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Weak-interaction effects in heavy atomic systems. II

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Evaluations of parity-violating and charge-parity—violating effects in heavy one-valence-electron atoms, employing the Hartree-Fock potential and several model potentials, are extended to include first-order electron-electron Coulomb corrections using many-body perturbation theory. Parity-conserving quantities, including valence energies, hyperfine splittings, and oscillator strengths, are also calculated and compared with experiment to determine the reliability of the weak-interaction calculations. It is found that the spread between calculations carried out in first-order perturbation theory starting from different potentials is of the same order of magnitude as the spread between the corresponding lowest-order evaluations. It is concluded that second-order many-body perturbation theory must give significant contributions. Some technical problems associated with going to second order are discussed.

I. INTRODUCTION

In a previous paper,[1] referred to in the following as I, we evaluated weak-interaction effects along with various standard atomic properties in the heavy one-valence-electron atoms Rb, Cs, Au, and Tl. The weak-interaction effects were, first, the induction of a nonvanishing electric dipole matrix element between states of the same nominal parity due to exchange of a Z_0 boson and, second, the induction of an enhanced atomic electric dipole moment (EDM) by a charge-parity (CP)—violating electron EDM. We employed several different potentials in I to estimate the reliability of the calculations. Our principal result was that different potentials gave predictions for parity-violating and CP-violating dipole matrix elements having a spread of up to 20%, which we used as a measure of the accuracy of our predictions. While this accuracy is sufficient to establish qualitatively the existence of neutral weak-current effects, greater accuracy is clearly desirable. In particular, recent measurements of parity violation in Cs have reached the 8% level,[2] and if atomic theory can achieve the same accuracy, information about the Weinberg angle[3] and one-loop radiative corrections to weak interactions[4] in a low-energy regime, complementary to high-energy probes of the weak interactions can be obtained. Furthermore, if an atomic EDM were to be discovered, a 20% theoretical calculation of the enhancement factor would permit a determination of the electron EDM only at the 20% level, so improvements in theoretical EDM predictions are also desirable. While our program is directed at weak-interaction effects, we note that any techniques developed to predict these effects to within a few percent should also be applicable to accurate studies of other atomic properties such as hyperfine splittings, valence energies, and oscillator strengths, which are of considerable interest in their own right.

While other approaches could be applied to study heavy atomic systems, for example, multiconfiguration Hartree-Fock methods,[5] we have chosen to employ many-body perturbation theory.[6] Our hope is that, if we make a physically sensible lowest-order approximation, two or three orders of perturbation theory will suffice to achieve a few percent accuracy. In this paper we present the results of calculations carried out to first order in the electron-electron interaction. As described in I, our plan is to carry out calculations starting from several different potentials and to use the spread in values between the calculations in a given order of perturbation theory to measure the reliability of the corresponding calculations. Presumably, this spread will vanish as higher-and-higher-order perturbations are included. For this purpose we employ three model potentials (described in the Appendix) together with the Hartree-Fock potential. The principal result of our first-order calculations is that there is a great deal of sensitivity to core polarization, so that the spread in values between quantities calculated in first order starting from different potentials ranges up to 20%, comparable to the spread found in lowest order. While somewhat better results were obtained for excited-state properties, due to the diminished effect of core polarization, it is clear that predictions of ground-state atomic properties at a level well under 10% will require the use of second-order perturbation theory and perhaps some form of infinite summation.

During the past decade, various many-body calculations of parity violation in heavy atoms have appeared, several of which go beyond the present calculations and include second-order correlation corrections. Closest to the present approach is the calculation of Mårtensson-Pendrill,[7] which is a complete first-order calculation of the parity violation in Cs starting from a Hartree-Fock potential. The first-order parity-violating matrix element in Cs based on the Hartree-Fock potential in the present paper agrees very well with the result of Ref. 7. Indeed, such agreement is expected since the principal difference between the present calculation and that of Mårtensson-Pendrill concerns the way in which perturbation theory is implemented. We also mention the elegant work of Dzuba *et al.*[8] on Cs which also starts with a Hartree-Fock potential and includes both first- and second-order correlation corrections. In first order, our calculations for Cs, as well as those of Mårtensson-Pendrill, agree with those of Dzuba *et al.* The calculations of Ref. 8 indicate that the second-order corrections in the Hartree-Fock case are substantially smaller than the 20% mentioned above. It should therefore be very interesting to do second-order calculations starting from other potentials to determine whether or not the results cluster around the Hartree-Fock values. Last, we mention the second-order calculations of Das *et al.*[9] and of Das[10] on Tl and Cs, respectively. While these calculations also start from a Hartree-Fock potential, we have discrepancies (of unknown origin) with the corresponding matrix elements even in lowest order.

The paper is organized as follows. In Sec. II various results from many-body perturbation theory are derived and a discussion of the corresponding angular-momentum factors is presented. In Sec. III the use of one-particle radial Green's functions to evaluate the formulas developed in Sec. II is described, and formulas for excitation energies, oscillator strengths, hyperfine splittings, parity-violating matrix elements, and EDM enhancement factors are presented. In Sec. IV we give the results of the numerical evaluation of these quantities. In Sec. V we summarize our results and describe several technical points that must be addressed before extending our calculations to second order.

II. RESULTS FROM MANY-BODY PERTURBATION THEORY

A. Basic formulas

In this section we employ first-order perturbation theory to determine the corrections to energies and matrix elements arising from the difference between the electron-electron Coulomb interaction and the model potential. While there is a well-known graphical representation for many-body perturbation theory,[6] it has usually been applied to cases where the model potential is a Hartree-Fock (HF) potential, leading to cancellations of many graphs. We present our results for the model potential case first and then describe the modifications required to treat the HF potential.

For atoms with a single valence electron, the ground state can be represented as

$$|v\rangle = a_v^\dagger |0_c\rangle , \tag{2.1}$$

where $|0_c\rangle$ represents the filled Fermi sea of the atomic core. Using the labels $a,b,c,\ldots$ for core states, and $n,m,\ldots$ for all other states, we see that $a_n|0_c\rangle=0$ and $a_a^\dagger|0_c\rangle=0$. We also use the labels $i,j,\ldots$ to represent either core or excited states. Now, in I we decomposed the full Hamiltonian H into H_0 and V_c, where

$$\begin{aligned} H_0 &= \sum_i [\boldsymbol{\alpha}_i\cdot\mathbf{p}_i + m\beta_i + V(r_i)] , \\ V_c &= \tfrac{1}{2}\alpha \sum_{i,j} \frac{1}{|\mathbf{r}_i-\mathbf{r}_j|} - \sum_i U(r_i) , \\ U(r) &= V(r) + \frac{Z\alpha}{r} . \end{aligned} \tag{2.2}$$

In second quantization these terms have the form

$$\begin{aligned} H_0 &= \sum_i \varepsilon_i a_i^\dagger a_i , \\ V_c &= \tfrac{1}{2} \sum_{i,j,k,l} g_{ijlk} a_i^\dagger a_j^\dagger a_k a_l - \sum_{i,j} U_{ij} a_i^\dagger a_j , \\ g_{ijlk} &= \int \frac{d\mathbf{r}\,d\mathbf{r}'}{|\mathbf{r}-\mathbf{r}'|} \psi_i^\dagger(\mathbf{r}')\psi_l(\mathbf{r}')\psi_j^\dagger(\mathbf{r})\psi_k(\mathbf{r}) . \end{aligned} \tag{2.3}$$

Here $\psi(\mathbf{r})$, which satisfies $(\boldsymbol{\alpha}\cdot\mathbf{p}+m\beta+V)\psi=\varepsilon\psi$, has the explicit form

$$\psi(\mathbf{r}) = \frac{1}{r} \begin{bmatrix} g(r)\chi_\kappa^\mu(\hat{r}) \\ if(r)\chi_{-\kappa}^\mu(\hat{r}) \end{bmatrix} .$$

The model potential $V(r)$ is chosen to give a reasonable overall description of the atom: An important special case is when V is the HF potential $V^{\rm HF}$, defined by

$$V_{ij}^{\rm HF} = \sum_a (g_{iaja} - g_{iaaj}) .$$

As in I, we consider three potentials in addition to $V^{\rm HF}$: the Tietz potential,[11] the Norcross potential,[12] and the Green potential.[13] We have changed some of the parameters used in I to define these potentials, in order to model the core states of the atoms more accurately: The parameters used in the present work are given in the Appendix.

We are interested in the effect of a single action of the perturbation V_c. This can be accounted for by forming the first-order correction to the wave function $|\delta v\rangle$,

$$\begin{aligned} |\delta v\rangle = &\sum_{a,n,m} \frac{g_{nmva}}{\varepsilon_v+\varepsilon_a-\varepsilon_n-\varepsilon_m} a_n^\dagger a_m^\dagger a_a |0_c\rangle \\ &+ \tfrac{1}{2} \sum_{a,b,n,m} \frac{g_{nmab}}{\varepsilon_a+\varepsilon_b-\varepsilon_n-\varepsilon_m} a_v^\dagger a_b a_a a_n^\dagger a_m^\dagger |0_c\rangle \\ &+ \sum_{n(\neq v)} \frac{U_{nv}-V_{nv}^{\rm HF}}{\varepsilon_n-\varepsilon_v} a_n^\dagger |0_c\rangle \\ &+ \sum_{a,n} \frac{U_{na}-V_{na}^{\rm HF}}{\varepsilon_a-\varepsilon_n} a_v^\dagger a_a a_n^\dagger |0_c\rangle . \end{aligned} \tag{2.4}$$

B. First-order energies and one-body matrix elements

All calculations presented here are based on either the first-order energy $\langle v|V_c|v\rangle$ or corrections to the matrix element of a one-body operator t, $\langle\delta v|t|v\rangle+\langle v|t|\delta v\rangle$. The operator t, defined by $t=\sum_{i,j} a_i^\dagger a_j t_{ij}$, where $t_{ij}=\langle i|t|j\rangle$, and where $|i\rangle$ refers to a single-particle wave function, is in our applications proportional to either the hyperfine operator $e\boldsymbol{\alpha}\cdot\mathbf{A}$, $\mathbf{A}=\boldsymbol{\mu}\times\mathbf{r}/r^3$, or to the electric dipole operator ez. Using Eq. (2.3) we find that the first-order correction to the energy is

$$\begin{aligned} E_1 &= E_1^v + \sum_a E_1^a , \\ E_1^v &= (V^{\rm HF}-U)_{vv} , \\ E_1^a &= (\tfrac{1}{2}V^{\rm HF}-U)_{aa} \end{aligned} \tag{2.5}$$

and that the first-order correction D_1 to the matrix element of t is given by

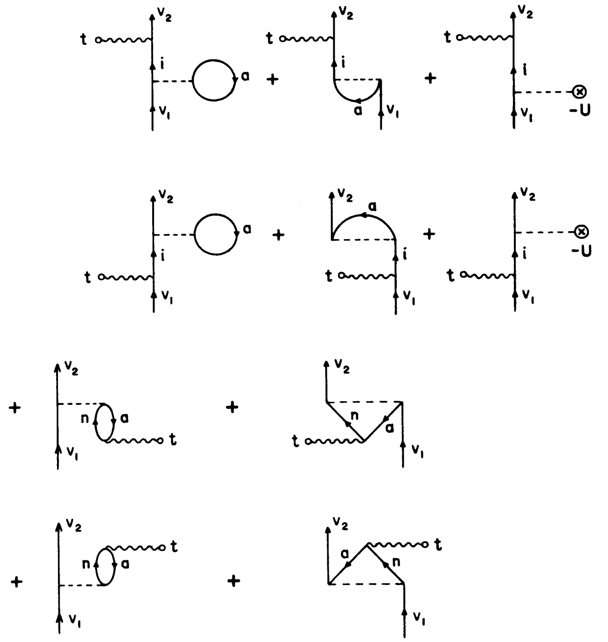

FIG. 1. Brueckner-Goldstone graphs for the first-order Coulomb corrections to the matrix element of a one-body operator t.

$$D_1 = D_1^v + \sum_a D_1^a ,$$

$$D_1^v = \sum_{i(\neq v_1)} \frac{t_{v_2 i}(V^{\mathrm{HF}}-U)_{iv_1}}{\varepsilon_{v_1}-\varepsilon_i} + \sum_{i(\neq v_2)} \frac{(V^{\mathrm{HF}}-U)_{v_2 i}t_{iv_1}}{\varepsilon_{v_2}-\varepsilon_i} , \tag{2.6}$$

$$D_1^a = \sum_n \frac{t_{an}(g_{v_2 n v_1 a}-g_{v_2 n a v_1})}{\varepsilon_a+\varepsilon_{v_1}-\varepsilon_n-\varepsilon_{v_2}} + \sum_n \frac{(g_{v_2 a v_1 n}-g_{v_2 a n v_1})t_{na}}{\varepsilon_a+\varepsilon_{v_2}-\varepsilon_n-\varepsilon_{v_1}} .$$

Note that the sum over n in the expression for D_1^a can be extended to a complete sum over i. The corrections given in Eq. (2.6) are illustrated in Fig. 1. In the HF case D_1^v vanishes. Furthermore, it is necessary to modify the lowest-order HF matrix element by including core random-phase-approximation (RPA) contributions from second- and higher-order perturbation theory in order to ensure gauge independence, as we discuss in more detail in Sec. III B. The modified matrix element t satisfies the equations

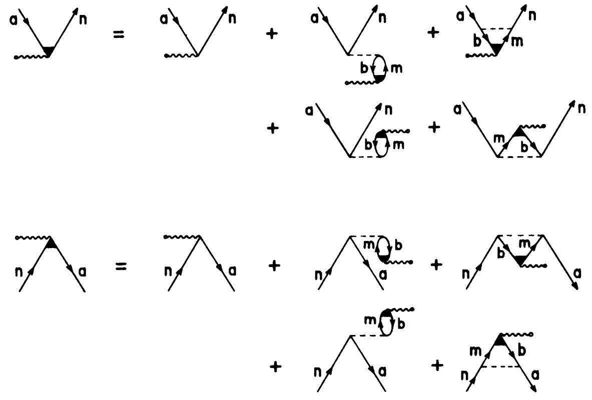

FIG. 2. Graphical representation of the RPA modifications of a one-body vertex.

$$\begin{aligned} t_{an}^{\mathrm{RPA}} &= t_{an} + \sum_{b,m} \frac{t_{bm}^{\mathrm{RPA}}(g_{ambn}-g_{mabn})}{\varepsilon_m-\varepsilon_b+\varepsilon_{v_2}-\varepsilon_{v_1}} + \sum_{b,m} \frac{t_{mb}^{\mathrm{RPA}}(g_{abmn}-g_{bamn})}{\varepsilon_m-\varepsilon_b+\varepsilon_{v_1}-\varepsilon_{v_2}} , \\ t_{na}^{\mathrm{RPA}} &= t_{na} + \sum_{b,m} \frac{t_{mb}^{\mathrm{RPA}}(g_{nbma}-g_{nbam})}{\varepsilon_m-\varepsilon_b+\varepsilon_{v_1}-\varepsilon_{v_2}} + \sum_{b,m} \frac{t_{bm}^{\mathrm{RPA}}(g_{nmba}-g_{mnba})}{\varepsilon_m-\varepsilon_b+\varepsilon_{v_2}-\varepsilon_{v_1}} \end{aligned} \tag{2.7}$$

which are illustrated graphically in Fig. 2. It should be noted that the results in Eqs. (2.6) and (2.7) still remain valid for $v_2 = v_1$.

C. Angular-momentum factors

The next step in evaluating Eqs. (2.5)—(2.7) is to perform the sums over all magnetic quantum numbers. Here the simplicity of dealing with one-valence-electron atoms comes into play, since the summations in Eqs. (2.5)—(2.7) over these quantum numbers are complete, allowing for a full use of the following basic relations involving $3j$ symbols:

$$\sum_{m_1,m_2} \begin{pmatrix} j_1 & j_2 & j_3 \\ m_1 & m_2 & m_3 \end{pmatrix} \begin{pmatrix} j_1 & j_2 & j_3' \\ m_1 & m_2 & m_3' \end{pmatrix} = \frac{1}{2j_3+1}\delta_{j_3 j_3'}\delta_{m_3 m_3'} ,$$

$$\sum_{M_1,M_2,M_3} (-1)^{J_1+J_2+J_3+M_1+M_2+M_3} \begin{pmatrix} J_1 & J_2 & j_3 \\ M_1 & -M_2 & m_3 \end{pmatrix} \begin{pmatrix} J_2 & J_3 & j_1 \\ M_2 & -M_3 & m_1 \end{pmatrix} \begin{pmatrix} J_3 & J_1 & j_2 \\ M_3 & -M_1 & m_2 \end{pmatrix} = \begin{pmatrix} j_1 & j_2 & j_3 \\ m_1 & m_2 & m_3 \end{pmatrix} \begin{Bmatrix} j_1 & j_2 & j_3 \\ J_1 & J_2 & J_3 \end{Bmatrix} . \tag{2.8}$$

We use standard graphical techniques[14] to implement these relations. The $3j$ symbols arise from performing the angular integrations in g_{abcd} after making a partial wave expansion of $1/|\mathbf{r}-\mathbf{r}'|$,

$$g_{abcd}=\sum_{l,q}(-1)^q\langle a|C_l^q|c\rangle\langle b|C_l^{-q}|d\rangle R_l(ab,cd)\ . \tag{2.9}$$

In Eq. (2.9),

$$\begin{aligned}R_l(ab,cd)=\int dr\int dr'\frac{r_<^l}{r_>^{l+1}}&\\ \times[g_a(r)g_c(r)+f_a(r)f_c(r)]&\\ \times[g_b(r')g_d(r')+f_b(r')f_d(r')]&\ ,\end{aligned} \tag{2.10}$$

where $r_<=\min(r,r')$, $r_>=\max(r,r')$, and $C_l^q(\hat{r})=[4\pi/(2l+1)]^{1/2}Y_l^q(\hat{r})$ is a conveniently normalized spherical harmonic. We may write

$$\langle a|C_l^q|b\rangle=(-1)^{j_a-m_a}\begin{pmatrix} j_a & l & j_b\\ -m_a & q & m_b\end{pmatrix}C_l(ab)\ , \tag{2.11}$$

where the reduced matrix element $C_l(ab)$ is

$$\begin{gathered}C_l(ab)=(-1)^{j_a+1/2}\sqrt{[a][b]}\begin{pmatrix} j_a & j_b & l\\ -\frac{1}{2} & \frac{1}{2} & 0\end{pmatrix}\pi(l_a,l_b,l)\ ,\\ \pi(l_a,l_b,l)=\begin{cases}1 & \text{if } l_a+l_b+l \text{ even}\\ 0 & \text{if } l_a+l_b+l \text{ odd}\end{cases}\end{gathered} \tag{2.12}$$

with $[a]=2j_a+1$. For later reference it is also useful to define

$$A(ab,cd,lJ)=(-1)^{l+J-j_b-j_c}C_l(ab)C_l(cd)\begin{Bmatrix} j_a & j_b & l\\ j_d & j_c & J\end{Bmatrix} \tag{2.13}$$

and

$$\Lambda_{alb}=\begin{pmatrix} j_a & j_b & l\\ -\frac{1}{2} & \frac{1}{2} & 0\end{pmatrix}^2\pi(l_a,l_b,l)\ . \tag{2.14}$$

In general, once the magnetic-quantum-number sum has been performed, the remaining angular-momentum quantum numbers are restricted by triangle conditions and by parity selection rules: We refer to the values of the allowed intermediate-state angular-momentum quantum numbers as channels. Each atomic property considered in the following will be characterized by a certain set of channels to be discussed in turn.

III. EVALUATION OF ENERGIES AND MATRIX ELEMENTS USING RADIAL GREEN'S FUNCTIONS

After performing the angle integrations and summing over magnetic quantum numbers, we are left with a sum over the orbital quantum numbers for the allowed channels of products of radial integrals. A powerful alternative to the direct approach of saturating the sum with bound and continuum states is use of the Green's-function method, in which a differential operator is applied to the sum in order to eliminate the denominator, allowing the use of completeness relations to perform the sum. The resulting differential equation can be solved numerically in a time on the order of 1 sec. Using this technique we are able to evaluate the sums in Eqs. (2.5)—(2.7) even when many shells are present with only modest computer resources.[15] Explicit forms of the resulting differential equations will be developed as we discuss the evaluation of oscillator strengths, hyperfine integrals, parity-violating matrix elements, and EDM enhancement factors. In the remainder of this section we present those formulas which we use to compute the atomic properties of interest. The results of numerical evaluation of these formulas will be given in Sec. IV. The equations appropriate for model potentials are presented first, followed by the somewhat different equations used in the HF case.

A. Corrections to valence energies

The simplest expression to consider is the first-order correction to the valence energy, which in fact does not require the use of Green's-function techniques, since the sums encountered range only over the finite core. This term vanishes in the case of the Hartree-Fock potential. The angular-momentum reduction is straightforward and leads to

$$E_1^v=\sum_a[a]\left[R_0(va,va)-\sum_l\Lambda_{vla}R_l(va,av)\right]-U_{vv}\ , \tag{3.1}$$

where the summation index $a=(n_a,\kappa_a)$ runs over all occupied core states. The allowed values of l are determined by the triangle relations $|j_v-j_a|\leq l\leq j_v+j_a$, with the restriction that $l+l_v+l_a$ be even: For valence s states, for example, only one l value, $l=l_a$, is allowed.

B. Corrections to transition matrix elements

We next consider the matrix element of ez between states of different parity, v_2 and v_1. Performing the angular-momentum reduction in Eq. (2.6) with $t=z=rC_1^0(\hat{r})$ and defining $\omega=\varepsilon_{v_2}-\varepsilon_{v_1}$ leads to

$$D_1=(-1)^{j_2-m_2}\begin{pmatrix} j_2 & 1 & j_1\\ -m_2 & 0 & m_1\end{pmatrix}\left[D_v+\sum_a D_a\right]\ , \tag{3.2a}$$

$$D_v=C_1(v_2v_1)(r_{\bar{v}_2v_1}+r_{v_2\bar{v}_1})\ , \tag{3.2b}$$

$$D_a=\sum_q C_1(aq)(r_{a_q^+a}+r_{aa_q^-})\ , \tag{3.2c}$$

where the perturbed orbitals $|\bar{v}\rangle$ and $|a_q^{\pm}\rangle$ associated with the excitation channels allowed by electric dipole selection rules obey the radial equations

$$(H_v-\varepsilon_v)\,|\,\bar{v}\rangle=U\,|\,v\rangle-\sum_a[a]\left[v_0(aa,r)\,|\,v\rangle-\sum_l\Lambda_{vla}v_l(va,r)\,|\,a\rangle\right]+\lambda_v\,|\,v\rangle\ , \tag{3.3a}$$

$$[H_q-(\varepsilon_a+\omega)]\,|\,a_q^+\rangle=-\tfrac{1}{3}C_1(aq)C_1(v_1v_2)v_1(v_1v_2,r)\,|\,a\rangle+\sum_l A(av_1,qv_2,l1)v_l(v_1a,r)\,|\,v_2\rangle\ , \tag{3.3b}$$

$$[H_q-(\varepsilon_a-\omega)]\,|\,a_q^-\rangle=-\tfrac{1}{3}C_1(aq)C_1(v_1v_2)v_1(v_2v_1,r)\,|\,a\rangle+\sum_l(-1)^{j_2-j_1}A(av_2,qv_1,l1)v_l(v_2a,r)\,|\,v_1\rangle\ . \tag{3.3c}$$

The Lagrange multiplier λ_v in Eq. (3.3a) is introduced to account for the restriction $i\neq v$ in Eq. (2.6). The Coulomb interaction potentials $v_l(ab,r)$ in Eqs. (3.3a)–(3.3c) are given by

$$v_l(ab,r)=\int_0^\infty dr'\frac{r_<^l}{r_>^{l+1}}(g_ag_b+f_af_b)'\ .$$

Our procedure in this and subsequent calculations is to determine first the channels q allowed by the angular momentum and parity selection rules. The differential equations in Eq. (3.3) are then solved numerically for each channel and the matrix elements in Eqs. (3.2) are evaluated. Because of the complexity of the calculations, all results presented were evaluated by at least two of us, using independently written computer codes. In evaluating matrix elements we extend the summation over excited states n in Eq. (2.6) to a complete sum, which as mentioned above is permitted because of cancellation of core contributions. This extension simplifies the right-hand side of the resulting differential equation to the form given in Eqs. (3.3b) and (3.3c).

When the HF potential is used, D_v vanishes. While D_a is still given by Eq. (3.2c), the perturbed orbitals $|\,a_q^\pm\rangle$ associated with the excitation channels now satisfy the coupled equations

$$\begin{aligned}
&[H_q-(\varepsilon_a+\omega)]\,|\,a_q^+\rangle\\
&\quad=-\tfrac{1}{3}C_1(aq)C_1(v_1v_2)v_1(v_1v_2,r)\,|\,a\rangle\\
&\qquad+\sum_l A(av_1,qv_2,l1)v_l(v_1a,r)\,|\,v_2\rangle-\sum_{b,p}\tfrac{1}{3}C_1(aq)C_1(bp)[v_1(bb_p^+,r)+v_1(b_p^-b,r)]\,|\,a\rangle\\
&\qquad+\sum_{b,p,l}[A(ab,qp,l1)v_l(ba,r)\,|\,b_p^+\rangle+(-1)^{j_b-j_p}A(ap,qb,l1)v_l(b_p^-a,r)\,|\,b\rangle]\ ,
\end{aligned} \tag{3.4a}$$

$$\begin{aligned}
&[H_q-(\varepsilon_a-\omega)]\,|\,a_q^-\rangle\\
&\quad=-\tfrac{1}{3}C_1(aq)C_1(v_1v_2)v_1(v_2v_1,r)\,|\,a\rangle\\
&\qquad+\sum_l(-1)^{j_2-j_1}A(av_2,qv_1,l1)v_l(v_2a,r)\,|\,v_1\rangle-\sum_{b,p}\tfrac{1}{3}C_1(aq)C_1(bp)[v_1(b_p^+b,r)+v_1(bb_p^-,r)]\,|\,a\rangle\\
&\qquad+\sum_{b,p,l}[A(ab,qp,l1)v_l(ba,r)\,|\,b_p^-\rangle+(-1)^{j_b-j_p}A(ap,qb,l1)v_l(b_p^+a,r)\,|\,b\rangle]\ .
\end{aligned} \tag{3.4b}$$

The summations over b and p in Eqs. (3.4) extend over all channels p associated with excitation of each core orbital b.

In the studies of transition matrix elements we find it useful to compare calculations made using the "length" form, ez, of the dipole matrix element with corresponding calculations using the "velocity" form, ev_z/ω, where $v_z=i\alpha_3$. It is easily established that these two forms lead to identical results in lowest order for the model potential cases, but not for the Hartree-Fock case. In first order one can prove from Eqs. (3.2) and (3.3) that

$$D_1(z)=D_1(v_z/\omega_0)-\omega_1/\omega_0D_0(v_z/\omega_0)\ , \tag{3.5}$$

where ω_1 is the first-order correction to the transition energy. This relation is the obvious extension of the lowest-order length-velocity matrix element identity, with proper account being taken of the first-order shift ω_1 in transition energy. In the Hartree-Fock case, where $\omega_1=0$, the length and velocity forms of the dipole matrix elements differ in zeroth order and in first order. If we are willing to go beyond first-order perturbation theory, however, and include those terms from second and higher order which are obtained by simply iterating the first-order terms, then we can reestablish agreement between length and velocity forms of the dipole operator. The result of the iteration procedure is to replace the first-order core-excitation amplitudes t_{an} in Eq. (2.6) by the corresponding RPA amplitudes given in Eq. (2.7), which include some but not all of the terms from second- and higher-order perturbation theory. Using RPA core-excitation amplitudes in the evaluation of the first-order dipole matrix element D_1 in Eq. (2.6), one finds

$$D_0(z)+D_1(z)=D_0(v_z/\omega_0)+D_1(v_z/\omega_0)\ ,$$

so that the length-velocity agreement is restored. It should be mentioned that the parity-violation calculations of Mårtensson-Pendrill[7] carried out in the Hartree-Fock approximation also employ the RPA amplitudes to describe core excitations. The use of RPA amplitudes has also been advocated by Feldman and Fulton[16] who derive equations equivalent to Eqs. (2.6) and (2.7) in their field-theoretic studies of gauge independence.

C. Hyperfine-splitting corrections

Hyperfine splitting (hfs) is a particularly important aspect of atomic structure for us to understand, since the effect of parity violation is very similar in character to hfs: Both effects arise from an interaction localized at the nucleus, and both affect the valence electron. The very accurate experimental measurements of hyperfine intervals for one-valence-electron atoms provide a stringent test of atomic calculation methods. For hfs calculations the operator $t=-e\boldsymbol{\alpha}\cdot\mathbf{A}$ is conveniently described using vector spherical harmonics,

$$t=-i\sqrt{2}\frac{e\mu}{r^2}\boldsymbol{\alpha}\cdot\mathbf{C}_{10}^{(0)} .$$

Here $\mathbf{C}_{10}^{(0)}$ is defined following the convention of Akhiezer and Berestetskii.[17] Since we deal with atomic states with angular momentum $J=\frac{1}{2}$, there are only two hyperfine levels, described by $F=I\pm\frac{1}{2}$, which are split by $\Delta\nu_{\mathrm{hfs}}=(I+\frac{1}{2})A$, where

$$A=13\,074.7g_I\frac{4\kappa_v}{4\kappa_v^2-1}R_0 \text{ MHz} , \tag{3.6a}$$

$$R_0=2\int dr\, f(r)g(r)/r^2 . \tag{3.6b}$$

First-order perturbation theory leads to a shift in R_0 which can be written as $R_0\rightarrow R_0+R_v+\sum_a R_a$, where

$$R_v=2\int\frac{dr}{r^2}[g_v(r)f_{\bar{v}}(r)+f_v(r)g_{\bar{v}}(r)] \tag{3.7a}$$

and

$$\begin{aligned}R_a=-\frac{2|\kappa_v|+1}{2\kappa_v}&\begin{bmatrix} j_v & 1 & j_v\\ -j_v & 0 & j_v\end{bmatrix}\\ &\times\sum_q C_1(-aq)(\kappa_a+\kappa_q)\\ &\qquad\times 2\int\frac{dr}{r^2}[g_a(r)f_{a_q}(r)+f_a(r)g_{a_q}(r)] .\end{aligned} \tag{3.7b}$$

Here $|\bar{v}\rangle$ satisfies Eq. (3.3a) and the core-excitation channel orbitals $|a_q\rangle$ satisfy

$$\begin{aligned}(H_q-\varepsilon_a)&|a_q\rangle\\ &=\sum_l A(av,qv,l1)v_l(va,r)|v\rangle+\lambda_a|a\rangle\delta_{q\kappa_a} .\end{aligned} \tag{3.8}$$

The corrected hyperfine constant becomes

$$A=13\,074.70g_I\frac{4\kappa_v}{4\kappa_v^2-1}\left[R_0+R_v+\sum_a R_a\right] \text{ MHz} . \tag{3.9}$$

When the potential is a HF potential, R_v vanishes; the correction R_a is still given by Eq. (3.7b), but the perturbed orbitals $|a_q\rangle$ now satisfy the coupled equations

$$\begin{aligned}[H_q-\varepsilon_a]|a_q\rangle=&\sum_l A(av,qv,l1)v_l(va,r)|v\rangle+\sum_{b,p,l}[A(ab,qp,l1)v_l(ba,r)|b_p\rangle+(-1)^{j_b-j_p}A(ap,qb,l1)v_l(b_pa,r)|b\rangle]\\ &+\sum_c\lambda_{cq}|c\rangle\delta_{q\kappa_c} ,\end{aligned} \tag{3.10}$$

where the Lagrange multipliers λ_{cq} are introduced to ensure orthogonality of the perturbed orbitals with occupied orbitals having the same angular symmetry.

D. Parity-violating electric dipole matrix elements

The treatment of parity-violating electric dipole matrix elements parallels the treatment in Sec. III B of transition matrix elements. In this case, however, the transition is between states of the same nominal parity. As explained in I, however, we include in our lowest-order Hamiltonian the weak Hamiltonian

$$H_w=-\frac{G_F}{\sqrt{8}}Q_w\begin{bmatrix} 0 & \rho(\mathbf{r})\\ \rho(\mathbf{r}) & 0\end{bmatrix} . \tag{3.11}$$

Therefore, all states involved have a small admixture of the opposite parity, which allows the dipole matrix element to be nonvanishing. This admixture can enter in either one of the valence states, in the core states, or in the intermediate states. For model potentials the result can be expressed as

$$D=D_0+D_v+D_v'+\sum_a(D_a+D_a') .$$

The term D_0 is the lowest-order contribution evaluated in I,

$$D_0=\begin{bmatrix} j_v & 1 & j_v\\ -j_v & 0 & j_v\end{bmatrix}C_1(-vv)(r_{\tilde{v}_2v_1}+r_{v_2\tilde{v}_1}) , \tag{3.12}$$

where $|\tilde{i}\rangle$ is the weakly perturbed part of the wave function $|i\rangle$. D_v, D_v', D_a, and D_a' are the first-order corrections. Here

$$D_v=\begin{bmatrix} j_v & 1 & j_v\\ -j_v & 0 & j_v\end{bmatrix}C_1(-vv)(r_{\bar{v}_2\tilde{v}_1}+r_{\tilde{v}_2\bar{v}_1}) , \tag{3.13}$$

where the radial functions $|\bar{v}_1\rangle$ and $|\bar{v}_2\rangle$ satisfy Eq. (3.3a). The term D_v' is given by

$$D_v'=\begin{bmatrix} j_v & 1 & j_v\\ -j_v & 0 & j_v\end{bmatrix}C_1(-vv)(r_{v_2\tilde{\bar{v}}_1}+r_{\tilde{\bar{v}}_2v_1}) , \tag{3.14}$$

where $|\tilde{\bar{v}}\rangle$ is the weakly perturbed part of $|\bar{v}\rangle$ and satisfies

$$
\begin{aligned}
(H_{-\dot{v}}-\varepsilon_v)\,|\,\tilde{\bar{v}}\rangle
&=-H_w\,|\,\bar{v}\rangle+(U-V_{\mathrm{HF}})\,|\,\tilde{v}\rangle\\
&\quad+\sum_{b,l}(2j_b+1)[\Lambda_{vlb}v_l(bv,r)\,|\,\tilde{b}\rangle\\
&\qquad+\Lambda_{vl-b}v_l(\tilde{b}v,r)\,|\,b\rangle]+\lambda\,|\,\tilde{v}\rangle\ .
\end{aligned}
\tag{3.15}
$$

The core contributions are

$$
D_a=\begin{pmatrix} j_v & 1 & j_v\\ -j_v & 0 & j_v\end{pmatrix}\sum_q C_1(-aq)(r_{a_q^+\tilde{a}}+r_{\tilde{a}a_q^-})\ ,
\tag{3.16}
$$

where the $|\,a_q^{\pm}\rangle$ satisfy

$$
[H_q-(\varepsilon_a+\omega)]\,|\,a_q^+\rangle=\sum_l A(av,qv,l1)v_l(v_1a,r)\,|\,v_2\rangle\ ,
\tag{3.17a}
$$

$$
[H_q-(\varepsilon_a-\omega)]\,|\,a_q^-\rangle=\sum_l A(av,qv,l1)v_l(v_2a,r)\,|\,v_1\rangle\ ,
\tag{3.17b}
$$

and

$$
D_a'=\begin{pmatrix} j_v & 1 & j_v\\ -j_v & 0 & j_v\end{pmatrix}\sum_q C_1(-aq)(r_{\tilde{a}_q^+a}+r_{a\tilde{a}_q^-})\ ,
\tag{3.18}
$$

where the $|\,\tilde{a}_q^{\pm}\rangle$ satisfy

$$
\begin{aligned}
[H_{-q}-(\varepsilon_a+\omega)]\,|\,\tilde{a}^{+}_{-q}\rangle=-H_w\,|\,a_q^+\rangle-\tfrac{1}{3}C_1(a-q)C_1(v-v)[v_1(\tilde{v}_1v_2,r)+V_1(v_1\tilde{v}_2,r)]\,|\,a\rangle&\\
+\sum_l A(-av,-qv,l1)[v_l(v_1\tilde{a},r)+v_l(\tilde{v}_1a,r)]\,|\,v_2\rangle+\sum_l A(av,qv,l1)v_l(v_1a,r)\,|\,\tilde{v}_2\rangle\ ,&
\end{aligned}
\tag{3.19a}
$$

$$
\begin{aligned}
[H_{-q}-(\varepsilon_a-\omega)]\,|\,\tilde{a}^{-}_{-q}\rangle=-H_w\,|\,a_q^-\rangle-\tfrac{1}{3}C_1(a-q)C_1(v-v)[v_1(\tilde{v}_2v_1,r)+v_1(v_2\tilde{v}_1,r)]\,|\,a\rangle&\\
+\sum_l A(-av,-qv,l1)[v_l(v_2\tilde{a},r)+v_l(\tilde{v}_2a,r)]\,|\,v_1\rangle+\sum_l A(av,qv,l1)v_l(v_2a,r)\,|\,\tilde{v}_1\rangle\ .&
\end{aligned}
\tag{3.19b}
$$

When the potential used is a Hartree-Fock potential, the first-order valence correction vanishes as in the previous problems, and the equations for the core terms are unchanged, except that $|\,a_q^{\pm}\rangle$ and $|\,\tilde{a}_q^{\pm}\rangle$ now satisfy

$$
\begin{aligned}
&[H_q-(\varepsilon_a+\omega)]\,|\,a_q^+\rangle\\
&\quad=\sum_l A(av,qv,l1)v_l(v_1a,r)\,|\,v_2\rangle+\sum_{b,p,l}[A(ab,qp,l1)v_l(ba,r)\,|\,b_p^+\rangle+(-1)^{j_b-j_p}A(ap,qb,l1)v_l(b_p^-a,r)\,|\,b\rangle]\ ,
\end{aligned}
\tag{3.20a}
$$

$$
\begin{aligned}
&[H_q-(\varepsilon_a-\omega)]\,|\,a_q^-\rangle\\
&\quad=\sum_l A(av,qv,l1)v_l(v_2a,r)\,|\,v_1\rangle+\sum_{b,p,l}[A(ab,qp,l1)v_l(ba,r)\,|\,b_p^-\rangle+(-1)^{j_b-j_p}A(ap,qb,l1)v_l(b_p^+a,r)\,|\,b\rangle]\ ,
\end{aligned}
\tag{3.20b}
$$

$$
\begin{aligned}
&[H_{-q}-(\varepsilon_a+\omega)]\,|\,\tilde{a}^{+}_{-q}\rangle\\
&\quad=-H_w\,|\,a_q^+\rangle-V_{\mathrm{HF}}'\,|\,a_q^+\rangle-\tfrac{1}{3}C_1(a-q)C_1(v-v)[v_1(\tilde{v}_1v_2,r)+v_1(v_1\tilde{v}_2,r)]\,|\,a\rangle\\
&\qquad+\sum_l A(-av,-qv,l1)[v_l(v_1\tilde{a},r)+v_l(\tilde{v}_1a,r)]\,|\,v_2\rangle+\sum_l A(av,qv,l1)v_l(v_1a,r)\,|\,\tilde{v}_2\rangle\\
&\qquad-\sum_{b,p}\tfrac{1}{3}C_1(a-q)C_1(-bp)[v_1(\tilde{b}b_p^+,r)+v_1(b_p^-\tilde{b},r)+v_1(b\tilde{b}^{+}_{-p},r)+v_1(\tilde{b}^{-}_{-p}b,r)]\,|\,a\rangle\\
&\qquad+\sum_{b,p,l}\{A(-ab,-qp,l1)[v_l(\tilde{b}a,r)+v_l(b\tilde{a},r)]\,|\,b_p^+\rangle+A(ab,qp,l1)v_l(ba,r)\,|\,\tilde{b}^{+}_{-p}\rangle\\
&\qquad\qquad+(-1)^{j_b-j_p}A(-ap,-qp,l1)[v_l(\tilde{b}^{-}_{-p}a,r)+v_l(b_p^-\tilde{a},r)]\,|\,b\rangle\\
&\qquad\qquad+(-1)^{j_b-j_p}A(ap,qb,l1)v_l(b_p^-a,r)\,|\,\tilde{b}\rangle\}\ ,
\end{aligned}
\tag{3.20c}
$$

$$
\begin{aligned}
&[H_{-q}-(\varepsilon_a-\omega)]\,|\,\tilde{a}^{-}_{-q}\rangle\\
&\quad=-H_w\,|\,a_q^-\rangle-V_{\mathrm{HF}}'\,|\,a_q^-\rangle-\tfrac{1}{3}C_1(a-q)C_1(v-v)[v_1(\tilde{v}_2v_1,r)+v_1(v_2\tilde{v}_1,r)]\,|\,a\rangle\\
&\qquad+\sum_l A(-av,-qv,l1)[v_l(v_2\tilde{a},r)+v_l(\tilde{v}_2a,r)]\,|\,v_1\rangle+\sum_l A(av,qv,l1)v_l(v_2a,r)\,|\,\tilde{v}_1\rangle\\
&\qquad-\sum_{b,p}\tfrac{1}{3}C_1(a-q)C_1(-bp)[v_1(\tilde{b}^{+}_{-p}b,r)+v_1(b\tilde{b}^{-}_{-p},r)+v_1(b_p^+\tilde{b},r)+v_1(\tilde{b}b_p^-,r)]\,|\,a\rangle\\
&\qquad+\sum_{b,p,l}\{A(-ab,-qp,l1)[v_l(\tilde{b}a,r)+v_l(b\tilde{a},r)]\,|\,b_p^-\rangle+A(ab,qp,l1)v_l(ba,r)\,|\,\tilde{b}^{-}_{-p}\rangle\\
&\qquad\qquad+(-1)^{j_b-j_p}A(-ap,-qb,l1)[v_l(\tilde{b}^{+}_{-p}a,r)+v_l(b_p^+\tilde{a},r)]\,|\,b\rangle\\
&\qquad\qquad+(-1)^{j_b-j_p}A(ap,qb,l1)v_l(b_p^+a,r)\,|\,\tilde{b}\rangle\}\ ,
\end{aligned}
\tag{3.20d}
$$

TABLE I. Valence energies (in a.u.) for the potentials employed in this work with and without first-order correlation corrections compared with experiment. First-order corrections vanish in the HF case. $5p^* = 5p_{1/2}$, $5p = p_{3/2}$, etc.

Atom			Tietz	Green	Norcross	HF	Expt.[a]
Rb	5*s*	0th	−0.154 14	−0.153 48	−0.153 45		
		1st	−0.008 85	0.022 58	0.011 96		
		Sum	−0.162 99	−0.130 90	−0.141 49	−0.139 29	−0.153 51
	$5p^*$	0th	−0.095 57	−0.096 15	−0.096 31		
		1st	−0.006 44	0.009 70	0.005 27		
		Sum	−0.102 00	−0.086 45	−0.091 04	−0.090 82	−0.096 19
	5*p*	0th	−0.093 98	−0.094 80	−0.094 94		
		1st	−0.007 12	0.008 98	0.004 64		
		Sum	−0.101 11	−0.085 82	−0.090 31	−0.089 99	−0.095 11
	6*s*	0th	−0.061 40	−0.062 15	−0.061 97		
		1st	−0.002 29	0.005 31	0.002 79		
		Sum	−0.063 70	−0.056 84	−0.059 18	−0.58 70	−0.061 77
	$6p^*$	0th	−0.045 05	−0.045 70	−0.045 56		
		1st	−0.002 12	0.003 20	0.001 58		
		Sum	−0.047 16	−0.042 51	−0.043 98	−0.043 89	−0.045 45
	6*p*	0th	−0.044 56	−0.045 26	−0.045 12		
		1st	−0.002 30	0.003 00	0.001 42		
		Sum	−0.046 86	−0.042 26	−0.043 70	−0.043 60	−0.045 10
	7*s*	0th	−0.033 45	−0.033 82	−0.033 72		
		1st	−0.000 92	0.002 11	0.001 09		
		Sum	−0.034 37	−0.031 72	−0.032 63	−0.032 44	−0.033 62
Cs	6*s*	0th	−0.143 43	−0.143 09	−0.143 01		
		1st	0.003 16	0.026 08	0.017 63		
		Sum	−0.140 27	−0.117 01	−0.125 38	−0.127 37	−0.143 10
	$6p^*$	0th	−0.092 47	−0.092 23	−0.092 50		
		1st	0.001 71	0.012 14	0.009 24		
		Sum	−0.090 77	−0.080 09	−0.083 26	−0.085 62	−0.092 17
	6*p*	0th	−0.088 92	−0.089 15	−0.089 28		
		1st	−0.000 13	0.010 25	0.007 42		
		Sum	−0.089 05	−0.078 90	−0.081 86	−0.083 79	−0.089 64
	7*s*	0th	−0.058 27	−0.059 01	−0.058 83		
		1st	0.000 46	0.006 07	0.004 19		
		Sum	−0.057 81	−0.052 95	−0.054 63	−0.055 19	−0.058 65
	$7p^*$	0th	−0.043 79	−0.044 24	−0.044 10		
		1st	0.000 26	0.003 91	0.002 78		
		Sum	−0.043 53	−0.040 33	−0.041 32	−0.042 02	−0.043 93
	7*p*	0th	−0.042 70	−0.043 23	−0.043 07		
		1st	−0.000 19	0.003 43	0.002 33		
		Sum	−0.042 89	−0.039 80	−0.040 74	−0.041 37	−0.043 10
	8*s*	0th	−0.032 13	−0.032 51	−0.032 40		
		1st	0.000 16	0.002 43	0.001 67		
		Sum	−0.031 97	−0.030 07	−0.030 73	−0.030 95	−0.032 30
Au	6*s*	0th	−0.370 79	−0.369 75	−0.377 98		
		1st	−0.014 42	0.088 79	0.105 06		
		Sum	−0.385 20	−0.280 96	−0.272 92	−0.274 61	−0.339 04
	$6p^*$	0th	−0.187 07	−0.171 33	−0.176 26		
		1st	0.001 22	0.040 32	0.049 22		
		Sum	−0.185 86	−0.131 00	−0.127 04	−0.133 79	−0.168 82
	6*p*	0th	−0.159 07	−0.144 23	−0.148 52		
		1st	0.000 24	0.023 20	0.030 32		
		Sum	−0.158 83	−0.121 03	−0.118 20	−0.121 62	−0.151 43
	7*s*	0th	−0.093 83	−0.092 67	−0.095 24		
		1st	0.001 98	0.007 54	0.011 22		
		Sum	−0.091 85	−0.085 13	−0.084 02	−0.083 22	−0.090 79
	$7p^*$	0th	−0.064 41	−0.063 13	−0.064 26		
		1st	0.000 67	0.005 76	0.007 77		
		Sum	−0.063 74	−0.057 37	−0.056 49	−0.056 44	−0.065 51

TABLE I. (*Continued*).

Atom			Tietz	Green	Norcross	HF	Expt.[a]
	$7p$	0th	−0.059 90	−0.058 34	−0.059 20		
		1st	0.000 19	0.004 42	0.005 97		
		Sum	−0.059 71	−0.053 92	−0.053 23	−0.053 35	−0.062 34
	$8s$	0th	−0.044 98	−0.044 75	−0.045 42		
		1st	0.000 56	0.002 47	0.003 38		
		Sum	−0.044 42	−0.042 28	−0.042 04	−0.041 58	−0.044 05
Tl	$6p^*$	0th	−0.224 54	−0.224 46	−0.224 46		
		1st	0.034 47	0.068 27	0.035 91		
		Sum	−0.190 07	−0.156 20	−0.188 56	−0.199 68	−0.224 46
	$6p$	0th	−0.183 20	−0.176 41	−0.180 03		
		1st	0.022 08	0.036 14	0.019 75		
		Sum	−0.161 12	−0.140 27	−0.160 28	−0.166 93	−0.188 96
	$7s$	0th	−0.101 91	−0.101 79	−0.105 78		
		1st	0.009 54	0.009 82	0.011 16		
		Sum	−0.092 37	−0.091 96	−0.094 61	−0.096 18	−0.103 82
	$7p^*$	0th	−0.069 32	−0.069 57	−0.071 11		
		1st	0.006 51	0.008 33	0.007 93		
		Sum	−0.062 82	−0.061 24	−0.063 18	−0.065 05	−0.068 82
	$7p$	0th	−0.063 91	−0.063 73	−0.064 81		
		1st	0.004 71	0.006 41	0.005 37		
		Sum	−0.059 20	−0.057 32	−0.059 44	−0.060 86	−0.064 26
	$8s$	0th	−0.047 55	−0.047 69	−0.048 51		
		1st	0.002 98	0.003 25	0.003 10		
		Sum	−0.044 57	−0.044 44	−0.045 41	−0.045 75	−0.047 92
	$8p^*$	0th	−0.036 26	−0.036 39	−0.036 84		
		1st	0.002 46	0.003 10	0.002 78		
		Sum	−0.033 80	−0.033 29	−0.034 07	−0.034 62	−0.035 98

[a] C. E. Moore, *Atomic Energy Levels*, Natl. Bur. Stand. Ref. Data Ser., Natl. Bur. Stand. (U.S.) Circ. No. 35 (U.S. GPO, Washington, D. C., 1971), Vol. III.

where

$$V'_{\mathrm{HF}}\,|a_q^{\pm}\rangle = -\sum_{b,l}(2j_b+1)[\Lambda_{-blq}v_l(\tilde{b}a_q^{\pm},r)\,|\,b\rangle + \Lambda_{blq}v_l(ba_q^{\pm},r)\,|\,\tilde{b}\rangle] \ . \tag{3.21}$$

E. EDM enhancement factors

In the presence of an external electric field $\mathbf{E}_{\mathrm{ex}}=E_{\mathrm{ex}}\hat{z}$ and a nonvanishing electron EDM d_e, the Dirac-Coulomb Hamiltonian becomes

$$\begin{aligned} H_{\mathrm{EDM}} &= H+H_1 \ , \\ H &= \sum_i(\boldsymbol{\alpha}_i\cdot\mathbf{p}_i+m\beta_i)+V \ , \\ H_I &= \sum_i(-eE_{\mathrm{ex}}z_i-d_e\gamma_0\boldsymbol{\Sigma}_i\cdot\mathbf{E}_T^i) \ , \end{aligned} \tag{3.22}$$

where

$$\begin{aligned} V &= \tfrac{1}{2}\alpha\sum_{i,j}\frac{1}{|\mathbf{r}_i-\mathbf{r}_j|}-\sum_i\frac{Z\alpha}{r_i} \ , \\ \mathbf{E}_T^i &= E_{\mathrm{ex}}\hat{z}_i-\frac{1}{e}\boldsymbol{\nabla}_iV \ . \end{aligned} \tag{3.23}$$

In the following we suppress the summation over i in H_1 for notational simplicity. The change in energy arising from H_1 that is linear in E_{ex} arises from both first and second orders of perturbation theory:

$$\begin{aligned} \Delta E = {} & -d_e\langle 0\,|\,\gamma_0\Sigma_3\,|\,0\rangle E_{\mathrm{ex}} \\ & -d_e\sum_n{}'\frac{1}{E_0-E_n}(\langle 0\,|\,z\,|\,n\rangle\langle n\,|\,\gamma_0\boldsymbol{\Sigma}\cdot\boldsymbol{\nabla}V\,|\,0\rangle \\ & +\langle 0\,|\,\gamma_0\boldsymbol{\Sigma}\cdot\boldsymbol{\nabla}V\,|\,n\rangle\langle n\,|\,z\,|\,0\rangle)E_{\mathrm{ex}} \ . \end{aligned} \tag{3.24}$$

In order to make the calculation parallel the parity-violating one, we use Eq. (3.22) to write

$$\gamma_0\boldsymbol{\Sigma}\cdot\boldsymbol{\nabla}V = i\left[\gamma_0\boldsymbol{\Sigma}\cdot\mathbf{p},H-\sum_i(\boldsymbol{\alpha}_i\cdot\mathbf{p}_i+m\beta_i)\right] \ . \tag{3.25}$$

The part of the commutator involving H cancels the energy denominator, allowing the sum over n to be completed (the excluded term $|\,n\rangle=|\,0\rangle$ gives a vanishing contribution). The resulting commutator precisely cancels the first term in Eq. (3.24). The presence of γ_0 in $\gamma_0\boldsymbol{\Sigma}\cdot\mathbf{p}$ is all that keeps the remaining part of the commutator from vanishing. Therefore, in the nonrelativistic limit the ef-

TABLE II. Reduced matrix elements (in a.u.) for allowed transitions with and without first-order correlation corrections for the different potential models employed here compared with experiment. Experimental values are obtained from measured oscillator strengths.

Atom	Transition		Tietz	Green	Norcross	HF	Expt.
Rb	$5s \rightarrow 5p^*$	0th	4.47	4.39	4.44	4.82	
		1st	−0.79	0.21	−0.15	−0.21	
		Sum	3.68	4.60	4.29	4.61	4.11(0.06)[a]
	$5s \rightarrow 5p$	0th	−6.28	−6.19	−6.25	−6.80	
		1st	1.05	−0.31	0.19	0.30	
		Sum	−5.23	−6.50	−6.07	−6.51	−5.90(0.07)[a]
Cs	$6s \rightarrow 6p^*$	0th	4.85	4.74	4.81	5.28	
		1st	−0.82	0.16	−0.22	−0.30	
		Sum	4.04	4.90	4.58	4.97	4.52(0.01)[b]
	$6s \rightarrow 6p$	0th	−6.77	−6.64	−6.72	−7.43	
		1st	1.00	−0.30	0.23	0.41	
		Sum	−5.77	−6.94	−6.50	−7.01	−6.36(0.01)[b]
Au	$6s \rightarrow 6p^*$	0th	2.41	2.31	2.30	2.71	
		1st	11.60	−1.02	−1.04	−0.49	
		Sum	14.01	1.29	1.26	2.22	1.83(0.09)[c]
	$6s \rightarrow 6p$	0th	−3.29	−3.11	−3.10	−3.70	
		1st	−0.67	1.32	1.35	0.63	
		Sum	−3.96	−1.79	−1.75	−3.07	−2.56(0.09)[c]
Tl	$6p^* \rightarrow 7s$	0th	−1.70	−1.64	−1.75	−2.05	
		1st	−0.25	−0.63	−0.24	0.14	
		Sum	−1.94	−2.28	−1.99	−1.91	−1.84(0.06)[d]
	$6p \rightarrow 7s$	0th	−3.52	−3.61	−3.72	−3.97	
		1st	0.06	−0.53	0.11	0.35	
		Sum	−3.46	−4.14	−3.61	−3.61	−3.29(0.09)[d]

[a] A. Gallagher and E. Lewis, Phys. Rev. A **10**, 231 (1974).

[b] L. Shabanova, Yu. Monakov, and A. Khlyustalov, Opt. Spectrosc. (USSR) **47**, 1 (1979).

[c] N. Penkin and I. Slavenas, Opt. Spectrosc. (USSR) **15**, 3 (1963).

[d] Obtained from N. P. Penkin and I. N. Shabanova, Opt. Spectrosc. (USSR) **14**, 87 (1963), and M. Norton and A. Gallagher, Phys. Rev. A **3**, 915 (1971). See J. Migdalek and W. E. Baylis, J. Phys. B **12**, 2595 (1979).

fect vanishes, in accordance with Schiff's theorem.[18] We now have

$$\Delta E = -2id_e E_{\mathrm{ex}} \sum_n \frac{1}{E_0 - E_n} (\langle 0 | z | n \rangle \langle n | \gamma_0 \gamma_5 \mathbf{p}^2 | 0 \rangle + \langle 0 | \gamma_0 \gamma_5 \mathbf{p}^2 | n \rangle \langle n | z | 0 \rangle) \tag{3.26}$$

which is the change of energy associated with the one-body Hamiltonian

$$h_W = 2i \frac{d_e}{e} \gamma_0 \gamma_5 \mathbf{p}^2 . \tag{3.27}$$

Once this equivalent one-body form is adopted, the calculation parallels the previous parity-violation one entirely, so the equations of Sec. III D apply also to this case, with the slight modification that $v_2 = v_1$ and $\omega = 0$. The reduction of the EDM interaction Hamiltonian to one-body form carried out above follows the procedure outlined by Sandars in his seminal work on atomic electric dipole moments.[19]

IV. TABULATION OF RESULTS

In the paragraphs below we give the results of numerical evaluation of the various atomic properties discussed in Sec. III. The lowest-order values in the present tables are in some cases different from the corresponding values given in I, since changes have been made both in the treatment of nuclear finite size and in the model potential parameters. The new parameters and nuclear charge distributions are presented in the Appendix along with a discussion of the importance of modeling core states when constructing model potentials.

In Table I we give the low-lying spectra of the four elements studied as calculated in each of the four potentials considered. The rather good agreement between the lowest-order model potential results and the experimental values is not surprising since these low-lying energies were used to determine the parameters for the potentials. By comparing with the corresponding HF results and with the first-order corrections it becomes evident that the relatively precise agreement between the zeroth-order spectrum and experiment achieved by fitting the experimental spectra is illusory. First-order perturbation theory intro-

TABLE III. Magnetic dipole hyperfine constants (A) in MHz calculated using various potentials. In the HF case, the first-order term includes core polarization corrections to all orders. The nuclear g factors used are the following: Rb, 0.541 208 (Ref. a); Cs, 0.737 7208 (Ref. a); Au, 0.098 772 (Ref. b); and Tl, 3.276 4268 (Ref. c).

Isotope	State	Term	Tietz	Green	Norcross	HF	Expt.
^{84}Rb	$5s$	0th	1098.0	1067.0	1051.0	643.9	
		1st	111.2	−386.4	−175.4	135.8	
		Sum	1209.2	680.6	875.6	779.7	1011.9108[d]
	$6s$	0th	252.8	255.0	253.2	172.0	
		1st	6.8	−55.1	−29.6	35.9	
		Sum	259.6	199.9	223.4	207.9	239.3(12)[e]
^{133}Cs	$6s$	0th	2405.0	2408.0	2346.0	1435.0	
		1st	9.6	−978.0	−469.9	289.2	
		Sum	2414.6	1430.0	1876.1	1724.2	2298.1579[f]
	$7s$	0th	565.1	576.0	569.1	393.9	
		1st	−11.3	−123.9	−69.0	80.1	
		Sum	553.8	452.1	500.1	474.0	546.3(30)[e]
^{197}Au	$6s$	0th	3036.0	3504.0	3550.0	2175.0	
		1st	663.3	−805.7	−912.7	251.2	
		Sum	3699.3	2698.3	2637.3	2426.2	3049.6601[g]
	$7s$	0th	333.0	320.6	335.1	266.1	
		1st	−20.8	−15.3	−27.6	29.1	
		Sum	312.3	305.3	307.5	295.2	
^{205}Tl	$6p^*$	0th	−23 488.0	−28 238.7	−25 931.0	−17 656.0	
		1st	4616.3	16 413.3	7166.6	−4315.2	
		Sum	−18 871.7	−11 825.4	−18 764.4	−21 971.2	−21 310.835[h]
	$7p^*$	0th	−2729.0	−2875.0	−3153.0	−1968.0	
		1st	1093.0	924.5	1370.0	−61.4	
		Sum	−1636.0	−1950.5	−1783.0	−2029.4	−2131(60)[i]

[a] C. W. White, W. M. Hughes, G. S. Hayne, and H. G. Robinson, Phys. Rev. **174**, 23 (1968); Phys. Rev. A **7**, 1178 (1973).

[b] A. Narath, Phys. Rev. **163**, 232 (1967); **175**, 696 (1968).

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duces corrections which modify the empirically determined unperturbed spectra, away from experiment and typically closer to the HF spectra. There are, of course, no first-order corrections in the HF case.

Our results for parity-conserving transition matrix elements are presented in Table II. Note that the experimental values given in the table, which are inferred from measured oscillator strengths and the experimentally determined transition frequencies, are relatively poorly known except for the case of Cs. This is then one case where a theoretical program capable of predictions of a few percent accuracy could provide guidance to experiments. At the present level, however, our theoretical predictions are accurate only at about the 20% level. The most successful model is the Norcross potential, in which the first-order corrections always take the lowest-order value in the direction of experiment: This feature is also present in the HF case, which in the Au and Tl cases is actually superior to the Norcross potential. The Tietz and Green potentials have more erratic behavior, but are fairly successful except for the $6s \rightarrow 6p_{1/2}$ transition in Au for the Tietz potential. The large first-order correction there arises from the $5d_{3/2} \rightarrow p_{1/2}$ excitation channel. Slight changes in the Tietz parameters lead to large changes in this correction. Particular attention will be paid to this transition in the ensuing second-order calculation to see if the cause of this unstable behavior is neglect of some important second-order effect.

Table III contains our hyperfine-splitting results. A notable feature is that the excited-state hyperfine constants are in general under better control than those of the ground state, with roughly 20% spreads reduced to 10%. A similar effect was noted for transition matrix elements. We attribute this effect to a decreased sensitivity to the structure of the core of the atom in excited states. This suggests that it may be of value, if experimentally feasible, to study weak-interaction effects in excited states rather than the ground state. Comparing the potentials, we see

TABLE IV. Parity-violating dipole matrix elements in units of $iea_0(Q_w/N)10^{-11}$ calculated in various models.

	Tietz	Green	Norcross	HF	Expt.	Others
Rubidium $5s \rightarrow 6s$, $A=85$						
0th	0.168	0.164	0.160	0.139		
1st	-0.056	-0.030	-0.029	-0.004		
Sum	0.112	0.134	0.131	0.135		
Cesium $6s \rightarrow 7s$, $A=133$						
0th	1.079	1.073	1.056	0.927	0.93(12)[a]	0.880[c]
1st	-0.325	-0.197	-0.200	-0.037	1.00(8)[b]	0.996[d]
Sum	0.754	0.876	0.856	0.890		0.972[e]
						1.06[f]
						0.89[g]
Gold $6s \rightarrow 7s$, $A=197$						
0th	5.18	5.69	5.59	6.85		
1st	-1.21	-2.79	-2.60	-1.13		
Sum	3.97	2.90	2.99	5.72		
Thallium $6p^* \rightarrow 7p^*$, $A=205$						
0th	-9.64	-10.19	-10.07	-10.01	$-7.8(1.4)$[h]	-8.73[i]
1st	0.86	-0.57	0.66	0.30		-6.83[j]
Sum	-8.78	-10.76	-9.41	-9.71		

[a] M. A. Bouchiat *et al.*, Phys. Lett. **134B**, 463 (1984); **117B**, 358 (1982). $Q_w = -68.6$, $\beta = 26.8$.

[b] Reference 2. $Q_w = -68.6$, $\beta = 27.3$.

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[h] P. Drell and E. D. Commins, Phys. Rev. A **32**, 2196 (1985); C. E. Tanner and E. D. Commins, Phys. Rev. Lett. **56**, 332 (1986). $Q_w = 110$, $\beta = 205$.

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[j] Reference 9.

again that HF always tends toward the experimental result, but because the lowest-order HF results are quite far off, the final agreement is only moderately good. In this case the Norcross potential tends to again have corrections of the correct sign, but of large magnitude, so that the final result "overshoots" the experimental value. The Tietz and Green potentials are again somewhat erratic, with the Green potential overshooting the experimental values significantly.

Parity-violating transition matrix elements are presented in Table IV. A striking feature of the predictions is that, if the Tietz potential is left out, the remaining three potentials agree within 10%. However, given the behavior in the preceding three tables, we do not wish to claim this as our level of accuracy and in particular would not want to infer a Weinberg angle from experiment with accuracy of under 20%. If, however, a second-order calculation were to give results for the first three tables consistent with experiment and one another at, say, the 5% level, and the results of this table were consistent with one another at this same level, we would then feel in a position to make statements about the agreement or disagreement with the Weinberg-Salam model at this level of accuracy.

As mentioned in the Introduction, there have been several calculations done on Cs and Tl using the HF potential. In the case of Cs, we have found excellent agreement with the first-order results of Dzuba *et al.*[8] and with the results of Mårtensson-Pendrill[7] when we use the nuclear distributions of these authors. However, we have serious discrepancies with the calculation of Das[10] already in the lowest-order calculation. The source of this discrepancy is not understood at this time. Similar remarks apply to Tl, where we have good agreement with a calculation of Mårtensson-Pendrill[20] and discrepancies already in lowest order with Das.[9]

Finally, in Table V we present our EDM enhancement factor calculations. The same general pattern of spreads of values is present here as in the previous cases. Here, however, because one is looking for a nonzero electron EDM rather than attempting a precision evaluation, our results can be interpreted in a more positive fashion, that is, that the atomic-physics uncertainties are certainly under 50%. In I, a very large correction to the EDM enhancement factor of Tl was noted in the HF case that made it seem possible that even such a relatively large uncertainty might not be attainable. However, it is seen that once first-order corrections are added in, a large cancellation takes place, and the HF result is consistent with the other potential results. A similar effect takes place in the parity calculation, but there the cancellation takes place

TABLE V. Electric dipole moment enhancement factors $R = D_{\text{atom}}/d_e$ with and without first-order correlation corrections for the various potential models employed here.

Atom	State		Tietz	Green	Norcross	HF	Other[a] (Unshielded)	Other[a] (Shielded)
Rb	5*s*	0th	34.0	29.7	30.0	26.6		
		1st	−17.0	−5.9	−8.0	−2.0		
		Sum	16.1	23.7	22.0	24.6	27.5	24
Cs	6*s*	0th	158.2	139.4	144.1	126.6		
		1st	−77.9	−33.5	−43.7	−11.7		
		Sum	80.3	106.0	100.4	114.9	133	119
Au	6*s*	0th	379.7	371.7	368.4	340.2		
		1st	−437.2	−240.9	−234.2	−90.3		
		Sum	−57.4	130.8	134.2	249.9		
Tl	6*p**	0th	−679.0	−689.0	−687.0	−1906.0		
		1st	177.0	83.0	125.0	865.0		
		Sum	−502.0	−607.0	−562.0	−1041.0		−700[b]

[a] P. G. H. Sandars, Phys. Lett. **22**, 290 (1966) (with and without shielding).
[b] P. G. H. Sandars and R. M. Sternheimer, Phys. Rev. A **11**, 473 (1975).

between different parts of the first-order calculation: This phenomenon was first observed by Mårtensson-Pendrill.[21]

V. SUMMARY

The most obvious conclusion from examination of our results is that many-body perturbation theory is quite sensitive and can produce 20% corrections. The problem is that the core is now being allowed to interact with the valence electron, and a poor description of a core state can result in large corrections, as perturbation theory is in some sense trying to adjust the core into its actual form. It is extremely difficult to simultaneously model all core states with a single potential: It is typical in adjusting parameters to see one set of core energies improve, while another set deteriorates. Table VI shows the core energies of Cs and how well different models do. Note that the unusually good description of the deep core states in the Hartree-Fock model is accompanied by very bad results for the outer core, which is related to the unusually poor lowest-order HF predictions. It can also be seen that the Norcross potential is unusually good, except that the very outer core is relatively poorly described. However, all of the models considered, after adjusting parameters as described in the Appendix, give qualitatively a correct picture of the core, so we expect that higher orders of perturbation theory will give progressively better results. We are particularly encouraged by the fact that the treatment

TABLE VI. Core energies for Cs in various potentials employed in this work (in a.u.).

State	Tietz	Green	Norcross	HF	Expt.[a]
1*s*	1362	1336	1316	1330	1322
2*s*	223	213	210	213	210
2*p**	214	202	199	199	196
2*p*	200	189	186	186	184
3*s*	45.4	44.3	44.4	46.4	44.7
3*p**	41.0	39.5	39.6	40.4	39.1
3*p*	38.1	36.9	37.0	37.9	36.6
3*d**	29.5	27.8	28.4	28.3	27.1
3*d*	28.8	27.3	27.8	27.8	26.6
4*s*	7.68	8.44	8.27	9.51	8.48
4*p**	5.94	6.65	6.47	7.45	6.33
4*p*	5.41	6.12	5.94	6.92	5.93
4*d**	2.37	3.07	2.96	3.49	2.90
4*d*	2.28	2.98	2.87	3.40	2.81
5*s*	0.876	1.09	0.994	1.49	0.83
5*p**	0.487	0.624	0.553	0.908	0.48
5*p*	0.438	0.555	0.493	0.840	0.42

[a] J. A. Bearden and A. F. Burr, Rev. Mod. Phys. **39**, 125 (1967).

of excited-state hyperfine splitting gives a set of results that are within 10% of each other and experiment: As discussed above, we attribute this situation to a lessened dependence on the core.

It is clearly necessary to go to second order to make predictions of atomic properties that are better than 10%. In some respects, while considerably more complicated, the calculations involved are similar to those described above. However, a novel feature of second-order calculations is the presence of double excitations, which can arise, for example, from the second term in Eq. (2.4). The sums over single excitations encountered in the present work were handled with Green's-function techniques that are well understood. Less is known of Green's-function techniques for application to these double excitations: While the nonrelativistic problem has been understood for some time,[22] there has been little work on the relativistic problem.[23] Sucher[24] has pointed out that straightforward extension of many-body perturbation theory to second order leads to terms that are not well defined which involve simultaneous excitation of positive and negative energy states; such terms must be explicitly removed. One approach that avoids this problem is direct saturation of the double sum with positive energy discrete and continuum states: This approach is used by Dzuba *et al.*[8] and Das.[9,10] We are exploring this approach, the two-particle Green's-function approach, and also a method proposed by Mohr[25] involving a contour integration over a product of one-particle Green's functions of complex energy. Whatever technique we employ, we expect that at the level of second-order perturbation theory, accuracies of a few percent should result. This expectation is based on the experience of Dzuba *et al.*,[8] who achieve this level of agreement with hyperfine constants, oscillator strengths, and valence energies in the Hartree-Fock treatment of Cs. What is not known is how the model potentials will behave. It is possible that at this level they will be far more accurate than Hartree-Fock; the general question of what potential leads to the most rapid convergence of many-body perturbation theory has never been addressed for heavy atoms.

Any complete second-order calculation is likely to involve extremely massive computation, due both to the large numbers of graphs to be evaluated and the difficulty of evaluating double-excitation sums. Nevertheless, we consider the problems addressed here of sufficient interest to justify the work: The atomic systems dealt with here are, even though the fundamental interaction is well known, nonperturbative systems that have not been dealt with in the systematic fashion that one- and two-electron atoms have. In addition, a test of the Weinberg-Salam theory at low energies in conjunction with the high-energy tests implicit in the accurate measurements of the W and Z_0 mass at European Organization for Nuclear Research (CERN) (Geneva, Switzerland) would provide a stringent test of unified theories of weak and electromagnetic interactions. The discovery alone of a nonvanishing atomic electric dipole moment would be of extreme importance to various exotic theoretical models of unification: However, given that discovery, an accurate calculation of the enhancement factor will be the only way of accurately determining the magnitude of d_e, which would be one of the fundamental properties of the electron, such as its magnetic anomaly, and which would certainly play as important a role in elementary particle physics.

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APPENDIX

The lowest-order values in the tables in Sec. IV are in some cases different from the corresponding values given in I, since changes have been made both in the treatment of nuclear finite size and in the model potential parameters.

In the case of parity-violating matrix elements for heavy elements we found a sensitivity at the several percent level to the shape of the nuclear charge distribution. Since much of the accurate data on nuclear charge distributions are parametrized in terms of a Fermi distribution,

$$\rho_{\mathrm{nuc}}(r)=\frac{\rho_0}{1+e^{4\ln 3(r-c)/t}}\ ,$$

we adopt such a form in the present work instead of the uniform distribution used in I. We choose the parameters as

$$c=\begin{cases}4.871\ \mathrm{fm}, & t=2.3\ \mathrm{fm}\ \text{ for } {}^{85}\mathrm{Rb}\\ 5.674\ \mathrm{fm}, & t=2.3\ \mathrm{fm}\ \text{ for } {}^{133}\mathrm{Cs}\\ 6.555\ \mathrm{fm}, & t=2.3\ \mathrm{fm}\ \text{ for } {}^{197}\mathrm{Au}\\ 6.617\ \mathrm{fm}, & t=2.3\ \mathrm{fm}\ \text{ for } {}^{205}\mathrm{Tl}\ .\end{cases}$$

This change causes virtually no differences with any of the lowest-order results given in I except the parity-violating matrix elements.

A second change, which leads to more significant differences with I, is that we choose the model potential parameters differently; the new parameters are listed in Table VII. In I we adjusted the model potential parameters to fit the ground state and the low-lying excitation spectra without regard to the core orbitals. The hyperfine constants determined from the resulting valence wave functions were generally in good agreement with experiment. The first-order perturbations considered here involve excitations of these core orbitals, and we find that a poor description of the more weakly bound core orbitals can lead to (often disasterously) large first-order correc-

TABLE VII. Model potential parameters.

Tietz potential: $V(r)=-\frac{\alpha}{r}\left[1+\frac{Z-1}{(1+tr)^2}e^{-\gamma r}\right]$

	t	γ
Rb	1.9530	0.2700
Cs	2.0453	0.2445
Au	2.4310	0.3500
Tl	2.3537	0.3895

Green potential: $V(r)=-\frac{\alpha}{r}\left[1+\frac{Z-1}{H(e^{r/d}-1)+1}\right]$

	H	d
Rb	3.481 14	0.785 51
Cs	4.469 10	0.896 65
Au	4.456 00	0.716 00
Tl	4.453 00	0.723 38

Norcross potential: $V(r)=V_{\mathrm{TF}}(\lambda,r)-\frac{\alpha}{2r^4}\alpha_d(1-e^{-(r/r_c)^6})$

$-\frac{\alpha}{2r^6}(\alpha_q-6\beta a_0)(1-e^{-(r/r_c)^{10}})$

	λ	r_c	α_d	α_q	β
Rb	0.993 21	3.2318	9.076	35.41	4.41
Cs	1.062 52	3.5461	15.81	86.40	7.91
Au	0.954 27	4.5439	12.4	103.00	4.13
Tl	0.963 82	4.553 43	24.00	108.00	8.69

tions which make it doubtful that the corresponding perturbation series converges. To remedy this situation we included the outer $4f$ and $5d$ energies in Tl and Au along with the valence energies in determining the model potential parameters. One consequence of this change is that the hyperfine constants in lowest order are not in as good agreement with experiment as in I. Nevertheless, the first-order perturbations are under better control.

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