2$ [(a.u.) <sup>-3</sup> ]
	$R_c$ (fm)	$t$ (fm)			$np_{1/2}^2$	$np_{3/2}^2$	$(n-1)d_{3/2}^2$	$(n-1)d_{5/2}^2$	
$^4\text{Be}$	1.97	2.34	$2s^2^1S_0$	100					447.8675
				90.3	3.2	6.5			447.1048
$^{12}\text{Mg}$	2.85	2.6	$3s^2^1S_0$	100					14 541.152
				92.5	2.5	5.0			14 539.628
$^{20}\text{Ca}$	3.6	2.51	$4s^2^1S_0$	100					77 301.282
				91.8	2.7	5.2	0.1	0.2	77 297.680
$^{38}\text{Sr}$	4.8	2.3	$5s^2^1S_0$	100					765 147.49
				92.2	2.6	4.7	0.2	0.3	765 138.27
$^{56}\text{Ba}$	5.83	1.79	$6s^2^1S_0$	100					4 052 630.1
				91.8	2.8	4.4	0.3	0.6	4 052 608.5
$^{88}\text{Ra}$	6.7	2.3	$7s^2^1S_0$	100					56 397 338
				93.3	2.8	2.8	0.3	0.7	56 397 202
$^{30}\text{Zn}$	4.24	2.3	$4s^2^1S_0$	100					315 037.27
$^{48}\text{Cd}$	5.24	2.3	$5s^2^1S_0$	100					2 006 489.1
$^{80}\text{Hg}$	6.44	2.3	$6s^2^1S_0$	100					29 341 769
$^{70}\text{Yb}$	6.37	2.3	$6s^2^1S_0$	100					12 866 018
				92.8	2.9	4.2			12 865 956

TABLE II. Results for  $nsnp\ ^3P_1$  and  $nsnp\ ^1P_1$  states of IIa and IIb elements. (a) MCDF calculation with two configurations, (b) MCDF calculation with five configurations, (c) normalization of the mixing contribution of (b) to only mixing of  $nsnp_{1/2}$  and  $nsnp_{3/2}$  configurations, (d) mixing of  $nsnp_{1/2}$  and  $nsnp_{3/2}$  configurations as derived from the experimental energy-level scheme.

Element	Electron state		Configurations contributions (%)					$4\pi  \Psi(0) _{\text{tot}}^2$ (a.u.) <sup>-3</sup>	
	$nsnp$	$ SLJ\rangle$	$ns$ + $np_{1/2}$	$ns$ + $np_{3/2}$	$np_{1/2}$ + $(n-1)d_{3/2}$	$np_{3/2}$ + $(n-1)d_{3/2}$	$np_{3/2}$ + $(n-1)d_{5/2}$		
<sup>4</sup> Be	$2s\ 2p$	$^3P_1$	a	66.6	33.4				441.086
			d	67.04	32.96				
		$^1P_1$	a	33.6	66.4				443.25
			d	32.96	67.04				
<sup>12</sup> Mg	$3s\ 3p$	$^3P_1$	a	66.8	33.2				14 525.549
			d	67.03	32.97				
		$^1P_1$	a	33.2	66.8				14 531.822
			b	31.7	63.6	1.6	0.3	2.9	14 528.561
			c	33.3	66.7	(npnd configurations)			
			d	32.97	67.03				
<sup>20</sup> Ca	$4s\ 4p$	$^3P_1$	a	67.0	33.0				77 279.146
			b	65.7	31.3	0.5	1.7	0.9	77 277.843
			c	67.7	32.3				
			d	67.4	32.6				
		$^1P_1$	a	33.0	67.0				77 288.244
			b	26.6	54.7	6.3	1.2	11.2	77 277.332
			c	32.7	67.3				
			d	32.6	67.4				
<sup>38</sup> Sr	$5s\ 5p$	$^3P_1$	a	67.8	32.2				765 091.35
			b	67.0	29.5	0.7	1.9	0.8	765 087.65
			c	69.4	30.6				
			d	69.5	30.5				
		$^1P_1$	a	32.1	67.9				765 114.34
			b	24.5	55.2	7.0	1.3	12.0	765 086.32
			c	30.7	69.3				
			d	30.5	69.5				
<sup>56</sup> Ba	$6s\ 6p$	$^3P_1$	a	69.2	30.8				4 052 503.9
			b	69.1	26.0	1.3	2.8	0.9	4 052 492.4
			c	72.7	27.3				
			d	75.0	25.0				
		$^1P_1$	a	30.6	69.4				4 052 556.1
			b	17.7	51.8	11.5	1.7	17.4	4 052 471.9
			c	25.5	74.5				
			d	25.0	75.0				
<sup>88</sup> Ra	$7s\ 7p$	$^3P_1$	a	74.1	25.9				56 396 456
			b	77.3	18.8	1.6	2.0	0.2	56 396 397
			c	80.4	19.6				
			d	84.5	15.5				
		$^1P_1$	a	25.3	74.7				56 396 782
			b	13.4	67.1	8.6	0.9	10.0	56 396 386
			c	16.6	83.4				
			d	15.5	84.5				
<sup>30</sup> Zn	$4s\ 4p$	$^3P_1$	a	67.5	32.5				314 953.23
			d	68.0	32.0				
		$^1P_1$	a	32.6	67.4				314 975.61
			d	32.0	68.0				
<sup>48</sup> Cd	$5s\ 5p$	$^3P_1$	a	69.1	30.9				2 006 267.7
			b	71.0	29.0				
		$^1P_1$	a	30.9	69.1				2 006 321.9
			d	29.0	71.0				
<sup>80</sup> Hg	$6s\ 6p$	$^3P_1$	a	75.7	24.3				29 340 002
			d	80.7	19.3				
			a	24.2	75.8				
			d	19.3	80.7				

TABLE II. (Continued).

Element	Electron state		Configurations contributions (%)					$4\pi  \Psi(0) _{\text{tot}}^2$ (a.u.) <sup>-3</sup>		
	<i>nsnp</i>	$ SLJ\rangle$	<i>ns</i> $+np_{1/2}$	<i>ns</i> $+np_{3/2}$	<i>np</i> <sub>1/2</sub> $+(n-1)d_{3/2}$	<i>np</i> <sub>3/2</sub> $+(n-1)d_{3/2}$	<i>np</i> <sub>3/2</sub> $+(n-1)d_{5/2}$			
<sup>70</sup> Yb	6s6p	<sup>3</sup> P <sub>1</sub>	a	71.0	29.0				12 865 675	
			b	72.5	24.9	0.8	1.5	0.4	12 865 657	
			c	74.4	25.6					
			d	77.6	22.4					
		<sup>1</sup> P <sub>1</sub>	a	28.7	71.3				12 865 800	
			b	21.3	65.0	5.3	0.8	7.7	12 865 679	
			c	24.7	75.3					
			d	22.4	77.6					

$$\begin{aligned}
 |^3P_1\rangle' &= (1-\gamma^2)^{1/2} |^3P_1\rangle + \gamma |^1P_1\rangle, \\
 |^1P_1\rangle' &= -\gamma |^3P_1\rangle + (1-\gamma^2)^{1/2} |^1P_1\rangle,
 \end{aligned}
 \tag{19}$$

where the states to the right are pure *LS* states. This evaluation procedure normally includes the electrostatic and spin-orbit interaction, although recently some refined analysis has been performed with different spin-orbit parameters for the diagonal and off-diagonal matrix elements as well as the inclusion of the spin-spin interaction.<sup>64,65</sup> The parameters evaluated by the least-squares fit should be considered as *effective* ones within the analyzed *nsnp* configuration.

With the experimental energy levels given by Moore<sup>55</sup> and the formulas by Olsson and Salomonson,<sup>64</sup> one obtains the mixing parameter values given in row *d* for the <sup>3</sup>P<sub>1</sub> and <sup>1</sup>P<sub>1</sub> states in Table II. As a comparison we transformed these *LS*-coupled wave functions into *jj*-coupled ones. The values for  $\gamma$  [according to Eq. (19)] range from  $-0.004$  for <sup>4</sup>Be and <sup>12</sup>Mg up to  $-0.008$  for <sup>20</sup>Ca,  $-0.014$  for <sup>30</sup>Zn,  $-0.031$  for <sup>38</sup>Sr,  $-0.047$  for <sup>48</sup>Cd,  $-0.092$  for <sup>50</sup>Ba,  $-0.122$  for <sup>70</sup>Yb,  $-0.16$  for <sup>80</sup>Hg, and  $-0.21$  for <sup>88</sup>Ra. It can be seen that in this semiempirical analysis the mixing is rather close to the pure *LS* coupling for elements up to Sr, while a significant breakdown of *LS* coupling takes place in the heavier elements like <sup>56</sup>Ba, <sup>88</sup>Ra, <sup>80</sup>Hg, and <sup>70</sup>Yb.

A comparison of the mixing obtained from the analysis in row *d* with the renormalized *ab initio* values in row *c* shows that the inclusion of the mixing with the *np(n-1)d* configuration in the calculations leads to a much better agreement than what is achieved with the calculations in row *a*. This semiempirical analysis was done only within the *nsnp* <sup>3</sup>P<sub>1</sub> and <sup>1</sup>P<sub>1</sub>  $|SLJ\rangle$  states. A better approach would be a new analysis with the inclusion of the configurations used in row *b*.

Concluding, one may say that the agreement of the MCDF calculations with the semiempirical analysis is very promising.

#### B. Electronic charge density at the nucleus

The single-configuration DF and MCDF results presented in Tables I and II also include the electronic charge density at the nucleus (at  $r=0$ ). The last columns of these tables give the value  $a_0 = 4\pi |\Psi(0)|_{\text{tot}}^2$  for the

<sup>1</sup>S<sub>0</sub>, <sup>3</sup>P<sub>1</sub>, and <sup>1</sup>P<sub>1</sub> states of the neutral systems.  $a_0$  is the first of the expansion coefficients of the charge density defined in Eq. (5).

As mentioned in the Introduction, the analysis of optical IS for the neutral systems is performed normally with reference to the ionized atom. We have, therefore, done single-configuration HF and DF calculations for the ground state *ns*<sup>2</sup>S<sub>1/2</sub> and first excited *np*<sup>2</sup>P<sub>1/2</sub> and *np*<sup>2</sup>P<sub>3/2</sub> states in the ions as well. The resulting total charge densities at the nucleus (at  $r=0$ ) are presented in Table III, columns 3 and 4, with the same nuclear parameters as for the neutral systems. The comparison of the nonrelativistic and relativistic results shows the dramatic increase of the charge density at the nucleus for higher *Z*, which usually is called the direct relativistic effect, and which is for Ra, e.g., already a factor of 9.3.

In an *ab initio* evaluation of the electronic part of the volume isotope shift it is not sufficient just to give the difference of the electronic charge density at  $r=0$ . The influence of the variation of the electronic charge over the nuclear volume must be discussed as well. As shown in formulas (6) and (7), this leads to additional contributions to the isotope shifts which are dependent on the change of the higher moments of the nuclear charge distribution as well as on the change of the expansion coefficients of the electronic charge distribution within the two configurations. The correction factor *K*, defined in Eq. (7), summarizes all these contributions. As nearly 90% of the charge density at the nucleus originates from the two 1s electrons, and the main part of the remaining 10% comes from the other *s* electrons, the change of the electron charge density has an *s*-wave-function behavior and is almost independent of the specific configuration. The coefficients  $C_2/C_1$  and  $C_3/C_1$  [defined in Eqs. (6) and (7), which were first introduced by Seltzer<sup>42</sup>] were calculated by us for many elements and transitions. Table IV presents these values together with those of Ref. 42, which include the contribution from only the 1s<sup>2</sup> electrons. In Fig. 2 a visual presentation of these coefficients is given as a function of the atomic number *Z*. The coefficients  $C_2/C_1$  appear to be very linear, while  $C_3/C_1$  show a somewhat structured behavior.

If the values for  $\Delta\langle r^4 \rangle^{A_1 A_2}$ ,  $\Delta\langle r^6 \rangle^{A_1 A_2}$ , etc., in Eq. (7) were known, the total correction factor *K* would be known as well. There is the possibility of getting realistic



TABLE III. Dirac-Fock results for the singly ionized ions of the group-IIa and -IIb elements. In addition, semiempirical charge densities as well as a comparison of relativistic and nonrelativistic parameters for the volume isotope shift and the magnetic dipole interaction are given.

Ion	Config. state	$4\pi  \Psi(0) ^2_{\text{tot}} \text{ (a.u.)}^{-3}$		$4\pi  \Psi(0) ^2 \text{ (a.u.)}^{-3}$		$F(ns_{1/2})(1-\delta)$		$R_{\text{tot}}$	$R_{\text{ns}}$	$\beta$					
		Nonrel.	Rel.	<i>ab initio</i>		<i>ab initio</i>		<i>ab initio</i>		Eq. (22)	Nonrel. Rel.				
				Eq. (18)	Eq. (17)	Eq. (17)	Hydrogenic <sup>b</sup>	Eq. (20)	Eq. (21)		Eq. (12)				
$^4\text{Be}^+$	$2s^2S_{1/2}$	440.612	443.907	10.155	10.235	10.170	12.5	12.5 <sup>c</sup>	1.001	1.002	1.007	1.008	1.008	1.042	1.043
	$2p^2P_{1/2}$		433.231								1.007				
$^{12}\text{Mg}^+$	$2p^2P_{3/2}$	430.019	433.214												
	$3s^2S_{1/2}$	13 716.19	14 531.58	21.341	22.735	21.677	28.7	27.0 <sup>d</sup>	1.02	1.01	1.059	1.065	1.064	1.097	1.097
	$3p^2P_{1/2}$	13 692.77	14 506.64								1.059				
$^{20}\text{Ca}^+$	$3p^2P_{3/2}$		14 506.64												
	$4s^2S_{1/2}$	66 669.02	77 287.49	24.186	28.532	25.318	33.9		1.05	1.03	1.159	1.180	1.170	1.116	1.114
	$4p^2P_{1/2}$	66 642.02	77 255.70								1.159				
$^{38}\text{Sr}^+$	$4p^2P_{3/2}$		77 255.67												
	$5s^2S_{1/2}$	475 265.14	765 111.50	41.220	71.298	49.256	55.5	56.8 <sup>e</sup>	1.20	1.14	1.610	1.730	1.657	1.110	1.101
	$5p^2P_{1/2}$	475 219.38	765 033.02								1.610				
$^{56}\text{Ba}^+$	$5p^2P_{3/2}$		765 032.52												
	$6s^2S_{1/2}$	1 546 317.7	4 052 548.3	50.861	157.695	77.007	77.0	74.4 <sup>d</sup>	1.51	1.37	2.621	3.100	2.780	1.111	1.087
	$6p^2P_{1/2}$	1 546 261.2	4 052 376.9								2.621				
$^{88}\text{Ra}^+$	$6p^2P_{3/2}$		4 052 373.8												
	$7s^2S_{1/2}$	6 058 540.6	56 396 722	74.561	1102.00	246.96	124.7		3.31	2.35	9.309	14.780	10.837	1.106	1.039
	$7p^2P_{1/2}$	6 058 458.1	56 395 577								9.309				
$^{70}\text{Zn}^+$	$7p^2P_{3/2}$		56 395 504												
	$4s^2S_{1/2}$	230 816.17	314 972.92	75.286	109.209	85.398	98.3		1.13	1.08	1.365	1.451	1.389	1.099	1.095
	$4p^2P_{1/2}$		314 853.37								1.365				
$^{48}\text{Cd}^+$	$4p^2P_{3/2}$	230 733.43	314 852.97												
	$5s^2S_{1/2}$	967 895.25	2 006 313.4	111.281	274.330	157.720	147.6	158.3 <sup>g</sup>	1.42	1.25	2.073	2.465	2.166	1.096	1.082
	$5p^2P_{1/2}$	967 773.27	2 006 016.5								2.073				
$^{80}\text{Hg}^+$	$5p^2P_{3/2}$		2 006 013.0												
	$6s^2S_{1/2}$	4 545 278.9	29 340 140	189.338	2079.96	572.74	303.0 <sup>h</sup>	329.0 <sup>i</sup>	3.03	1.99	6.455	10.985	7.290	1.088	1.038
	$6p^2P_{1/2}$	4 545 072.8	29 337 980								6.455				
$^{70}\text{Yb}^+$	$6p^2P_{3/2}$		29 337 879												
	$6s^2S_{1/2}$	3 035 943.8	12 865 772	79.59	449.416	160.30	128.1	123.8 <sup>j</sup>	2.01	1.66	4.238	5.647	4.693	1.099	1.050
	$6p^2P_{1/2}$	3 035 856.3	12 865 300								4.238				

<sup>a</sup>Reference 66 (nuclear moments and spins).

<sup>b</sup>According to Eqs. (26.18) and Fig. 71 in Ref. 1.

<sup>c</sup>Reference 67.

<sup>d</sup>Reference 68.

<sup>e</sup>Reference 69.

<sup>f</sup>Reference 70.

<sup>g</sup>Reference 71.

<sup>h</sup>Reference 72.

TABLE IV. Electronic coefficients in the parameter  $\lambda$  in Eq. (7) from MCDF calculations and Ref. 44.

Element	Source	$C_2/C_1$ ( $10^{-4} \text{ fm}^{-2}$ )	$C_3/C_1$ ( $10^{-6} \text{ fm}^{-4}$ )	$K$ [Eq. (9)]
${}^4\text{Be}$	Ref. 42			
	MCDF	0.26	0.41	0.999 87
${}^{12}\text{Mg}$	Ref. 42			
	MCDF	1.12	1.02	0.998 85
${}^{20}\text{Ca}$	Ref. 42			
	MCDF	2.14	1.37	0.996 4
${}^{30}\text{Zn}$	Ref. 42	3.40	1.39	
	MCDF	3.70	1.87	0.991 5
${}^{38}\text{Sr}$	Ref. 42	4.45	1.56	
	MCDF	4.75	1.97	0.986 0
${}^{48}\text{Cd}$	Ref. 42	5.96	1.88	
	MCDF	6.42	2.37	0.978
${}^{56}\text{Ba}$	Ref. 42	7.03	2.04	
	MCDF	7.47	2.38	0.968
${}^{70}\text{Yb}$	Ref. 42	9.29	2.54	
	MCDF	9.46	2.75	0.952
${}^{80}\text{Hg}$	Ref. 42	10.9	2.90	
	MCDF	12.0	3.66	0.938
${}^{88}\text{Ra}$	Ref. 42	12.1	3.16	
	MCDF	13.5	4.06	0.926

values for  $K$  by use of changes of the nuclear moments from other experiments, like electron scattering.<sup>25</sup> However, this is probably too much of an effort as  $K$  is only a small correction factor. Most likely it is quite sufficient to use the nuclear droplet model with a nuclear radius  $R$  proportional to  $A^{1/3}$ . The resulting equation for  $K$  is

then Eq. (9). The values for  $K$  calculated in this way are given in Table IV. Figure 3 shows the function  $1-K$  as function of  $Z$  which presumably is correct within  $\pm 10\%$  or better.

Having this in mind we are able to calculate the electronic factors  $F_i$  according to the definition of Eq. (8),

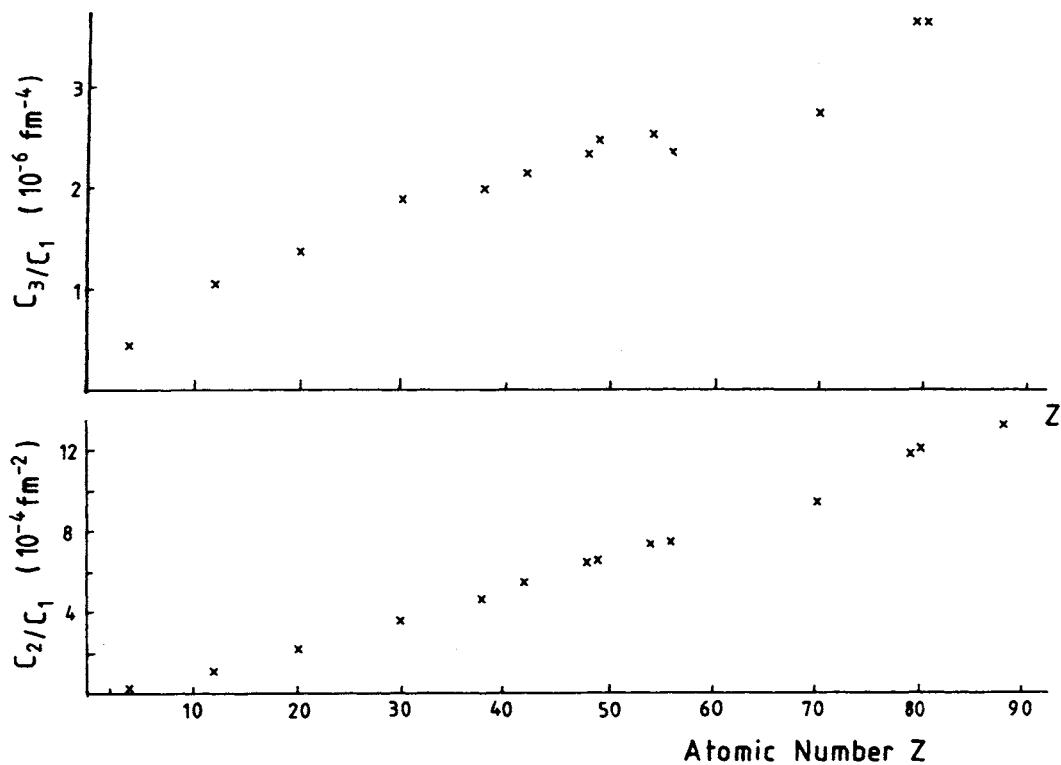


FIG. 2. Theoretical values of the factors  $C_2/C_1$  and  $C_3/C_1$  which describe the change of the electron density over the nuclear radius as function of the atomic number  $Z$ .

where only the change of the electron charge density at  $r=0$  is needed. Before we discuss these results from the *ab initio* calculations in Tables V and VI we will first analyze in Table III the semiempirical calculation of the charge density  $|\Psi_{ns}(0)|^2$  for the ions as well as the various correction factors which are needed in this evaluation.

In order to check and discuss the semiempirical approach reviewed in Sec. IIC, we present in Table III the relativistic charge densities  $4\pi|\Psi_{ns}(0)|^2$  for the outermost  $ns$  electron as well, using our *ab initio*  $ns$  wave functions. The first method is the evaluation of the value according to Eq. (18) in column 6, and the second is the magnetic dipole interaction integral according to Eq. (17) in column 7. In addition, analogous nonrelativistic calculations were done, which we present in column 5. We notice that the magnetic dipole integral and the relativistic  $ns$  charge density at  $r=0$  are almost the same for elements up to  $\text{Ca}^+$ . For heavier elements like  $\text{Hg}^+$  and  $\text{Ra}^+$  the volume isotope-shift values are bigger than the magnetic ones by approximately a factor of up to 5. We present the *ab initio* values of  $|\Psi_{ns}(0)|^2$  in Table III only to give a comparison with the semiempirical approximation as we use only the results of  $\Delta|\Psi(0)|_{\text{tot}}^2$  to calculate the  $F_i$  factors.

As reviewed in Sec. IIC, essentially two types of semiempirical approaches are used to calculate the electronic charge density at the nucleus. The most straightforward one is to estimate  $|\Psi_{ns}(0)|_{\text{nr}}^2$  from the atomic energy levels for the alkali-metal-like systems according to the Goudsmit-Fermi-Segré formula [Eq. (14)]. In the calculation the quantity  $(1-d\sigma/dn)$  is evaluated from the energy levels given by Moore.<sup>55</sup> The resulting values are given in the eighth column. It was found that the value of  $(1-d\sigma/dn)$  for  $\text{Hg}^+$  of 1.248 given by Kopfermann<sup>1</sup> is not correct, it should be 1.17 instead.

The other approach is to use the experimental magnetic dipole interaction constants  $a_{ns}^{\text{expt}}$  for the ground state of the  $^2S_{1/2}$  state of the ion according to Eq. (15). As the Bohr-Weisskopf correction factor  $(1-\delta)$  and the Breit-Rosenthal correction  $(1-\epsilon)$  are taken from Kopfermann<sup>1</sup> we have to use the hydrogenic factor  $F(ns_{1/2})$  discussed below to be consistent.

Unfortunately the experimental magnetic dipole interaction constants are only known for some isotopes of the elements (quoted in Table III) and a complete comparison is therefore not possible. The resulting values according to this second semiempirical procedure are given in column 9.

It is astonishing that the semiempirical values of  $|\Psi_{ns}(0)|_{\text{nr}}^2$  in columns 8 and 9 are so similar while they are evaluated in such different ways. The difference to the *ab initio* nonrelativistic values in column 5 makes clear that there are contributions beyond Hartree-Fock. As these semiempirical values contain experimental information they include part of higher polarization and correlation effects which for the ionic transitions are not contained in our *ab initio* values. On the other hand, the semiempirical values also include theoretical values like, e.g.,  $F(ns_{1/2})$ , from rather simple calculations, whereas the *ab initio* calculations do not contain any correction factors [like, e.g.,  $(1-\delta)$ ] as they are automatically included. The semiempirical evaluation of  $F_i$  in addition needs the value for  $\beta$  in Eq. (12) and  $f(Z)$  in Eq. (11).

In addition, in order to give a complete comparison with the semiempirical approach, we present in Table III values of the various correction factors like  $F(ns_{1/2})$ ,  $R$ , and  $\beta$  which we do not need in our *ab initio* evaluation of the electronic factors.

The correction factors  $F(ns_{1/2})(1-\delta)$ , as defined in Eq. (17), are given in column 10 from DF results in columns 5 and 7, whereas column 11 presents analogous values calculated with hydrogenic wave functions according to Kopfermann.<sup>1</sup>

In columns 12 and 13 we present the relativistic correction factors  $R$  from our *ab initio* calculation which are defined as follows:

$$R_{\text{tot}} = \frac{|\Psi(0)|_{\text{tot,rel}}^2}{|\Psi(0)|_{\text{tot,nr}}^2} \quad (20)$$

and

$$R_{ns} = \frac{|\Psi_{ns}(0)|_{\text{rel}}^2}{|\Psi_{ns}(0)|_{\text{nr}}^2} \quad (21)$$

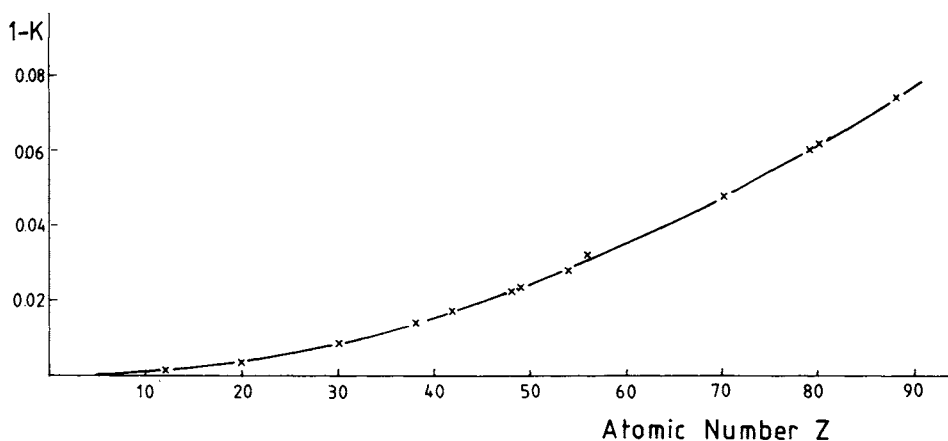


FIG. 3. The correction factor  $K$  as defined in Eq. (9), presented here as  $1-K$ .

TABLE V. A summary of *ab initio* charge density differences and  $F_i$  values for transitions in neutral atoms and singly ionized group-IIa elements. In addition, semiempirical and experimental  $F_i$  values are presented.

Spectra	Transition	Wave-length (nm)	$4\pi\Delta  \Psi(0) ^2$ (a.u.) <sup>-3</sup>			$F_i$ (GHz/fm <sup>2</sup> )			Expt. Muonic x rays
			Nonrel. HF	Rel. DF	Rel. MCDF	Rel. DF	Rel. MCDF	Semiempirical GFS Eq. (14)	
Be II	$2s^2S_{1/2} \rightarrow 2p^2P_{1/2}$	313.2	10.593	10.676		-0.01672			
	$\rightarrow 2p^2P_{3/2}$	313.1	10.593	10.693		-0.01675			
Be I	$2s^2^1S_0 \rightarrow 2s2p^1P_1$	235		4.618	3.855	-0.00723	-0.00604		
	$\rightarrow 2s2p^3P_1$	455		6.782	6.019	-0.0106	-0.00943		
Mg II	$3s^2S_{1/2} \rightarrow 3p^2P_{1/2}$	280.4	23.42	24.94		-0.1172			
	$\rightarrow 3p^2P_{3/2}$	279.6	23.42	24.94		-0.1172			
Mg I	$3s^2^1S_0 \rightarrow 3s3p^1P_1$	285		9.33	11.067	-0.0438	-0.05201		
	$\rightarrow 3s3p^3P_1$	457		15.60	14.079	-0.0733	-0.06616		
Ca II	$4s^2S_{1/2} \rightarrow 4p^2P_{1/2}$	397.0	27.0	31.79		-0.2490			
	$\rightarrow 4p^2P_{3/2}$	393.5	27.0	31.82		-0.2492			
Ca I	$4s^2^1S_0 \rightarrow 4s4p^1P_1$	423.7		13.038	20.348	-0.1021	-0.1594		-0.176(10) <sup>a</sup>
	$\rightarrow 4s4p^3P_1$	657.3		22.136	19.837	-0.1734	-0.1554		-0.182(5) <sup>b</sup>
Sr II	$5s^2S_{1/2} \rightarrow 5p^2P_{1/2}$	421.6	45.76	78.48		-1.168		-1.49	
	$\rightarrow 5p^2P_{3/2}$	407.8	45.76	78.90		-1.174			
Sr I	$5s^2^1S_0 \rightarrow 5s5p^1P_1$	460.7		33.15	51.95	-0.4933	-0.7731		
	$\rightarrow 5s5p^3P_1$	689.4		56.14	50.62	-0.8354	-0.7533		
Ba II	$6s^2S_{1/2} \rightarrow 6p^2P_{1/2}$	493.4	56.5	171.4		-3.759		-4.98 <sup>c</sup>	-4.81
	$\rightarrow 6p^2P_{3/2}$	455.5	56.5	174.5		-3.827			
Ba I	$6s^2^1S_0 \rightarrow 6s6p^1P_1$	553.5		74.0	136.6	-1.623	-2.996	-3.93 <sup>e</sup>	
	$\rightarrow 6s6p^3P_1$	791.1		126.2	116.1	-2.768	-2.546		-3.80(33) <sup>d</sup>
Ra II	$7s^2S_{1/2} \rightarrow 7p^2P_{1/2}$	463.4	82.5	1145		-39.460			
	$\rightarrow 7p^2P_{3/2}$	381.6	82.5	1218		-41.976			
Ra I	$7s^2^1S_0 \rightarrow 7s7p^1P_1$	482.7		556	816.0	-19.160	-28.122		-3.04(26) <sup>f</sup>
	$\rightarrow 7s7p^3P_1$	714.3		882	805.0	-30.394	-27.743		-2.59(22) <sup>d</sup>

<sup>a</sup>Reference 74.

<sup>b</sup>Reference 75.

<sup>c</sup>Reference 76.

<sup>d</sup>Reference 26.

<sup>e</sup>Reference 77.

<sup>f</sup>Reference 78.

TABLE VI. A summary of *ab initio* charge density differences and  $F_i$  values for transitions in neutral atoms and singly ionized group-IIb elements. In addition, semiempirical and experimental  $F_i$  values are presented.

Spectra	Transition	Wave-length (nm)	$4\pi\Delta  \Psi(0) ^2$ (a.u.) <sup>-3</sup>			$F_i$ (GHz/fm <sup>2</sup> )				Experimental	
			<i>ab initio</i>			<i>ab initio</i>		Semiempirical		Electronic x rays	Muonic x rays
			Nonrel. HF	Rel. DF	Rel. MCDF	Rel. DF	Rel. MCDF	GFS Eq. (14)	hfs		
Zn II	$4s^2S_{1/2} \rightarrow 4p^2P_{1/2}$	206.3	82.74	119.55		-1.405					
	$\rightarrow 4p^2P_{3/2}$	203.6	82.74	119.95		-1.409					
Zn I	$4s^2^4S_0 \rightarrow 4s4p^1P_1$	213.9		61.66		-0.724					
	$\rightarrow 4s4p^3P_1$	307.7		84.04		-0.987					
Cd II	$5s^2S_{1/2} \rightarrow 5p^2P_{1/2}$	226.5	121.98	296.9		-5.581		-6.39	-6.67		
	$\rightarrow 5p^2P_{3/2}$	214.4	121.98	300.4		-5.647					
Cd I	$5s^2^1S_0 \rightarrow 5s5p^1P_1$	228.8		167.2		-3.143					
	$\rightarrow 5s5p^3P_1$	326.1		221.4		-4.162					
Hg II	$6s^2S_{1/2} \rightarrow 6p^2P_{1/2}$	194.2	206.1	2160		-67.67		-67.6	-73.4		
	$\rightarrow 6p^2P_{3/2}$	165.0	206.1	2261		-70.84					
Hg I	$6s^2^1S_0 \rightarrow 6s6p^1P_1$	184.9		1467		-45.96					
	$\rightarrow 6s6p^3P_1$	253.7		1767		-55.36		-44.9 <sup>a</sup> -48.8 <sup>c</sup>		-53.9(5.3) <sup>a</sup>	-57.4(2.9) <sup>b</sup>
Yb II	$6s^2S_{1/2} \rightarrow 6p^2P_{1/2}$		87.5	472		-12.94		-16.8	-16.2		
	$\rightarrow 6p^2P_{3/2}$		87.5	488		-13.38					
Yb I	$6s^2^1S_0 \rightarrow 6s6p^1P_1$	398.8		218	277	-5.98	-7.59				
	$\rightarrow 6s6p^3P_1$	555.65		343	299	-9.40	-8.20	-11.4 <sup>d</sup>	-10.9 <sup>d</sup>		

<sup>a</sup>Reference 72.

<sup>b</sup>Reference 25.

<sup>c</sup>Reference 79.

<sup>d</sup>Reference 80.

In the literature there exists another factor  $R_{ns}^{\text{hyd}}$  which is obtained with the hydrogenic wave functions<sup>81</sup> presented in column 14. As the relativistic  $ns$  wave function for a point nucleus diverges as  $r=0$ , the value at the nuclear radius  $r_N$  is used—thus it is defined at

$$R_{ns}^{\text{hyd}} = \frac{|\Psi_{ns}^{\text{hyd}}(r_N)|_{\text{rel}}^2}{|\Psi_{ns}^{\text{hyd}}(0)|_{\text{nr}}^2} \quad (22)$$

with  $r_N = 1.2 A^{1/3}$  fm. The comparison between the three columns shows that there are quite significant differences, especially for the high- $Z$  elements. The quantities  $R_{\text{tot}}$  and  $R_{ns}^{\text{hyd}}$  are relatively close to each other. There are two reasons for this. First, the choice to use the charge density at the distance  $r_N$  from the point-nucleus calculations is a good approximation for the total charge density at  $r=0$  from an extended-nucleus calculation. Second, the quantity  $R_{ns}^{\text{hyd}}$ , as defined in Eq. (22), is independent of  $n$  and as the total charge density is determined by the  $1s$  contribution—which in every type of calculation also is hydrogeniclike—both quantities are very comparable.

In columns 15 and 16 in Table III we present the screening constant  $\beta$ , defined in Eq. (12), as calculated from our *ab initio* HF and DF calculations, respectively. Similar calculations have been carried out earlier by Wilson<sup>82,83</sup> and Rajnak and Fred<sup>84</sup> which are in rather good agreement with our nonrelativistic results. A small difference exists between the relativistic and nonrelativistic values. The relativistic value is smaller because the relativistic  $ns$  wave function has a much higher probability in the vicinity of the nucleus and thus is screened less by the other electrons.

### C. Electronic factors in volume isotope shifts

Finally, the electronic factors  $F_i$  for the resonance transitions of ions, and neutral atoms of the *IIa* and *IIb* elements are presented in Tables V and VI, respectively. The experimental wavelengths for these transitions are given in column 3, the changes of the charge densities  $4\pi\Delta|\Psi(0)|^2$  are presented in columns 4–6. They have been calculated from the total charge densities at the nucleus (at  $r=0$ ) for the particular states with our *ab initio* HF, DF, and MCDF methods.

The comparison of nonrelativistic and relativistic single-configuration calculations of  $\Delta|\Psi(0)|^2$  shows an even larger increase of the charge densities at the nucleus for higher  $Z$  compared to the increase of the total charge density; for Ra it is a factor of 13 already. The electronic factors  $F_i$  from the relativistic DF and MCDF results calculated according to Eq. (8) are presented in columns 7 and 8. The MCDF calculations, which include the configurations discussed in Tables I and II, change the  $F_i$  factors quite drastically for all elements presented here. The difference between the  $F_i$  values for the  $^1S_0 \rightarrow ^1P_1$  and  $^1S_0 \rightarrow ^3P_1$  transitions, which can be seen for the DF results in column 7, becomes much smaller and is even inverted in the MCDF calculations in column 8. The reason for this behavior is the strong contribution of the  $np(n-1)d$  configuration in the  $^1P_1$  state which can be seen in Table II and which is not present very strongly in

the MCDF calculations of the  $^3P_1$  level.

These *ab initio* results of the  $F_i$  values will now be compared with the semiempirical estimates. To do this the semiempirical values  $|\Psi_{ns}(0)|_{\text{nr}}^2$  obtained in Table III, columns 8 and 9, have to be introduced in Eq. (12) and then in Eq. (11) to calculate the  $F_i$  factors. This means that the values for  $\beta$  and  $f(Z)$  have to be known as well. To be consistent we use the nonrelativistic  $\beta$  values in Table III, column 15, and  $f(Z)$  is taken from Babushkin.<sup>46</sup> The resulting  $F_i$  values for the transitions discussed are presented in columns 9 and 10 of Tables V and VI. We present in these tables only a few semiempirical  $F_i$  values because Babushkin has given his  $f(Z)$  values for only a small number of elements. Of course, one could use the values  $R_{ns}^{\text{hyd}}$  from Eq. (22) instead. To be consistent we leave it to the reader to calculate the  $F_i$  values in this different way from the values given in Table III. The  $F_i$  values of transitions in the neutral systems are usually connected to the  $F_i$  values of the ions by slopes in King plots. If such experimental data are not available the  $F_i$  values of the neutral systems can be connected to the  $F_i$  values of the ionized systems by use of Eq. (13).

As an overall trend it can be seen that the semiempirical  $F_i$  values are larger in nearly all cases than the *ab initio* ones. In most cases the difference amounts to about 30%. The only exception from this trend is Hg. It is impossible to comment these findings in a physical way. The *ab initio* calculation is a straightforward consistent method only dependent on the quality of the MCDF method to describe the charge density at the nucleus in two different fully self-consistent calculations. On the other hand, the semiempirical calculations, which we have discussed in detail, are a mixture of experimental, semiempirical, and partially simple theoretical results. It is astonishing that these semiempirical methods are at all good enough that the agreement with a purely theoretical method is within 30% or better. Of course, we do not want to claim that the MCDF method leads to exact results, but a careful analysis of the configurations used in the multiconfiguration calculations allows us to claim that most of the relevant configurations are taken into account. A bit of evidence that this statement may be correct comes from the comparison with the results of columns 12 and 13 where we present the experimental values of the electronic factors  $F_i$  derived from a combination of optical isotope-shift data and  $\delta\langle r^2 \rangle^{AA'}$  obtained in electronic and muonic x rays. The few values which are available show better agreement with our *ab initio* results than with the semiempirical results.

## IV. CONCLUSIONS AND OUTLOOK

Although *IIa* and *IIb* elements in first approximation are relatively simple two-electron systems, they are still not understood in great detail; the theoretical description is not yet very satisfying. On the other hand, from the experimental point of view we have a very good knowledge of the various different quantities like atomic energy levels, fine-structure splittings, hyperfine-structure constants, lifetimes, etc., for low-lying states as well as different

Rydberg series.

In the analysis of the experimental data it is essential to do the analysis on an *ab initio* basis and to try to explain as many experimental quantities as possible with the available theory. Isotope-shift and hyperfine-structure data are, in this respect, of special interest as they are especially sensitive to that part of the wave functions in the vicinity of the nucleus where the inclusion of relativity is especially important. This also means that results from other experimental areas like electronic and muonic x rays can be used as a test for the calculations.

The MCDF method used here is the most general *ab initio* relativistic method nowadays applicable to all elements of the Periodic Table. Of course, e.g., perturbation expansion methods or the random-phase approximation are even more accurate, but can be used only either in certain parts of the periodic system or in certain configurations. The practical problem and the disadvantage of the MCDF method is that additional configurations, which are used together with the main configuration, have to be picked from an infinite possible number of configurations. The hope is that the few configurations which are included in the calculations contribute dominantly.

The calculations presented here show that this procedure is possible more or less as far as the volume isotope shift is concerned. The  $F_i$  values of the resonance transitions of the *IIa* and *IIb* elements, discussed in this paper, are described fairly well, which definitely shows that the multiconfiguration contributions have to be included. Thus part of the correlation as well as the full effect of the extended nucleus and relativity is included fully self-consistently in such calculations. On the other hand, the

semiempirical method used to describe the  $F_i$  values shows that in this method a large number of approximations, experimental values, and semiempirical formulas are mixed together. It is astonishing that so many groups still use these semiempirical approaches which include such a large number of corrections, although it is possible today to use standard theoretical methods like DF or MCDF to obtain *ab initio* values of good accuracy.

With this review for all *IIa* and *IIb* elements we would like to urge the experimentalists to try to reanalyze their IS data in a consistent way with this complete set of  $F_i$  values for all these elements.

As an outlook one may say that the MCDF method is a very valid method to calculate electronic factors with relatively good success for a large number of configurations in the whole region of elements of the Periodic Table. A lot of work is needed to refine and ease this method as there are still practical problems with the picking of the configurations, its inclusion in all general cases, and its numerical convergence.

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