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AMPLIFICATION OF THE ELECTRON ELECTRIC DIPOLE MOMENT IN ATOMS

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Enhancement coefficients for the electron electric dipole moment (EDM) in U, Lu, Eu, Ce, Cs, Fe and Cr are calculated for the purpose of determining the most suitable substance for experiments on determination of the EDM. The amplification coefficients are estimated in the quasiclassical approximation, Coulomb functions being employed. Numerical calculations show that the amplification coefficient increases with Z as Z^4.

1. INTRODUCTION

VIOLATION of CP parity in K^0 meson decay indicates the possible existence of electric dipole moments (EDM) for elementary particles. In this connection, the search for EDM in elementary particles is of timely importance.

This paper is devoted to the problem of observing EDM in the electron, and is stimulated by the work of Sandars,[1] who has shown that if the electron has EDM then the atomic levels are shifted in an external electric field in proportion to the field, even in the absence of random degeneracy. In other words, the atom as a whole also acquires an EDM. It was shown in the same papers that the EDM of hydrogen-like atoms, such as the alkali elements, can be larger by many times than that of the electron, i.e., $d_a = Rd_e$, where d_e is the EDM of the electron and R is the enhancement coefficient. In the case of Cs atoms, for example, the enhancement coefficient is R = 130.

This raises the question whether the enhancement of the electron EDM in the atom can be used to perform a simple macroscopic experiment aimed at observing the EDM of the atom. This experiment may have the following scheme: a ferromagnetic sample is placed in a strong magnetic field, the spins of the sample are aligned, and an electric polarization appears:

$$P = d_a N = R d_e N; \tag{1}$$

Here d_a is the EDM of the atom, N is the number of atoms per unit volume of the sample, d_e is the EDM of the electron, and R is the enhancement coefficient. Assuming that $N = 10^{23}\ \text{cm}^{-3}$ and $d_e = 2\times10^{-23}$ e cm,[2] the potential difference produced on the end surfaces of a sample 1 cm thick amounts approximately to

$$V = 3\cdot10^{-6}\, R/\varepsilon \ \ [\text{в}], \tag{2}$$

where ϵ is the dielectric constant of the sample. If the magnetic field varies with a frequency ν, then an alternating current is produced with an amplitude

$$I = 3\nu R\cdot 10^{-19} \ \ [a]. \tag{3}$$

Another experimental formulation is also possible, wherein a ferromagnetic sample is placed in an electric field and the magnetization is measured. This variant turns out to be more convenient, since there exist at the present time sensitive magnetometers which make it possible to detect a variation of a magnetic field by 10^{-9} G against a background of several kilogauss.[3] It becomes possible here either to duplicate or improve the upper estimate of the electron EDM, which equals 2×10^{-23} cm.[2]

To estimate the sensitivity of these experiments, it is necessary to calculate the enhancement coefficients for the most convenient materials. One might use for this purpose the formulas published by Sandars[1] for the enhancement coefficient. However, since these formulas are valid only for alkali elements, and no derivation was presented for them in general, it is necessary to repeat the theoretical calculations, and to duplicate some of the numerical calculations by Sandars in order to check the validity of the numerical algorithm.1)

The calculation method and the results are given in Sec. 2. In Sec. 3 we discuss the causes of the enhancement and various corrections. The appendix presents a brief derivation of the single-particle enhancement coefficient.

2. CALCULATION OF THE ENHANCEMENT COEFFICIENT

According to formula (A.7) of the appendix, the enhancement coefficient of the electron EDM in an atom, in the case of a solitary unpaired electron with angular momentum l_0 and at small values of $(Z\alpha)^2$, is equal to a linear combination of the partial coefficients R_1 and R_2, where

$$R_{1,2} = 2(Z\alpha)^2 \int_0^\infty \psi_{1,2} \frac{d}{dr}\left(\frac{v}{r}\right)^2 \psi_0\, dr, \tag{4}$$

Z is the charge of the nucleus, α is the fine-structure constant, v/r is the self-consistent potential, ψ_0 is the solution of the equation

$$\left[\frac{1}{2}\frac{d^2}{dr^2} - \frac{1}{2}\frac{l_0(l_0+1)}{r^2} + \frac{v}{r} - \mathscr{E}_0\right]\psi_0 = 0, \tag{5}$$

and $\psi_{1,2}$ are subject to the equations

$$\left[\frac{1}{2}\frac{d^2}{dr^2} - \frac{l_{1,2}(l_{1,2}+1)}{2r^2} + \frac{v}{r} - \mathscr{E}_0\right]\psi_{1,2} = r\psi_0 \tag{6}$$

at $l_1 = l_0 - 1$ and $l_2 = l_0 + 1$.

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1) After submission of this article, Sandars published a new paper [4] containing a detailed theoretical derivation of the enhancement coefficient for alkali elements.

If the only unpaired electron is an s-electron, as in alkali metals, then the enhancement coefficient for this electron is determined only by the partial coefficient R_2, since obviously there is no correction ψ_1 with $l_1 = l_0 - 1$ when $l_0 = 0$. For multi-electrom atoms with several unpaired electrons, we shall use the same formula (4) for each electron, and the total coefficient will be made up of the individual coefficients in accordance with the law governing the formation of configurations.

The inhomogeneous equations (6) were solved by the Runge-Kutta method, the self-consistent potential v being taken from tables.[5] To calculate the right sides of (6), we solved also Eq. (5). The obtained solution ψ_0 was compared with the tabulated solution[5] in order to check on the method. The obtained agreement was usually good, accurate to rounding off one unit in the last significant figure given in [5]. A discrepancy was obtained only for the f-shell of U, for which the wave function ψ_0 differed from the tabulated values by 10%. The causes of the discrepancy were not established.

The equations were solved on the sides of small and large r up to the large point r_0, where

$$\frac{l_0(l_0+1)}{2r^2} - \frac{v}{r_0} + \mathscr{E}_0 = 0.$$

At the point $r = r_0$, the right-hand and left-hand solutions ψ_i were joined together using the continuity of the function and of the derivative, while ψ_0 was joined together only in accord with the continuity of the function. At the same time, the continuity of the logarithmic derivative of ψ_0 at the joining point was verified. The gap between the left and right logarithmic derivatives of ψ_0 turned out to be sufficiently small in all cases, except the one indicated above (the f shell of $_{92}U$).

Simultaneously with solving the equations, we obtained the normalization integral $\int_0^\infty \psi_0^2 dr$ and the coefficients $R_{1,2}$. The initial values of ψ_0, ψ_0', ψ_i, and ψ_i' ($i = 1, 2$) were determined analytically, by expansion in a Taylor series in powers of r^n at small values of r and in powers of r^{-n} at large values of r. Of the two possible solutions near zero and infinity, we chose only the decreasing solutions. The first coefficient in the expansion was specified arbitrarily, and this determined the solution of the equation in the entire region of integration from left to right (i.e., from small values of r to large ones), or from right to left, uniquely with accuracy to an arbitrary constant factor. The remaining expansion coefficients were determined with the aid of Eqs. (5) and (6). The two arbitrary constants for ψ_i ($i = 1, 2$) were then determined from the two conditions for the continuity of ψ_i and ψ_i' at the point $r = r_0$. For the function ψ_0, the constants were determined from the condition for the continuity of ψ_0 and from the normalization condition

$$\int_0^{r_{max}} \psi_0^2 dr = 1,$$

where r_{max} is the maximum value of r, up to which the numerical calculations were performed; it amounted to[5] $r_{max} \approx 117\, Z^{-1/3}$.

The calculation results are gathered in Table I, which indicates also the values of $r_{1,2}$ (in units of $h/mcZ\alpha$), at which the integrals (4) for $R_{1,2}$ with an upper integration limit $r_{1,2}$ (rather than r_{max}) reach approximately half the final value. It is seen from Table I that the enhancement coefficients of almost all the elements are much smaller than those of Cs. Nonetheless, the macroscopic experiment, owing to the large magnetic susceptibility of the ferromagnetic materials, still turns out to be quite sensitive. It was noted that the smaller l_0 and the closer to zero the energy level of the corresponding electron, the larger the enhancement coefficient.

3. CAUSES OF ENHANCEMENT AND CORRECTIONS TO ENHANCEMENT COEFFICIENT

A. Relativistic Corrections

The partial enhancement coefficients (4) were calculated in an approximation in which $(Z\alpha)^2 \ll 1$, whereas in fact $Z\alpha$ is not small for most elements listed in Table I. This raises the question of what happens if the relativistic effect is completely taken into account.

The relativistic effect changes formula (4) in two ways. First, it is necessary to use for its derivation a relativistic self-consistent potential, for which there are still no tables in the case of the heavy elements; second, it is necessary to use for a specified potential the exact Dirac equation. As shown in [1] calculations with the aid of the exact Dirac equation lead to the appearance of relativistic factors in the partial enhancement coefficients. For the partial enhancement coefficients R_1 and R_2, these factors are given by

$$\eta_i = \frac{|\varkappa_i| (4\varkappa_i^2 - 1)}{\rho_i (4\rho_i^2 - 1)}, \quad i = 1, 2,$$

where

$$\rho_i = (\varkappa_i^2 - (Z\alpha)^2)^{1/2}, \quad \varkappa_1 = -l, \quad \varkappa_2 = l + 1.$$

However, the values of these factors, for the electrons with orbital angular momentum l equal to 2 and 3, which are of interest to us here, differ little from unity, and have therefore been disregarded everywhere except for

Table I

	Element							
	$_{92}U$		$_{71}Lu$	$_{63}Eu$	$_{58}Ce$	$_{55}Cs$	$_{26}Fe$	$_{24}Cr$
	f-electron	d-electron						
R_1	9.1	74.3	45.1	1,8	1.95		0,76	0.56
r_1	6	2	2	6	6		2.5	2
R_2	0.05	—11.44	1,7	0,3	0.003	283 *	0.01	0.1
r_2	20	6	5	20	15	1	10	10

*Taking into account the factors (relativistic – 1.39 and orbital – 1/3), the enhancement coefficient R_{Cs} is equal to 131; Sandars [1] gives $R_{Cs} = 133$.

Cs, where the value of the coefficient served as a criterion for the correctness of the numerical calculations.

It remains to determine the extent to which it is legitimate to use the nonrelativistic self-consistent potential. It would be useful in this case to employ results obtained with the aid of the same nonrelativistic potential and which at the same time are in sufficient agreement with experiment. The only precedent in this respect, similar to the case considered here, is the enhancement of nuclear quadrupole moments, calculated by Sternheimer.[6] The situation in that calculation is somewhat similar to ours.

Indeed, when a nucleus has a quadrupole moment $Q^0_{\mu\nu}$, there is added to the Coulomb interaction between the electron and the nucleus also an interaction

$$U = Q_{\mu\nu}{}^0 r_\mu r_\nu / r^5. \tag{7}$$

This interaction in itself does not lead to a shift of the atomic levels, if the electron shells are closed. On the other hand, if an external charge is placed at a distance R from the atom, then a shift of the atomic levels does take place, and it turns out to be

$$\begin{aligned}\Delta\mathscr{E} &= \left\langle 0 \left| Q_{\mu\nu}{}^0 \frac{R_\mu R_\nu}{R^5} \right| 0 \right\rangle \\ &+ 2\sum_i \frac{1}{\mathscr{E}_i - \mathscr{E}_0} \left\langle 0 \right| Q_{\mu\nu}{}^0 \frac{r_\mu r_\nu}{r^5} \left| i \right\rangle \left\langle i \right| \frac{(r^2\delta_{\mu\nu} - {}^1/_3 r_\mu r_\nu) R_\mu R_\nu}{R^5} \left| 0 \right\rangle \\ &= Q_{\mu\nu} \frac{R_\mu R_\nu}{R^5},\end{aligned} \tag{8}$$

where $Q_{\mu\nu}$ is the effective quadrupole moment of the atom:

$$Q_{\mu\nu} = Q_{\mu\nu}{}^0 (1 + \varkappa). \tag{9}$$

If we estimate $Q_{\mu\nu}$ by the same method as used to estimate the dipole moment, then it turns out that only the outer shells make the main contribution to $Q_{\mu\nu}$, and this contribution for each individual electron is the same as for the dipole moment. The difference between them lies in the fact that whereas for the enhancement of the dipole moment it is necessary to take into account only the unpaired electrons, for the enhancement of the quadrupole moment it is necessary to take into account all the outer electrons. It is seen from (8) that inasmuch as $\Delta\mathscr{E}$ contains matrix elements of the type $\langle 0 | r^{-3} | i \rangle$, the largest contribution to $\Delta\mathscr{E}$ is made by the region $r \sim 1/Z$ where, generally speaking, relativistic effects are important. Therefore, if complete allowance for the relativistic effects, particularly the use of a relativistic self-consistent potential, can radically change the entire picture, then the predictions concerning the quadrupole moments would likewise be incorrect. In fact, however, the presence of the strong quadrupole enhancement predicted by the theory is observed in many experiments,[7] thus confirming the theory at least qualitatively. In some cases there is even quantitative agreement.

For the reader's convenience, we present a few values of the enhancement of the quadrupole moments κ (see (9)):[7]

	Experiment	Theory
Na^+:	7—11	5.6
Rb^+:	40—50	50
Cs^+ :	90	90

We show here only the values in best agreement with experiment. Discrepancies also occur: thus, for example, for Cl^- we have $\kappa_{\text{exp}} = 10$ and $\kappa_{\text{theor}} = 58$; however, the presence of large enhancement has been noted qualitatively in many experimental investigations, particularly, for example, in [8].

B. Causes of Enhancement

Let us discuss now the causes of the enhancement. Salpeter[9] has shown that for a hydrogen-like atom, the enhancement can be due in principle to the fact that there are almost degenerate levels with different parity, the distance between which (the Lamb shift) is small. Therefore the mixing of two functions with different parities will be of the order of

$$\langle i | H_1 | j \rangle / (\mathscr{E}_i - \mathscr{E}_j), \tag{10}$$

where H_1 is the energy of the interaction of the electron EDM with the nucleus, and i and j are two states whose energy levels are separated by the Lamb shift $\mathscr{E}_i - \mathscr{E}_j$, for example the states $2S_{1/2}$ and $2P_{1/2}$. Owing to the smallness of $\mathscr{E}_i - \mathscr{E}_j$, a strong mixing of the wave functions of different parity takes place.

In the case of real atoms, enhancement can be expected only at a large value of the nuclear charge. An estimate of the dependence of the EDM of the atom on the nuclear charge Z can be obtained in the following manner. According to expression (A.6), the EDM of the atom is determined in terms of the matrix elements of the operator $(\sigma \cdot \nabla u^2)$, for which the most important region is $r \sim 1/Z$, where the wave functions must be chosen by starting from the unscreened field of the nucleus. If quasiclassical wave functions are chosen,[10, 11] then the matrix elements of the operator $(\sigma \cdot \nabla u^2)$ turn out to be proportional to Z^3 in both the relativistic and nonrelativistic approximation. On the other hand, if Coulomb wave functions are chosen, then the same matrix elements turn out to be proportional to Z^5 in both the relativistic and nonrelativistic approximation. A similar dependence on Z should be expected for the EDM of the atom, since the matrix elements of the operator $(\mathbf{E} \cdot \mathbf{r})$ are determined principally by the region $r \sim 1$, where the field of the nucleus is completely screened, and therefore the elements turn out to be proportional to unity; in addition, the energy difference in the denominator of (10) is proportional to unity for real atoms. It follows from the foregoing reasoning that the enhancement coefficient depends on the charge of the nucleus like Z^γ, where γ is expected to lie in the interval between 3 and 5.

A direct numerical calculation shows that the enhancement coefficients of heavy atoms actually fit well the interpolation curve

$$R \approx R_{\text{int}} = A_{l_0} Z^4,$$

where A_{l_0} is a constant that depends on the angular momentum of the electron whose enhancement coefficient is sought. Table II lists values of R and R_{int} with the value of A_{l_0} determined for the extreme left element of the corresponding column. The enhancement coefficients R for alkali elements were taken from [1].

C. Multiparticle Behavior

If several unpaired electrons are present in excess

Table II

	$A_0 = 1{,}4\cdot 10^{-5}$			$A_2 = 1{,}7\cdot 10^{-6}$				$A_3 = 1\cdot 10^{-7}$	
	$_{37}$Rb	$_{55}$Cs	$_{87}$Fr	$_{24}$Cr	$_{26}$Fe	$_{71}$Lu	$_{92}$U (d)	$_{63}$Eu	$_{92}$U (f)
R_{int}	27.5	127	1002	0.56	0.78	48	137	1.8	7.1
R	27.5	133	1150	0.56	0.76	45	74	1.8	9.1

of the filled shells, rather than a single one, then the total dipole moment of the atom should be made up of the dipole moments produced by the individual electrons. The resultant dipole moment of the atom turns out to be proportional, in natural fashion, to the total angular momentum **J**. Thus, for example, the EDM of the ions Eu^{+2}, Fe^{+3}, and U^{+3} are respectively equal to

$$0.51 d_e \mathbf{J} \quad (J = 7/2), \quad 0.21 d_e \mathbf{J} \quad (J = 5/2), \quad 0.6 d_e \mathbf{J} \quad (J = 9/2).$$

In all the foregoing examples, the presence of a Russel-Sanders coupling was assumed.

The influence of the electrons from the closed shells on the electrons of the unclosed shells can appear both via exchange interaction, which was taken into account in the present paper to the same extent as it is taken into account in the self-consistent potential, and via polarization of the internal shells (core). The polarization of the core can be taken into account in the following manner.

When the atom is acted upon by an external field **E**, the internal shells of the atom are polarized, $\mathbf{P} = \alpha_c \mathbf{E}$, where α_c is the polarizability of the core. As a result, the field acting on the external electron will no longer be equal simply to $\varphi = \mathbf{E}\cdot\mathbf{r}$, but changes in accordance with the polarization of the internal shells. If account is taken of the polarization of the core in the same manner as used by Sandars,[1] then it is easy to estimate directly the changes that this will bring about in the final value of $\Delta\mathscr{E}$. Indeed, allowance for the core polarization signifies that the field **E** must be multiplied by the screening factor

$$[\alpha_c + (Z-1)r^3] \,/\, [Z\alpha_c + (Z-1)r^3].$$

As a result, the matrix element $\langle 0|\mathbf{E}\cdot\mathbf{r}|i\rangle$ in (A.6) is replaced by

$$\left\langle 0 \left| \frac{\alpha_c + (Z-1)r^3}{Z\alpha_c + (Z-1)r^3} \mathbf{E}\mathbf{r} \right| i \right\rangle.$$

but since the decisive region for this matrix element is $r \sim 1$, and since $Z \gg 1$, this entire matrix element turns out to be simply equal to

$$\langle 0|\mathbf{E}\mathbf{r}|i\rangle \,/\, (1 + \alpha_c).$$

Judging from the table given by Dalgarno,[12] the polarization of the core in all elements is of the order of unity, i.e., we see that allowance for the polarization of the core does not change the enhancement coefficient appreciably.

D. Influence of Crystal Field

Finally, let us stop to discuss the influence of the crystal field on the EDM of the atom. This influence becomes manifest in two ways. First, the crystal-lattice potential is added to the self-consistent potential. Second, the crystal lattice distorts the external field acting on the given atom. With respect to the first, we can state the following: since the EDM of the atom is strongly influenced by the region r_α of the order of $1/Z$, near the nucleus of the given atom, where the Coulomb potential of this atom is equal to Z^2 and the potential due to the remaining lattice atoms is proportional to unity, it follows that the influence of the crystal field on the EDM of the atom is smaller by a factor Z^2 than the influence of the field of the nucleus itself.

Let us see now how the crystal lattice influences the distortion of the external field acting on the atom. For a cubic crystal, this influence is well known and reduces to the fact that each atom is acted upon by a local field

$$\mathbf{E}_{\text{loc}} = \mathbf{E}_0 + (4\pi/3)\mathbf{P}, \tag{11}$$

where $\mathbf{E}_0$ is the external field and $(4\pi/3)\mathbf{P}$ is the Lorentz field due to the polarization $\mathbf{P} = \alpha \mathbf{E}_{\text{loc}}$ of the medium (α—polarizability). By introducing the dielectric constant

$$\varepsilon = \left(1 + \frac{8\pi}{3}\alpha\right) \Big/ \left(1 - \frac{4\pi}{3}\alpha\right),$$

it can be shown that the field acting on the given atom is[13]

$$|\mathbf{E}_{\text{loc}}| = \frac{\varepsilon + 2}{3} |\mathbf{E}_0|. \tag{12}$$

If the crystal is not cubic and consists of elements with different polarizabilities, it is first necessary to find the polarization of the atoms of each element. As shown by ferroelectrics as an example, the effective field may in this case greatly exceed the applied field.

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APPENDIX

The Dirac equation in a self-consistent field can be written in the following symbolic form:

$$[L_D + \xi\alpha^2\gamma_0\Sigma\nabla u(r)]\psi(r) = 0, \tag{A.1}$$

where

$$L_D = p_0 + \alpha^2 u + \gamma_0 - \alpha\gamma_0(\gamma\mathbf{p}), \quad p_0 = 1 - \alpha^2\mathscr{E},$$

α is the fine-structure constant, $\mathscr{E}$ the energy level in units of $\alpha^2 mc^2$, u(r) the self-consistent potential with asymptotic form z/r at zero and 1/r at infinity, $\mathbf{p} = -i\partial/\partial\mathbf{r}$; all the lengths are measured in units of $\hbar/mc\alpha$; γ_0, γ, and Σ are Dirac matrices; ξ is a dimensionless constant connected with the electron EDM in the following manner:

$$\mathbf{d}_e = \xi \frac{e\hbar}{mc\alpha} \Sigma.$$

As $\alpha^2 \to 0$, we get $L_D \to \alpha^2 L_{Sh}$, where $L_{Sh} = -p^2/2 + u - \mathscr{E}$, and Eq. (A.1) goes over into

$$[L_{\mathrm{Sch}} + \xi(\sigma \nabla u)]\psi = 0 \tag{A.2}$$

(σ – Pauli matrices).

Since, according to the Schiff theorem the interaction of the electron EDM with the field can be eliminated from Eq. (A.2), in first order in ξ, by using the transformation

$$\psi = e^{\xi} \quad \psi', \tag{A.3}$$

it is convenient to perform a similar transformation in (A.1)

$$\psi = e^{\xi \Sigma \nabla} \psi' \tag{A.4}$$

so that in the nonrelativistic limit the interaction of the EDM with the field drops out automatically. Equation (A.1) then reduces to the form

$$[L_{\mathrm{D}} + \xi\alpha^2(\gamma_0 - 1)(\Sigma \nabla u)]\psi = 0. \tag{A.5}$$

In the presence of an external homogeneous field **E**, the electron levels are shifted by an amount determined, in the first order in **E** and $\xi\alpha^2$, principally by the terms

$$\Delta\mathscr{E} \approx 2\xi\alpha^2 \sum_i \frac{\langle 0|\mathbf{E}r|i\rangle\langle i|\sigma\nabla u^2|0\rangle}{\mathscr{E}_i - \mathscr{E}_0}. \tag{A.6}$$

For numerical calculations it is convenient to reduce this equation to the form

$$\Delta\mathscr{E} = \frac{2\xi}{2l+1}\left[\frac{l^2 - m^2}{2l-1}R_1 + \frac{(l+1)^2 - m^2}{2l+3}R_2\right]\mu|\mathbf{E}|, \tag{A.7}$$

where

$$R_i = 2(Z\alpha)^2 \int \bar{\psi}_i(r)\left[\frac{d}{dr}\left(\frac{v}{r}\right)^2\right]\psi_0(r)\,dr, \quad i = 1, 2; \tag{A.8}$$

$\bar{\psi}_i(r)$ obey the equations

$$\left[\frac{1}{2}\frac{d^2}{dr^2} - \frac{l_i(l_i+1)}{2r^2} + u(r) - \mathscr{E}_0\right]\bar{\psi}_i(r) = r\psi_0(r); \tag{A.9}$$

l, m, and μ are respectively the angular momentum of the electron, the projection of the angular momentum, and the projection of the electron spin on the external field **E**; $l_1 = l - 1$, $l_2 = l + 1$, and v/r is connected with u(r) by the relation

$$u(r) = v/r. \tag{A.10}$$

As seen from formula (A.8) the parameter of the expansion of $\Delta\mathscr{E}$ in powers of α^2 is actually $(Z\alpha)^2$, and therefore formula (A.7) is valid only when $(Z\alpha)^2 \ll 1$. However, as shown in the text, for electrons with angular momentum $l \geq 2$, formula (A.7) turns out to be a sufficiently good approximation even for heavy atoms. The wuantities R_1 and R_2 are called in the test the partial enhancement coefficients.

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