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RELATIVISTIC EFFECTS IN THE HYPERFINE STRUCTURE OF THE SECOND SPECTRUM OF THE Bi II ION*

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The hyperfine structure splitting of the line 2368 Å ($6s^26p^2\,{}^1S_2 \to 6s6p^3\,{}^5S_2^0$) was determined experimentally and used to analyse the ground configuration $6s^26p^2$ of Bi II, in the intermediate coupling scheme. It is experimentally proved that the relativistic effects occurring in the configuration investigated are practically identical to those of the ground configuration $6s^26p^3$ of the Bi I atom and are in agreement with the predictions of the theoretical OHFS and HF procedures. The experimental values obtained for the radial parameters are: $\langle r^{-3}\rangle_{6p}^{01} = 35.4(3)$, $\langle r^{-3}\rangle_{6p}^{12} = 72(1)$, $\langle r^{-3}\rangle_{6p}^{10} = -21(1)$ (all values in a.u.). Configuration interactions (core polarization effect) are shown to participate to forty percent in the parameter $\langle r^{-3}\rangle_{6p}^{10}$.

1. *Introduction*

For the interpretation of the hyperfine structure (hfs), it is necessary to take relativistic effects into account, primarily for heavy elements. Normally, this is done by corrrecting the non-relativistic results by some multiplicative "relativistic correction factors (RCF)".

The simplest approach is to use hydrogenic wavefunctions, and the first of this type is due to Breit [1]. Correction factors based on this kind of calculation have also been analyzed by Casimir [2] and have ever since been used almost exclusively in hyperfine-structure analysis. A convenient tabulation is due to Kopfermann [3]. A significant improvement in this theory was made by Schwartz [4] who, for the p-electrons, calculated the ratio C''/C' of normalization factors versus the atomic effective numbers.

Recently, new methods of relativistic calculation of the hyperfine structure have been developed, mainly by Lindgren and Rosén [5]. They performed an *ab initio* theoretical calculation of the radial parameters of the hyperfine structure for a variety of elements. For the heavy elements, it is very interesting to compare their results, obtained by the use

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of restricted relativistic Hartree–Fock wave functions (HF method) or by the "optimized Hartree–Fock–Slater" (OHFS) method with the experimental data.

In this work we performed a hyperfine structure analysis of the $6s^2 6p^2$ ground configuration of the Bi II ion in order to find which of the above mentioned theoretical methods is the best. For this purpose, the hyperfine structure of the level $^1D_2$ on the line $\lambda = 2368$ Å was investigated experimentally. Unfortunately, the levels $^3P_{1,2}$ were not accessible by our equipment and the experimental data for these levels were taken from the work of Cole [6]. A scheme for the energy levels of Bi II is given in Fig. 1.

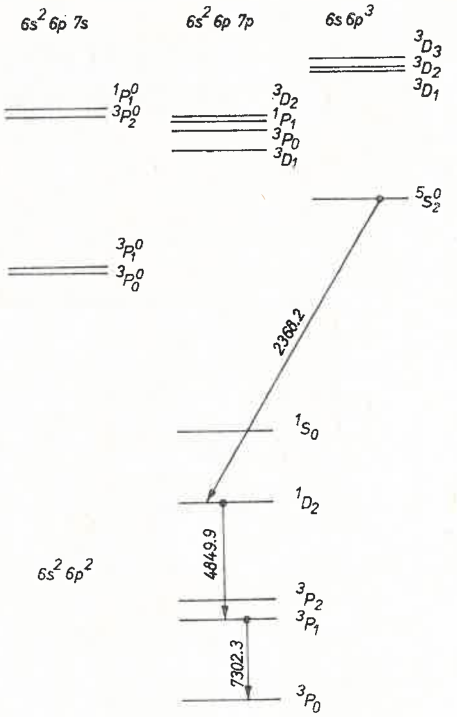

Fig. 1. Diagram of the electronic levels of the Bi II ion

2. Experimental results

The hyperfine structure of the Bi II spectrum obtained with a hollow cathode was investigated on the 2368 Å line with a PGS-2 plane grating spectrograph (1300 grooves/mm). In the sixth order, where we worked, the reciprocal dispersion at the position of the strongest component was 0.233697 (18) Ångstroms per millimeter. The linear change in reciprocal dispersion within the hyperfine structure multiplet was determined as 276.49(65) $\times 10^{-6}$ Å/mm^2. The practical resolving power was near 5×10^5. This permitted the splitting of components separated by 0.005 Å (or 85×10^{-3} cm^{-1}). The microphotometer trace of this line and the scheme of the $6s6p^3\ ^5S_2$–$6p^2\ ^1D_2$ transition are given in Fig. 2.

All possible combinations of the distances between components of this line were measured by means of an Abbé comparator. For each distance, 30 independent measurements were performed and the average value, as well as its standard error, were computed. For a check of these measurements, thirty microphotometer traces enlarged 800 times were taken and the same distances were measured. This procedure permitted, utilizing our modest equipment, to determine hfs intervals with an average accuracy of 5×10^{-5} Å or $1\times10^{-3}\ \mathrm{cm}^{-1}$. This accuracy is indispensable to achieve a theoretical interpretation of the experimental hfs data.

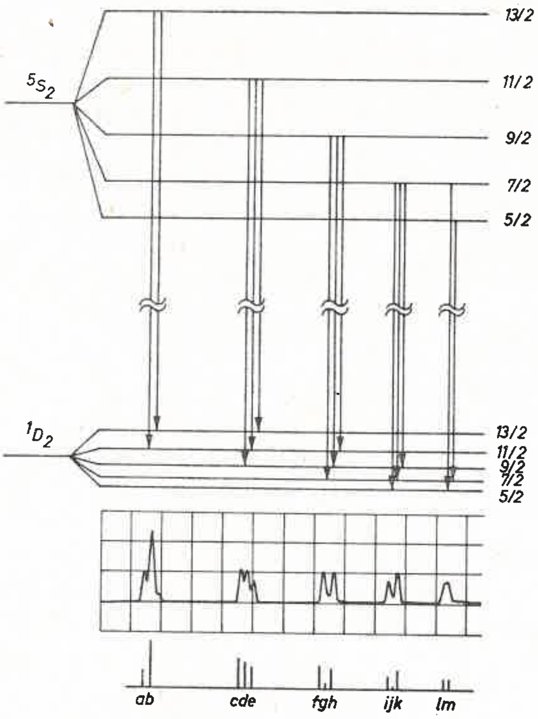

Fig. 2. The hyperfine structure of the line $\lambda = 2368.2$ Å of the Bi II ion

Each W_{ij} distance between hfs components according to e.g. Kopfermann [3] can be written as:

$$W_{ij} = \tfrac{1}{2}K_iA - \tfrac{1}{2}K_j^*A^* + Q_iB - Q_j^*B^*, \tag{1}$$

where:

$$K = F(F+1) - J(J+1) - I(I+1), \qquad Q = \frac{3K(K+1) - 2J(J+1)2I(I+1)}{8I(2I-1)2J(2J-1)},$$

and F, I and J denote the quantum numbers, A is the magnetic dipole constant, and B the electric quadrupole hfs constant. The symbols with an asterisk refer to an upper level. Using the above equation the 55 measured distances between hfs components on the spectroscopic plate were expressed by a set of constants appropriate to its structure. These constants are $A(^1D_2)$, $B(^1D_2)$, $A(^5S_2)$ and $B(^5S_2)$.

The expressions involving the constants were fitted to the experimental values of the hfs intervals. The results of the least squares fit are (in 10^{-3} cm^{-1})

$$A(^1D_2) = 27.18(11), \quad B(^1D_2) = -8.3(1.6),$$

$$A(^5S_2) = 401.35(11), \quad B(^5S_2) = -9.4(1.6). \tag{2}$$

The values in parentheses are the standard errors in the last digits of each value quoted.

We also intended to determine the precise value of the $6s6p^3\ ^5S_2$ energy level. By means of the Fe I and Fe II standard spectra [6], we determined the wavelength of the strongest component of the 2368 Å line. Converted to vacuum, it has the value: $\lambda_{\mathrm{vac}} = 2368.877\ (3)$ Å. Using the values [2] and [3], the difference in energy between the centres of gravity of the hfs patterns of the $^5S_2$ and $^1D_2$ levels (see Fig. 1) were calculated as:

$$E(^5S_2)-E(^1D_2) = 42\,210.728(10)\ \mathrm{cm}^{-1}.$$

Assuming for the $^1D_2$ level the value

$$E(^1D_2) = 33\,938.86(2)\ \mathrm{cm}^{-1}$$

given by Kołyniak et al. [8], the position of the $6s6p^3\ ^5S_2$ level on the energy scheme is:

$$E(6s6p^3\ ^5S_2) = 76\,149.588(30)\ \mathrm{cm}^{-1}$$

with reference to the $6p^2\ ^3P_0$ ground level of Bi II. This value is larger than the one, 76 147 cm^{-1}, given by Moore [9].

3. *Discussion of the results*

Once the experimental values of the hyperfine interaction constants for many levels of one configuration are available, it becomes possible to test the theory. The generally used procedure is (*i*) to develop intermediate coupling wave functions in the SL basis for the states of interest, (*ii*) to postulate the effective hyperfine Hamiltonians, (*iii*) to work out the matrix element of these operators between the SL basis states, (*iv*) from these matrix elements and wave functions, to develop expressions for the hfs interaction constants, and finally (*v*) to least-squares fit the theoretical expressions to the experimental values of the hfs constants using the least squares method varying the parameters occurring in the effective Hamiltonians.

The form of the hyperfine Hamiltonians, introduced by Sandars and Beck [10], and the procedure for obtaining the theoretical parametrized expressions for the hyperfine interaction constants have already been published in Acta Physica Polonica [11, 12]. Therefore, we shall give only a Table of the matrix elements (Table IV) and the final form of the theoretical expression for the magnetic-dipole constants A and electric-quadrupole constants B. The procedure for developing intermediate-coupling wave functions always involves fine structure analysis and is described below.

3.1. Fine structure analysis and wave functions

It has been shown in Ref. [13] that the accuracy of the eigenvectors has a peculiarly strong been influence on the values obtained for the hyperfine structure parameters, as well as on the interpretation of the hyperfine structure interactions. The accuracy of the eigenvectors can be improved by taking into account all the possible kinds of interactions predicted theoretically for the configuration considered.

The ground configuration of the Bi II ion is $6s^26p^2$ (see Fig. 1). Energy-level calculations of heavy atoms are usually based on a Hamiltonian comprising only a Coulomb interaction between the electrons and their individual spin-orbit interactions. These interactions are represented by the Slater parameters F_k and the spin-orbit constant ζ_{nl}. Generally, results of such calculations are in rather poor agreement with the energy levels observed.

In second-order perturbation theory, the following interactions are predicted for the p^2 configuration: (*i*) two-electron effective electrostatic interactions with distant configuration [14], which are associated with an $\alpha L(L+1)$ term, where L is the total orbital quantum number for the particular state, (*ii*) electrostatically correlated spin-orbit interactions [15], and, to first-order perturbation theory, (*iii*) two-electron magnetic interaction [16] within one configuration as orbit-orbit, spin-other orbit, and spin-spin interaction.

As follows from theoretical calculations by Fraga, Karwowski and Saxena [17], the contribution of the interactions cited under (*iii*) to the fine structure of the heavy atoms is practically zero. Moreover, the orbit-orbit interaction includes a term proportional to $L(L+1)$ as for the configuration interactions and is thus inseparable from them.

The present work is a first treatment taking into consideration electrostatically correlated spin-orbit interactions in the fine structure analysis of any heavy element. We calculate the matrix elements using formula (12) of Ref. [15]. The parameter $Q^{(2)}$, in the case of the $6s^26p^2$ configuration, is defined as follows:

$$Q^{(2)} = \tfrac{6}{5} \sum_{n'} \frac{R^{(2)}(6p6p,\, 6pn'p)\zeta_p(6p,\, n'p)}{\Delta E(6p,\, n'p)}$$

with $R^{(2)}$ and ζ_p, respectively, Slater's parameter and the spin-orbit parameter, and $\Delta E(6p, n'p)$ the energy separation between the configuration $6s^26p^2$ and an excited configuration $6s^26pn'p$. We introduced the second-order corrections (*i*) and (*ii*) successively in order to investigate their contributions to the fine structure splittings. Therefore, three versions of the calculations were performed:

In version I, the experimental values of the energy levels were fitted by the Slater integrals F_0 and F_2 and the spin–orbit parameter ζ_p.

In version II, besides the parameters F_0, F_2 and ζ_p, the parameter α was included.

Version III contains both kinds of configuration interactions with distant configurations. The results of our calculations are given in Tables II and III. It is seen from Table II that taking into account the α parameter improves the fit significantly. This conclusion is contrary to a conclusion of Rosén [18] for other heavy elements, such as Pb and Po. In version III the fit is perfect, because the number of energy levels is equal to the number of free parameters.

TABLE I

Nonzero matrix elements of the fine structure Hamiltonian for the np^2 configuration

$(SLJ,\ S'L'J')$	Matrix elements
$^3P_0,\ ^3P_0$	$F_0-5F_2-\zeta_p+2\alpha-\frac{1}{6}Q^{(2)}$
$^3P_1,\ ^3P_1$	$F_0-5F_2-\frac{1}{2}\zeta_p+2\alpha-\frac{1}{12}Q^{(2)}$
$^3P_2,\ ^3P_2$	$F_0-5F_2+\frac{1}{2}\zeta_p+2\alpha+\frac{1}{12}Q^{(2)}$
$^1D_2,\ ^1D_2$	$F_0+F_2+6\alpha$
$^1S_0,\ ^1S_0$	F_0+10F_2
$^3P_2,\ ^1D_2$	$\frac{\sqrt{2}}{2}\zeta_p+\frac{\sqrt{2}}{60}Q^{(2)}$
$^1S_0,\ ^3P_0$	$-\sqrt{2}\zeta_p+\frac{\sqrt{2}}{3}Q^{(2)}$

TABLE II

Experimental and calculated values of the energy levels of the ground configuration of the Bi II ion (in cm^{-1})

Level	Experimental	Experimental minus calculated		
		Version I	Version II	Version III
$^3P_0$	0	75.78	87.13	0
$^3P_1$	13325.57	64.22	−61.00	0
$^3P_2$	17031.78	−223.55	−113.51	0
$^1D_2$	33938.86	65.87	105.39	0
$^1S_0$	44173.85	176.71	−17.99	0

TABLE III

Values of the fine structure parameters for the $6p^2$ configuration of the Bi II ion (in cm^{-1})

Version	Parameters				
	F_0	F_2	ζ_p	α	$Q^{(2)}$
I	24975	1170.5	11722	—	—
II	25175	1144.0	11792	−86.5	—
III	25568	1139	12001	−215	1371

To illustrate the second-order perturbations, the shift of the level $^3P_1$ caused by them was calculated. The contributions are $-431\ \text{cm}^{-1}$ from the electrostatic configuration interaction and $-114\ \text{cm}^{-1}$ from the correlated spin-orbit interaction, respectively.

The eigenvectors determined in version III were used in the subsequent hfs analysis. The eigenvectors for the levels with $J = 2$ are:

$$|{}^3P_2\rangle = 0.704176|{}^3P_2^{SL}\rangle - 0.710025|{}^1D_2^{SL}\rangle,$$
$$|{}^1D_2\rangle = 0.710025|{}^3P_2^{SL}\rangle + 0.704176|{}^1D_2^{SL}\rangle. \tag{3}$$

The superscript "SL" is used to distinguish the SL-basis state from the actual state of the atom. We see above that SL coupling is a very poor approximation for the levels ($J = 2$) of Bi II.

In order to complete our fine structure analysis we also give the eigenvectors in j–j coupling notation:

$$|{}^3P_2\rangle = 0.165024|3/2\ 3/2;\ 2\rangle + 0.986289|3/2\ 1/2;\ 2\rangle,$$
$$|{}^1D_2\rangle = 0.986289|3/2\ 3/2;\ 2\rangle - 0.165024|3/2\ 1/2;\ 2\rangle. \tag{4}$$

We see that the Bi II states are much closer to the j–j coupled states. This remark will be useful in the interpretation of hfs splitting.

3.2. Results of the hfs analysis

As already mentioned, the detailed derivation of the theoretical expressions for the hfs constants is omitted. We give only indispensable matrix elements of the hfs operators in Table IV and the final form of these expressions.

TABLE IV

Matrix elements diagonal in J for magnetic-dipole and electric-quadrupole hfs interactions in the p^2 electron configuration

$A(SLJ,\ S'L'J)$	a_p^{01}	a_p^{10}	a_p^{12}
${}^3P_2,\ {}^3P_2$	0.5	0.5	0.1
${}^1D_2,\ {}^1D_2$	1	0	0
${}^3P_2,\ {}^1D_2$	0	0	$-\frac{3\sqrt{2}}{20}$
${}^3P_1,\ {}^3P_1$	0.5	0.5	-0.5

$B(SLJ,\ S'L'J)$	b_p^{02}	b_p^{11}
${}^3P_2,\ {}^3P_2$	-0.4	$\frac{\sqrt{5}}{15}$
${}^1D_2,\ {}^1D_2$	0.8	0
${}^3P_2,\ {}^1D_2$	0	$-\frac{\sqrt{10}}{30}$
${}^3P_1,\ {}^3P_1$	0.2	$\frac{\sqrt{5}}{30}$

The A-constants found for the magnetic dipole interactions are:

$$A(^3P_1) = 0.5a^{01}+0.5a^{10}-0.5a^{12}, \tag{5}$$

$$A(^3P_2) = 0.75207a^{01}+0.24793a^{10}+0.26171a^{12}, \tag{6}$$

$$A(^1D_2) = 0.74793a^{01}+0.25207a^{10}-0.16171a^{12}, \tag{7}$$

where a^{01} represents the interaction between the nuclear magnetic-dipole moment $\hat{\mu}_I$ and the magnetic field produced by purely orbital motion of the electron; a^{12} represents the interaction between $\hat{\mu}_I$ and the magnetic moment of the electron $-2\mu_B\hat{S}$; and a^{10} — the effects of relativity and configuration interaction ("core polarization effect").

The above set of equations was solved taking for $A(^1D_2)$ the value determined in this work (Eq. (2)) whereas for $A(^3P_1)$ and $A(^3P_2)$ the values given by Cole [6], which are $-82.5(7)\times 10^{-3}$ cm^{-1} and $115.1(2.8)\times 10^{-3}$ cm^{-1}, respectively. The obtained experimental values of the hfs parameters $a^{k_sk_l}$ are the following (in 10^{-3} cm^{-1}):

$$a^{01} = 101.1(7), \quad a^{12} = 206.1(2.4), \quad a^{10} = -60.1(3.1). \tag{8}$$

The hfs parameters $a^{k_sk_l}$ according to e.g. Armstrong [19] can be expressed as follows:

$$a^{k_sk_l} = 3.18263g_I\langle r^{-3}\rangle^{k_sk_l} = 3.18263g_I\langle r^{-3}\rangle_{6p}F^{k_sk_l}, \tag{9}$$

where $a^{k_sk_l}$ are in 10^{-3} cm^{-1} and all radial parameters $\langle r^{-3}\rangle$ in atomic units; $\langle r^{-3}\rangle^{k_sk_l}$ are relativistic radial parameters, $F^{k_sk_l}$ are relativistic correction factors (RCF), and g_I is the nuclear g-factor, which for ^{209}Bi has a value of 0.898 n.m. [20].

Hence, the experimentally determined values of the relativistic radial integrals for the configuration $6s^26p^2$ of Bi II amount to:

$$\langle r^{-3}\rangle^{01}_{6p} = 35.4(3), \quad \langle r^{-3}\rangle^{12}_{6p} = 72(1), \quad \langle r^{-3}\rangle^{10}_{6p} = -21(1). \tag{10}$$

We shall now use these values to study the relativistic and configuration effects in the hyperfine structure.

The electric-quadrupole interaction constants B, similarly as Eqs. (5)–(7), can also be written as functions of the hfs parameters:

$$B(^3P_1) = 0.2b^{02}+0.07454b^{11}, \tag{11}$$

$$B(^3P_2) = 0.20496b^{02}+0.17932b^{11}, \tag{12}$$

$$B(^1D_2) = 0.19504b^{02}-0.03025b^{11}, \tag{13}$$

where

$$b^{k_sk_l} = 7.83788Q\langle r^{-3}\rangle^{k_sk_l} = 7.83788Q\langle r^{-3}\rangle_{6p}R^{k_sk_l}, \tag{14}$$

Q is the nuclear quadrupole moment, and $R^{k_sk_l}$ are relativistic correction factors (RCF).

In this work we refrain from a detailed analysis of the quadrupole interactions, because the errors in the experimental data are about 20% making it impossible to draw precise conclusions. Roughly, the constant $B(^1D_2)$ can be used to test the breakdown of the SL--coupling. Its value changes very strongly from the SL-limit to the j–j limit of coupling. In the case of Bi II it ranges from about 80×10^{-3} cm^{-1} for SL to zero for j–j coupling. The small experimental value $-8.3(1.6) \times 10^{-3}$ cm^{-1} confirms that the level denoted as "$^1D_2$" is very near the limit of j–j coupling, Eq. (4).

3.3. Examination of the relativistic effects

The latest *ab initio* theoretical calculations of the relativistic effects of hfs performed by Lindgren and Rosén [5] for many elements show significantly better agreement with the experimental data. The RCF given by Rosén and Lindgren [21] have been obtained as the ratio of relativistic and non-relativistic values of $\langle r^{-3} \rangle$ performing non-relativistic and relativistic calculations by the same Self-Consistent-Field method. Those calculations were performed mostly for the ground configurations of free atoms. According to the above, these RCF cannot be automatically applied to other electron configurations or ions of the same atoms in contradistinction to the RCF given by Kopfermann, which are n-independent. It can be seen from Eqs. (9) and (14) that each hfs parameter determined from experiment is the product of the nuclear moment, the non-relativistic value $\langle r^{-3} \rangle_{nl}$, and the RCF. Likewise, the fine structure spin-orbit parameter ζ is defined as:

$$\zeta = \alpha^2 R_\infty Z_i \langle r^{-3} \rangle_{nl} H_r,$$

where H_r is also a RCF, Z_i the effective nuclear charge, α the fine structure constant and R_∞ the Rydberg constant.

In order to decide whether the relativistic effects on the $6s^26p^3$ configuration of Bi I and on the $6s^26p^2$ configuration of the Bi II ion are identical or different, we form the ratios of the hfs, or the fs parameters. As can be seen from Table V, the differences in the experimental values of the parameters obtained for Bi I and for Bi II, respectively, originate only in the non-relativistic parameters $\langle r^{-3} \rangle_{6p}$. Thus, we draw the conclusion that the relativistic effects in Bi I and Bi II are practically identical.

TABLE V

Comparison between the spin-orbit parameters ζ_p and radial integrals of Bi I and Bi II

Configuration	Experimental				Theoretical nonrelativistic [17]	
	ζ_p [in cm^{-1}]	$\langle r^{-3} \rangle^{01}$	$\langle r^{-3} \rangle^{12}$	$\langle r^{-3} \rangle^{12}/\langle r^{-3} \rangle^{01}$	ζ_p [in cm^{-1}]	$\langle r^{-3} \rangle_{6p}$
$6s^26p^3$ Bi I	10093 [a]	30.3 [a]	61.7 [a]	2.036	6831	14.74
$6s^26p^2$ Bi II	12001	35.4(3)	72(1)	2.034	8118	17.51
The ratio Bi II/Bi I	1.189	1.17(2)	1.17(2)	1.001	1.188	1.188

[a] Values taken from Ref. [22].

To check the *ab initio* calculated RCF [21] with the experimental data, we propose to introduce "semiempirical RCF", which we define as follows:

$$\text{“Semiempirical RCF”} = \frac{\text{experimental } \langle r^{-3}\rangle^{k_s k_l}}{\text{theoretical non-relativistic } \langle r^{-3}\rangle_{nl}}.$$

We performed these calculations for Bi I and Bi II and the results are compared in Table VI with the RCF calculated *ab initio* by different methods. It can be remarked that "semiempirical RCF" F^{01} and F^{12}, and also their ratio F^{12}/F^{01} for Bi I and Bi II, are to within the experimental error identical and agree to within $1 \div 2\%$ with the RCF calculated by means of the OHFS or HF methods.

TABLE VI

Comparison between semi-empirical RCF with those calculated *ab initio*

Element	F^{01}	F^{12}	$-F^{10}$	F^{12}/F^{01}	H_r	Method
Bi II $6s^26p^2$	2.022	4.112		2.034	1.478	Semi-empirical calculation
Bi I $6s^26p^3$	2.056	4.186		2.036	1.478	
Bi I $6s^26p^3$	1.998	4.168	0.726	2.086		OHFS[a]
	2.068	4.128	0.678	1.996		HF[a]
	1.644	2.788	0.333	1.696	1.185[b]	Kopfermann[a]
	1.737	3.011	0.383	1.733		Breit[a]
				1.894		Schwartz [4]

[a] Values taken from Ref. [21]. [b] Value taken from Ref. [3].

We were unable to test the RCF F^{10}, because the experimental value $\langle r^{-3}\rangle^{10}$ is the sum of the relativistic and configuration interaction effects, as follows:

$$\langle r^{-3}\rangle^{10} = \langle r^{-3}\rangle^{10}_{\text{rel}} + \langle r^{-3}\rangle^{10}_{\text{conf}}.$$

Nonetheless, we can separate these two effects. The effect of relativity is defined as:

$$\langle r^{-3}\rangle^{10}_{\text{rel}} = \langle r^{-3}\rangle_{6p} F^{10} = -12.3 \text{ a.u.},$$

where we assume $F^{10} = -0.702$ as the average value of the RCF F^{10} from the OHFS and HF methods. The remaining

$$\langle r^{-3}\rangle^{10}_{\text{conf}} = \langle r^{-3}\rangle^{10} - \langle r^{-3}\rangle^{10}_{\text{rel}} = -8(1) \text{ a.u.}$$

is the "core polarization effect", caused by excitation of the inner *s*-electrons to the empty shells.

The contribution from the configuration interaction amounts to forty percent and is significantly smaller than for the configuration $6s^26p^3$ of Bi I, where it amounts to about 57% (~ -13.6 a.u.).

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