Numerical test of the equality of the "length" and "velocity" forms of oscillator strengths for Li I in the Dirac-Fock approximation

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Transition matrix elements and oscillator strengths are calculated for various transitions between discrete levels of the neutral Li atom in the Dirac-Fock approximation, in the "length" and "velocity" forms, using nonlocal effective currents determined from quantum field theory. The matrix elements and oscillator strengths are shown to be numerically equal in the two forms, for this specific example, thereby verifying a previously given general proof of this equality.

I. INTRODUCTION

A resolution of the problem of the difference between the "length" and "velocity" forms of the transition amplitude for single photons in the long-wavelength limit, as calculated in the nonrelativistic Hartree-Fock (NRHF) approximation, has recently been proposed.[1] The usual way of calculating transitions in the HF approximation has been to evaluate the dipole matrix element in the length or velocity forms, using appropriate single-particle HF wave functions, having the same argument $\mathbf{r}$ for the initial and final states. Recent work[2,3] has suggested that if one takes a quantum-field-theory approach and imposes the necessary requirement of gauge invariance, the correct form of the transition matrix element involves the reduced vertex operator. The alteration corresponds to the replacement of a local current by a nonlocal effective current, $\tilde{\Lambda}(\mathbf{r},\mathbf{r}')$, the matrix element of which must be taken between appropriate single-particle HF wave functions, which now have different arguments $\mathbf{r}$ and $\mathbf{r}'$. This nonlocal current, or reduced vertex operator, satisfies a Dyson equation. The local current appears as an inhomogeneous term in this equation. The formalism also necessarily forces the use of $V(N-1)$ HF wave functions, determined by solving the HF equations for the core electrons without regard to the valence electron, and then solving the valence-electron HF equations in the frozen potential of the $N-1$ core electrons. The field-theory approach is merely a systematic way of taking into account the multiparticle nature of the many-electron atoms, even if in certain approximations one-particle wave functions (in the HF case) or two-particle wave functions [in the random-phase-approximation (RPA) case] make their appearance in calculations.

The nonlocal current in the NRHF approximation and the equations which determine it were generated in recent work[2,3] by Feldman and Fulton, and the gauge invariance of transition matrix elements for general gauges was also proven there. The formalism was applied to the dipole limit of the NRHF case in Ref. 1. If one takes the dipole limit in the initial stages of the analysis, the length and velocity forms are connected by a gauge transformation and the proofs of Refs. 2 and 3 apply. A more conservative approach is used in Ref. 1; the analysis is carried out in a fixed gauge, the radiation gauge. The dipole limit is taken only in the final stage and only the commutator

$$\left[\left[\frac{p^2}{2m}+V_{\rm nucl}(r)\right],\mathbf{r}\right]=-\frac{i}{m}\mathbf{p}\ ,$$

an algebraic identity, is used to prove the identity of the final expressions for the matrix elements in the length and velocity forms. The conclusion is the same in both approaches.

An extension of the treatment of Refs. 2 and 3 to the relativistic [in particular the relativistic Hartree-Fock or Dirac-Fock (DF)] case was considered in Ref. 4. Positron states now enter into the analysis and, since more of the full QED theory appears, renormalization plays a role. After rather extensive arguments, which also involve the neglect of terms of higher leading order in a $Z\alpha$ expansion than the ones which are retained, the structures of both the relativistic random-phase approximation (RRPA) and DF equations turn out to be the same as those for the corresponding nonrelativistic formalism. The specific proof of the equivalence of the length and velocity forms of transition matrix elements, using a commutator approach[1] is not considered for the DF case. Since a proof of this equivalence in terms of gauge invariance is given in Ref. 4, a repetition of the work of Ref. 1 for the relativistic case is superfluous.

Perhaps a more useful development of the general formalism considered in Refs. 1–4 is to illustrate its validity, particularly as far as the nonlocal effective currents generated from the DF approximation are concerned, by numerically evaluating transition matrix elements which arise in a specific case. Besides demonstrating the numer-

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ical equivalence of the results for the length and velocity forms in this specific case, the present paper has three other aims: (a) to attempt to bridge the "language barrier" between the somewhat abstract and field-theoretically motivated formalism of Refs. 1–4 and the language and notation customarily used by atomic physicists; (b) to demonstrate that the approach of Refs. 1–4 can be successfully employed in specific numerical calculations; (c) to generate new values for transition matrix elements, differing from those for *both* the length and velocity forms, using only local currents and DF wave functions for the initial and final states. (These values provide the lowest-order contributions in a sequence involving increasing numbers of additional correlations, the sum of which is finally to be compared to experiment.)

In the following section we first briefly define the expressions relevant to the subsequent numerical calculations and give the equations which must be solved numerically. We then apply the general analysis to the case of various $E1$ transitions of Li I, and list the numerical results for these transitions. A brief discussion concludes the main part of our paper.

Appendix A is initially devoted to the realization of aim (a) listed above: the "translation" of the formalism of Refs. 1–4 to the more familiar language of atomic physics. More important, the differential equation, the solution of which plays an essential role in the numerical evaluations, is derived from the field-theory formalism.

The partial differential equations derived in Appendix A and given in the main body of the paper are only the starting points for the numerical analysis. The angular-momentum parts must be explicitly dealt with, and coupled differential equations in a single radial variable (for functions which enter the evaluation of the transition matrix elements) must be obtained before the numerical calculations can actually be implemented. These differential equations in the radial variable are obtained in Appendix B.

II. SUMMARY OF RELEVANT FORMALISM AND NUMERICAL RESULTS FOR $E1$ PHOTON ABSORPTION IN Li I

The relevant one-photon transition matrix element for absorption of a photon in the DF approximation is given by

$$\mathcal{M}_{fi}(k)=(2\pi)^4\delta(e_f-e_i-k)M_{fi}(k) . \quad (1)$$

This equation is Eq. (2.1) of Ref. 1 and is valid for the relativistic dipole case, as well as the nonrelativistic dipole case.[1] The letters f (i) refer to the final (initial) atomic state, e_f and e_i are the corresponding DF one-particle energy levels, and k is the energy of the absorbed photon. (We take $\hbar=c=1$ in all equations.)

The matrix element $M_{fi}(k)$ of Eq. (1), with the energy conservation taken out, is a specific case of a more general matrix element, $M_{pq}(k)$, in the dipole approximation:

$$M_{pq}(k)=(2k)^{-1/2}\int u_p^\dagger(\mathbf{r}_1)d^3r_1\widetilde{\Lambda}(\mathbf{r}_1,\mathbf{r}_2;k)\times d^3r_2u_q(\mathbf{r}_2) , \quad (2)$$

where p and q label discrete DF states (among which are f and i), $(2k)^{-1/2}$ is the photon wave-function normalization, and $\widetilde{\Lambda}(\mathbf{r}_1,\mathbf{r}_2;k)$ is the nonlocal effective dipole current,[5] which depends on the *two* spatial variables, $\mathbf{r}_1$ and $\mathbf{r}_2$. The function $\widetilde{\Lambda}$ satisfies an inhomogeneous integral equation, Eq. (A6). This equation has different inhomogeneous terms, corresponding to the different local photon-electron interactions for the different "forms":

$$[\widetilde{\Lambda}(\mathbf{r}_1,\mathbf{r}_2;k)]^{(\text{form})}=\delta^{(3)}(\mathbf{r}_1-\mathbf{r}_2)\lambda_k^{(\text{form})}(\mathbf{r}_1) , \quad (3)$$

where (form) stands for (velocity) or (length), and

$$\lambda_k^{(\text{velocity})}(\mathbf{r}_1)=e\boldsymbol{\epsilon}_k\cdot\boldsymbol{\alpha} , \quad (4)$$

$$\lambda_k^{(\text{length})}(\mathbf{r}_1)=iek\boldsymbol{\epsilon}_k\cdot\mathbf{r}_1 \quad (5)$$

(with $\boldsymbol{\epsilon}_k$ the photon polarization vector), in the velocity and length forms of the dipole approximation, respectively. It is the velocity form which naturally arises if the dipole approximation is made in the radiation gauge. The length form can be made to appear in one of two ways: by a gauge transformation, carried out subsequent to the dipole approximation, following Lamb's point of view,[6,7] or by use of a commutator identity[1,3] connecting the two forms in both the nonrelativistic and relativistic cases. For the relativistic case, this commutator identity is

$$[h(\mathbf{r}),\mathbf{r}]=-i\boldsymbol{\alpha} ,$$
$$h(\mathbf{r})=\boldsymbol{\alpha}\cdot\mathbf{p}+m\beta+V_{\text{nucl}}(\mathbf{r}) . \quad (6)$$

The corresponding commutator in the nonrelativistic case is given in the introduction. In principle, $\mathcal{M}_{fi}$ of Eq. (1) and $M_{pq},\widetilde{\Lambda}$ of Eq. (2) should carry separate labels, distinguishing the length and velocity forms. In point of fact, *both* forms, which involve *different* inhomogeneous terms, Eqs. (4) and (5), yield the same $\mathcal{M}_{fi}$, M_{pq}, and $\widetilde{\Lambda}$, and thus these functions require no distinguishing labels.

The transition matrix elements corresponding to the local currents, described by Eqs. (3)–(5) are

$$[m_{pq}(k)]^{(\text{form})}=\int d^3r\, u_p^\dagger(\mathbf{r})\lambda_k^{(\text{form})}(\mathbf{r})u_q(\mathbf{r}) . \quad (7)$$

These matrix elements are the ones normally called the DF matrix elements, if $u_p^\dagger(\mathbf{r})$ and $u_q(\mathbf{r})$ are DF states. However, the matrix elements which naturally arise from DF theory contain contributions from *nonlocal* as well as local effective currents,[2–4] where the latter are generated from the *local* field-theoretical lepton-photon interaction. The sum of these local and nonlocal currents is $\widetilde{\Lambda}$ of Eq. (2). As previously stated, $\widetilde{\Lambda}$ satisfies Eq. (A6), which is the dipole approximation equivalent of Eq. (3.44) of Ref. 4. When this equation for $\widetilde{\Lambda}$ is used in Eq. (2), and the notation is translated from the abstract to the more concrete form, one obtains

$$M_{fi}(k)=[m_{fi}(k)]^{(\text{form})}+\sum\!\!\!\!\!\!\int_{a,j}\left[\frac{M_{aj}(k)V_{fi}^{ja}}{e_a-e_j-k}+\frac{V_{fi}^{aj}M_{ja}(k)}{e_a-e_j+k}\right] , \quad (8)$$

and

$$M_{aj}(k)=[m_{aj}(k)]^{(\mathrm{form})} + \sum\!\!\!\!\!\!\int_{b,l} \left[\frac{M_{bl}(k)V_{aj}^{lb}}{e_b-e_l-k} + \frac{V_{aj}^{bl}M_{lb}(k)}{e_b-e_l+k} \right] , \tag{9a}$$

$$M_{ja}(k)=[m_{ja}(k)]^{(\mathrm{form})} + \sum\!\!\!\!\!\!\int_{b,l} \left[\frac{M_{bl}(k)V_{ja}^{lb}}{e_b-e_l-k} + \frac{V_{ja}^{bl}M_{lb}(k)}{e_b-e_l+k} \right] . \tag{9b}$$

The indices i, f, j, and l above refer to DF electron states outside of the frozen relaxed core. [We calculate in the $V(N-1)$ DF approximation.] This core for Li I consists of two $1s$ electrons. The indices a and b refer to the core electron states α and to positron states $\bar{n}$, i.e.,

$$\{a\}=\{\alpha,\bar{n}\} , \qquad (e_a-e_j\mp k)\,|_{a=\bar{n}} \equiv -(e_{\bar{n}}+e_j\pm k)\approx -2m, \quad e_{\bar{n}}>0 . \tag{10}$$

The symbol $\sum\!\!\!\!\!\!\int$ indicates summation over discrete and integration over continuum states. The expressions V_{pq}^{st} which appear in Eqs. (8) and (9) above are overlap integrals of the electron-electron (e-e) Coulomb interaction with DF states:

$$V_{pq}^{st} \equiv \int V(|\mathbf{r}-\mathbf{r}'|)u_p^\dagger(\mathbf{r})u_s^\dagger(\mathbf{r}') \times[u_q(\mathbf{r})u_t(\mathbf{r}')-u_t(\mathbf{r})u_q(\mathbf{r}')]d^3r\,d^3r' , \qquad V(|\mathbf{r}-\mathbf{r}'|)=e^2/|\mathbf{r}-\mathbf{r}'| . \tag{11}$$

The first term in the curly brackets is the direct and the second the exchange contribution; $V(|\mathbf{r}-\mathbf{r}'|)$ is the e-e Coulomb interaction.

Equation (8) and the equations for M_{aj} and M_{ja} [Eqs. (9a) and (9b)] are useful to make the structure of the theory clear and also serve to point to possible approximations to be made in the numerical analysis. However, they are not the actual starting points for that analysis. As far as structure is concerned, we note that Eqs. (9a) and (9b) are a closed set of linear algebraic equations for the core-electron (or positron) to valence-electron transitions. The solution of these equations must then be substituted in Eq. (8), which can give valence-electron—valence-electron transitions.

A more important issue is the question of positron states. These states do not appear in the DF equations[4] for e_α, e_i, and the corresponding wave functions for two reasons: Renormalization effectively serves to take into account some of their contributions, and the remaining finite terms are of higher leading order than the contributions of the usual DF potential and are neglected.[4] It is thus consistent to neglect such contributions in Eqs. (8) and (9) (for the matrix elements) as well. One can see directly from the structure of Eqs. (8) and (9) that these contributions are negligible, since energy denominators from positron terms will be large [of $O(2m)$ compared to $O(m\alpha^2)$] and overlap integrals V_{pq}^{st} will be small (since they will involve overlaps between discrete and continuum states). Indeed, it is apparent from more recent work on atoms of much higher[8,9] Z than Li I that, not only positron states, but deeper-lying core-electron states can be neglected in numerical calculations. In other words, since small overlap integrals and large energy denominators are already seen to lead to negligible effects for deep-lying core-electrons, they are even more likely to be negligible for positron states.[10] We will therefore, at a minimum, here and henceforth neglect the positron states $\bar{n}$ and let

$$\{a\}\approx\{\alpha\} . \tag{12}$$

Equations (8) and (9) are inconvenient as a starting point for numerical calculations. They can be recast in an alternative and equivalent form which is much better suited to calculations. The details of this reordering are given in Appendix A. The transition matrix element M_{fi} (with positron states consistently neglected) is given by

$$M_{fi}(k)=[m_{fi}(k)]^{(\mathrm{form})}+(2k)^{-1/2}\sum_\alpha \int d^3r\,[u_\alpha^\dagger(\mathbf{r})\lambda_k^{(\mathrm{form})}(\mathbf{r})F_{\alpha-}^{(\mathrm{form})}(\mathbf{r})+F_{\alpha+}^{(\mathrm{form})\dagger}(\mathbf{r})\lambda_k^{(\mathrm{form})}(\mathbf{r})u_\alpha(\mathbf{r})] , \tag{13}$$

$$(\mathrm{form})\equiv(\mathrm{length})\ \text{or}\ (\mathrm{velocity}) .$$

The functions $F_{\alpha\pm}^{(\mathrm{form})}$ satisfy partial integro-differential equations, as follows:

$$[h(r)\mp k-e_\alpha]F_{\alpha\pm}(\mathbf{r})+\int V_{\mathrm{DF}}(\mathbf{r},\mathbf{r}')F_{\alpha\pm}(\mathbf{r}')d^3r'=-\int Q(\mathbf{r},\mathbf{r}')([\mathscr{o}_\pm+\mathscr{Y}_\pm]u_\alpha)(\mathbf{r}')d^3r' . \tag{14}$$

The inhomogeneous terms $[\mathscr{o}_\pm u_\alpha](\mathbf{r})$ are defined as

$$[\mathscr{o}_+u_\alpha](\mathbf{r})\equiv\int V(|\mathbf{r}-\mathbf{x}|)u_i^\dagger(\mathbf{x})[u_f(\mathbf{x})u_\alpha(\mathbf{r})-u_f(\mathbf{r})u_\alpha(\mathbf{x})]d^3x , \tag{15}$$

and

$$[\mathscr{o}_-u_\alpha](\mathbf{r})\equiv[\mathscr{o}_+u_\alpha](\mathbf{r}) \ \text{ with } f\leftrightarrow i . \tag{16}$$

The homogeneous terms $[\mathscr{Y}_\pm u_\alpha](\mathbf{r})$ are given by

$$[\mathscr{Y}_\pm u_\alpha](\mathbf{r})\equiv\sum_\beta \int V(|\mathbf{r}-\mathbf{x}|)\{u_\beta^\dagger(\mathbf{x})[F_{\beta\pm}(\mathbf{x})u_\alpha(\mathbf{r})-F_{\beta\pm}(\mathbf{r})u_\alpha(\mathbf{x})]+F_{\beta\mp}^\dagger(\mathbf{x})[u_\beta(\mathbf{x})u_\alpha(\mathbf{r})-u_\beta(\mathbf{r})u_\alpha(\mathbf{x})]\} . \tag{17}$$

The remaining expressions appearing in Eq. (14) are defined as follows: k is the photon energy, e_α the DF energy eigenvalue for the electron-core states $\{\alpha\}$, and $Q(\mathbf{r},\mathbf{r}')$ is a projection operator to electron states outside of the frozen core,

$$Q(\mathbf{r},\mathbf{r}')\equiv\sum\!\!\!\!\!\!\int_j u_j(\mathbf{r})u_j^\dagger(\mathbf{r}') . \tag{18}$$

The Hamiltonian $h(\mathbf{r})$ is local, excludes electron-electron interactions, and is defined in Eq. (6). Finally, $V_{\mathrm{DF}}(\mathbf{r},\mathbf{r}')$ is the DF potential,

$$V_{\mathrm{DF}}(\mathbf{r},\mathbf{r}')=\sum_{\gamma}\left[\delta^{(3)}(\mathbf{r}-\mathbf{r}')\int V(|\mathbf{r}-\mathbf{x}|)u_{\gamma}(\mathbf{x})u_{\gamma}^{\dagger}(\mathbf{x})d^3x - V(|\mathbf{r}-\mathbf{r}'|)u_{\gamma}(\mathbf{r})u_{\gamma}^{\dagger}(\mathbf{r}')\right] . \tag{19}$$

The orthogonality conditions

$$\int u_{\beta}^{\dagger}(\mathbf{r})F_{\alpha\pm}(\mathbf{r})d^3r=0 \tag{20}$$

immediately follow as a consequence of Eq. (14).

We note that Eqs. (13) and (14) are analogous to (8) and (9), respectively. Equation (14) must be solved and the solutions substituted into Eq. (13) to obtain M_{fi}, just as Eqs. (9) must be solved and the solutions substituted into (8) to obtain M_{fi}.

It is instructive to compare and contrast Eq. (14) above with the corresponding equation in the relativistic random-phase approximation[11] (RRPA). Equations (14) and (17) correspond to Eqs. (6) and (5) of Ref. 11, respectively, with the present $F_{\alpha\pm}(\mathbf{r})$ replaced by $w_{\alpha\pm}(\mathbf{r})$ of Ref. 11. (The label α, indicating core orbitals, corresponds to the label i of Ref. 11.) However, the inhomogeneous terms $[\nu_{\pm}u_{\alpha}](\mathbf{r})$ of our Eqs. (15) and (16) are absent in Eq. (6) of Ref. 11. Our Eq. (14) is an inhomogeneous equation, with k being an input parameter. Equation (6) of Ref. 11 is a homogeneous equation, and is an eigenvalue equation for the eigenvalue ω, which thus plays an entirely different role from k. The DF problem which underlies our Eq. (14) involves the $V(N-1)$ DF potential, while the DF problem underlying Eq. (6) of Ref. 11 does not. Finally, the projection operator $Q(\mathbf{r},\mathbf{r}')$ is implicit in Ref. 11 but is explicitly displayed in the present work. Thus, the orthogonality condition Eq. (20) is a consequence of Eq. (14) in the present work, while the corresponding orthogonality condition [Eq. (7) of Ref. 11] is a constraint on $w_{\alpha\pm}$, the satisfaction of which is assured by introducing Lagrange multipliers in Eq. (6) of Ref. 11. No such Lagrange multipliers appear either in Eq. (14), or in the homogeneous RPA and RRPA eigenvalue equations [which also contain $Q(\mathbf{r},\mathbf{r}')$] which arise in the analysis[12] of Refs. 3 and 4 and precisely correspond to Eq. (6) of Ref. 11.

Let us leave the comparison with the RRPA and return to our main argument. Equations (14)—(17) are still not in suitable form for the final computation. To achieve this form, it is necessary to take angular momentum into account specifically and to convert Eq. (14) to an integro-differential equation in a single radial variable. This task is carried out in a manner analogous to that of Ref. 11, and the results, together with the relevant definitions, are given in Appendix B.

The formalism outlined above and in Appendix B has been applied to numerical calculations for the case of photon absorption between various bound states of Li I. The values of $m_{fi}^{(\mathrm{length})}$ and $m_{fi}^{(\mathrm{velocity})}$ are given in the third and fourth columns of Table I, and those of the form-independent M_{fi} in the fifth column. The corresponding oscillator strengths are listed in the sixth, seventh, and eighth columns. The results of some other calculations are quoted for purposes of comparison in the last two columns. We would like to emphasize that the equality of the results of the calculation in the length and velocity forms is not assumed but numerically demonstrated for every transition considered. As an illustration, let us look in more detail at the matrix element entries in the first row of Table I. The customarily calculated local DF contributions [from the first term on the right-hand side of Eq. (13)] yield, for the $2s_{1/2}\rightarrow 2p_{1/2}$ transition, to the limit of accuracy of the computation,

$$m_{fi}^{(\mathrm{length})}=3.364\,367\,3 , \tag{21}$$

and

$$m_{fi}^{(\mathrm{velocity})}=3.430\,127\,2 . \tag{22}$$

The contributions of the new, nonlocal, part of the effective current are

$$\Delta M_{fi}^{(\mathrm{length})}=-0.013\,886\,5 , \tag{23}$$

and

$$\Delta M_{fi}^{(\mathrm{velocity})}=-0.079\,646\,9 . \tag{24}$$

The length and velocity forms of both the local and nonlocal contributions differ, but in such a way as to compensate for each other, so that the final matrix elements are

$$M_{fi}^{(\mathrm{length})}=3.350\,480\,8 , \tag{25}$$

and

$$M_{fi}^{(\mathrm{velocity})}=3.350\,480\,3 . \tag{26}$$

The numerical results (25) and (26) agree to about one part in 10^7, the accuracy of the computation, and make the separate labeling of M_{fi} by "form" irrelevant. The indications length and velocity are therefore dropped in Table I, and the numerical results are also rounded to four significant figures, since the greater accuracy is not needed.

III. DISCUSSION

As both the general proofs of Refs. 1—4 and the specific numerical calculations for Li I in the present work indicate, the argument between advocates of the length and velocity forms of the Hartree-Fock transition matrix elements is resolved by the statement: If the conventional Hartree-Fock form, with local current, is used, *neither* form is correct; if a nonlocal current, generated from a local field-theoretical photon-electron interaction in a generalized Hartree-Fock scheme, is used, *both* forms are correct and can be shown to lead both formally and numerically to the same result. We must reemphasize that the numerical values of the "form-independent" results are in general different from both the length and velocity forms of the conventional HF results. The fact that, in

TABLE I. Summary of dipole transition calculations for Li I. $m_{fi}^{(\text{length})}$ and $m_{fi}^{(\text{velocity})}$ designate reduced dipole matrix elements calculated in the $V(N-1)$ Hartree-Fock approximation using the length and velocity forms for the dipole operator, while M_{fi} designates the present form-independent result. $f^{(\text{length})}$, $f^{(\text{velocity})}$, and f_{nonlocal} are the corresponding oscillator strengths. All quantities are given in atomic units.

Transition	Energy	$m_{fi}^{(\text{length})}$	$m_{fi}^{(\text{velocity})}$	M_{fi}	$f^{(\text{length})}$	$f^{(\text{velocity})}$	f_{nonlocal}	f_{other}	
$2s_{1/2}\rightarrow 2p_{1/2}$	0.0677	3.364	3.430	3.350	0.255	0.265	0.253	0.255[a]	0.264[a]
$2s_{1/2}\rightarrow 2p_{3/2}$	0.0677	4.758	4.851	4.738	0.511	0.531	0.507	0.511[a]	0.524[a]
$2s_{1/2}\rightarrow$sum					0.766	0.796	0.760	0.762[b]	0.753[c]
								0.750[d]	0.759[e]
								0.758[f]	
$2s_{1/2}\rightarrow 3p_{1/2}$	0.1396	0.1551	0.1368	0.1611	0.001 12	0.000 87	0.001 21	0.001[a]	
$2s_{1/2}\rightarrow 3p_{3/2}$	0.1396	0.2193	0.1934	0.2277	0.002 24	0.001 74	0.002 41	0.002[a]	
$2s_{1/2}\rightarrow$sum					0.003 36	0.002 61	0.003 62	0.003 69[b]	0.004 54[d]
								0.004 88[e]	0.004 07[f]
$2s_{1/2}\rightarrow 4p_{1/2}$	0.1645	0.1458	0.1356	0.1494	0.001 17	0.001 01	0.001 22		
$2s_{1/2}\rightarrow 4p_{3/2}$	0.1645	0.2062	0.1917	0.2113	0.002 33	0.002 02	0.002 45		
$2s_{1/2}\rightarrow$sum					0.003 50	0.003 03	0.003 67	0.003 71[b]	0.004 16[d]
								0.004 34[e]	0.003 86[f]
$2s_{1/2}\rightarrow 5p_{1/2}$	0.1760	0.1106	0.1037	0.1131	0.000 72	0.000 63	0.000 75		
$2s_{1/2}\rightarrow 5p_{3/2}$	0.1760	0.1564	0.1467	0.1599	0.001 43	0.001 26	0.001 50		
$2s_{1/2}\rightarrow$sum					0.002 15	0.001 89	0.002 25	0.002 29[b]	
$2p_{1/2}\rightarrow 3d_{3/2}$	0.0731	5.182	5.180	5.178	0.654	0.654	0.653	0.654[a]	0.653[a]
$2p_{3/2}\rightarrow 3d_{3/2}$	0.0731	2.318	2.317	2.316	0.0654	0.0654	0.0653	0.065[a]	
$2p_{3/2}\rightarrow 3d_{5/2}$	0.0731	6.953	6.950	6.947	0.589	0.588	0.588	0.588[a]	
$2p_{3/2}\rightarrow$sum					0.654	0.654	0.653		

[a]Reference 17, Dirac-Hartree-Fock.
[b]Reference 18, many-body perturbation theory.
[c]Reference 19, basis set expansion.
[d]Reference 20, model potential.
[e]Reference 21, many-body perturbation theory.
[f]Reference 22, random-phase approximation.

the present case of Li I, the length form is closer to the form-independent result than is the velocity form presumably has no deeper significance. It could well be an accident of the particular example considered.

The successful achievement of form independence of transition matrix elements is required for the theory to be consistent, but is not *ipso facto* a guarantee of a good fit to experiment. In fact, accurate experiments[13] for the $2s_{1/2}\rightarrow 2p_{1/2}$ transition in Li I give a value

$$f_{(\text{expt})}^{(2s_{1/2}\rightarrow 2p_{1/2})}=0.2472\pm 0.0004 \tag{27}$$

for the oscillator strength, compared to our result (see the first row, the eighth column of Table I, with an additional significant figure given below),

$$f_{\text{nonlocal}}^{(2s_{1/2}\rightarrow 2p_{1/2})}=0.2533\ . \tag{28}$$

While the inclusion of the nonlocal contributions improves the conventional HF results, this improvement is not sufficient to lead to agreement with experiment. Presumably, additional correlations must be taken into account to improve theoretical results. The advantage of the present approach is that such additional correlations can be systematically included in the theory[3] and form independence is maintained at each successive level of approximation.

The new feature of the present analysis—the appearance of a nonlocal part to the current which enters the evaluation of transition matrix elements in the terms containing $F_{\alpha\pm}^{(\text{form})}$ in Eq. (13)—does not significantly change the numerical result due to the dominant $m_{fi}^{(\text{form})}$ term, the conventional HF result, for the particular case of Li I. Even though this contribution is small, it is numerically significant, in view of the accuracy of the calculation. It can well be much larger, relative to m_{fi}, for different atoms.[9] Somewhat surprisingly, the form-independent result does not lie between $m_{fi}^{(\text{length})}$ and $m_{fi}^{(\text{velocity})}$, as has been on occasion conjectured.[1] This again need not be inevitably so in all cases, and thus no special significance should be attached to it.

Finally, we would like to reemphasize the difference between the present approach, which should be called HF (even though it is not the conventional HF approach), and the RPA approach. As we said in Sec. II, on cursory view there are similarities between the equations which arise in the two approaches.[11,14,15] Indeed, if one considers Feynman diagrams generated by expansions in powers of the *e-e* Coulomb interaction, there is a topological equivalence between corresponding Feynman diagrams in the two approaches. The distinct origin of the two formalisms is made clear in Ref. 3. The HF formalism for the transition matrix is generated from the two-point function (the one-particle electron Green's function, $G^{(1)}$), from which the three-point function for the nonlocal effective current (irreducible vertex, $\tilde{\Lambda}$) is gotten by insertion of a photon line in the $G^{(1)}$ expansion (in powers of the *e-e* Coulomb potential) in all possible ways. This results in the *inhomogeneous* equation for the nonlocal ef-

fective current, given in the dipole approximation in the present work. The RPA formalism for the transition matrix is generated from the four-point function (the two-particle electron Green's function, $G^{(2)}$). The matrix element is expressible in terms of the Bethe-Salpeter wave function defined from $G^{(2)}$. This wave function satisfies a *homogeneous* equation. The structures of the kernel appearing in the homogeneous part of the HF equation for the effective current and the kernel of the Bethe-Salpeter equation, leading to the RPA approximation, are very similar, but not identical. As stated above, the present paper uses the HF rather than the RPA approach.

Although the present work has been limited to the case of Li I, applications to other one-valence-electron atoms have been carried out and lead to similar results. Applications of this method to study transitions in atoms with more than one valence electron, and to calculate photoionization amplitudes in one-valence-electron atoms are underway.

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APPENDIX A: RELATION OF FIELD-THEORY FORMALISM TO THE COMPUTATIONS

The purpose of this appendix is twofold: First, we "translate" the somewhat abstract notation of Refs. 3 and 4, which is a natural extension of the Dirac bra-ket notation, into the more familiar language of wave functions in coordinate space and integro-differential equations; secondly, we take advantage of the greater structural transparency of the abstract formalism (as was done in Refs. 3 and 4) to obtain the key equations relevant to the numerical computations [Eqs. (14)–(17)]. These two activities will be carried on in tandem.

Equation (2) in the more abstract notation is

$$M_{pq}(k)=(2k)^{-1/2}\left\langle \begin{matrix} p \\ q \end{matrix} \middle| \widetilde{\Lambda}_k \right\rangle , \tag{A1}$$

where

$$\left\langle \begin{matrix} p \\ q \end{matrix} \middle| \widetilde{\Lambda}_k \right\rangle = \int \left\langle \begin{matrix} p \\ q \end{matrix} \middle| \begin{matrix} \mathbf{r}_1 \\ \mathbf{r}_2 \end{matrix} \right\rangle d\mathbf{r}_1\, d\mathbf{r}_2 \left\langle \begin{matrix} \mathbf{r}_1 \\ \mathbf{r}_2 \end{matrix} \middle| \widetilde{\Lambda}_k \right\rangle . \tag{A2}$$

The expressions appearing in (A2) can be translated into more conventional notation, as follows:

$$\left\langle \begin{matrix} p \\ q \end{matrix} \middle| \begin{matrix} \mathbf{r}_1 \\ \mathbf{r}_2 \end{matrix} \right\rangle \equiv \langle p \mid \mathbf{r}_1 \rangle \langle q \mid \mathbf{r}_2 \rangle^* = \langle p \mid \mathbf{r}_1 \rangle \langle \mathbf{r}_2 \mid q \rangle , \tag{A3}$$

and

$$\left\langle \begin{matrix} \mathbf{r}_1 \\ \mathbf{r}_2 \end{matrix} \middle| \widetilde{\Lambda}_k \right\rangle \equiv \langle \mathbf{r}_1 \mid \widetilde{\Lambda}_k \mid \mathbf{r}_2 \rangle = \widetilde{\Lambda}(\mathbf{r}_1,\mathbf{r}_2;k) . \tag{A4}$$

The first form of (A4) is in a "one-sided," the second in a "two-sided" Dirac notation, and the last form is that given in Eq. (2). The standard Dirac notation is used on the right-hand side of (A3), i.e.,

$$\begin{aligned} \langle \mathbf{r}_2 \mid q \rangle &\equiv u_q(\mathbf{r}_2) , \\ \langle p \mid \mathbf{r}_1 \rangle &\equiv u_p^\dagger(\mathbf{r}_1) . \end{aligned} \tag{A5}$$

The equation satisfied by $\mid \widetilde{\Lambda}_k \rangle$ is the dipole equivalent of that for $\mid \overline{\Lambda}_k \rangle$ [Eq. (3.44) of Ref. 4—cf. also the nonrelativistic Eq. (3.38) of Ref. 3],

$$\mid \widetilde{\Lambda}_k \rangle = \mid \lambda_k^{(\mathrm{form})} \rangle + \mathscr{U} \frac{R}{k-\mathscr{H}} \mid \widetilde{\Lambda}_k \rangle , \tag{A6}$$

where

$$\left\langle \begin{matrix} \mathbf{r}_1 \\ \mathbf{r}_2 \end{matrix} \middle| \lambda_k^{(\mathrm{form})} \right\rangle = \langle \mathbf{r}_1 \mid \lambda_k^{(\mathrm{form})} \mid \mathbf{r}_2 \rangle = [\bar{\lambda}(\mathbf{r}_1,\mathbf{r}_2;k)]^{(\mathrm{form})} , \tag{A7}$$

and $\lambda_k^{(\mathrm{form})}, \bar{\lambda}^{(\mathrm{form})}$ are defined in Eqs. (4) and (5), and (3), respectively. The other symbols appearing in (A6) are defined as follows [cf. Eqs. (3.18) of Ref. 3 and (3.40) and (3.41) of Ref. 4]:

$$\left\langle \begin{matrix} \mathbf{r}_1 \\ \mathbf{r}_2 \end{matrix} \middle| \mathscr{U} \middle| \begin{matrix} \mathbf{r}_3 \\ \mathbf{r}_4 \end{matrix} \right\rangle = \delta^{(3)}(\mathbf{r}_1-\mathbf{r}_3)\delta^{(3)}(\mathbf{r}_2-\mathbf{r}_4)V(\mid \mathbf{r}_1-\mathbf{r}_2 \mid) - \delta^{(3)}(\mathbf{r}_1-\mathbf{r}_2)\delta^{(3)}(\mathbf{r}_3-\mathbf{r}_4)V(\mid \mathbf{r}_1-\mathbf{r}_3 \mid) , \tag{A8}$$

$$R = (P_U+\bar{Q}_U)Q_L - Q_U(P_L+\bar{Q}_L) , \tag{A9}$$

$$\mathscr{H} = H_U - H_L , \tag{A10}$$

where the subscript $U(L)$ indicates operations in the upper (lower) index space of the one-sided Dirac notation; H is the DF Hamiltonian [cf. Eqs. (6) and (19)]

$$H = h + V_{\mathrm{DF}} . \tag{A11}$$

R is a combination of projection operators, P, Q, and $\bar{Q}$ [cf. Eqs. (3.11) and (3.20) of Ref. 4], where

$$P = \sum_\alpha P_\alpha , \quad P_\alpha = \mid \alpha \rangle \langle \alpha \mid , \tag{A12}$$

$$Q = \sum_j Q_j , \quad Q_j = \mid j \rangle \langle j \mid , \tag{A13}$$

$$\bar{Q} = \sum_{\bar{n}} \bar{Q}_{\bar{n}} , \quad \bar{Q}_{\bar{n}} = \mid \bar{n} \rangle \langle \bar{n} \mid . \tag{A14}$$

In (A12)–(A14), α or j labels electron states (inside or outside the core, respectively) and $\bar{n}$ labels positron states. The completeness of states leads to the identity [Eq. (3.21), Ref. 4]

$$P+Q+\bar{Q}=\mathbb{1}\ , \tag{A15}$$

and, thus, to the alternative form of R:

$$R=(P_U+\bar{Q}_U)\mathbb{1}_L-\mathbb{1}_U(P_L+\bar{Q}_L)\approx P_U\mathbb{1}_L-\mathbb{1}_U P_L\ , \tag{A16}$$

where the last, approximate, form arises when positron states are neglected.

To illustrate the translation procedure, consider (A6) in the coordinate basis. We have, inserting complete sets of state on the right-hand side of (A6),

$$\left\langle \begin{matrix}\mathbf{r}_1\\ \mathbf{r}_2\end{matrix}\middle|\tilde{\Lambda}_k\right\rangle=\left\langle \begin{matrix}\mathbf{r}_1\\ \mathbf{r}_2\end{matrix}\middle|\lambda_k^{(\mathrm{form})}\right\rangle+\int\left\langle \begin{matrix}\mathbf{r}_1\\ \mathbf{r}_2\end{matrix}\middle|\mathscr{U}\middle|\begin{matrix}\mathbf{r}_3\\ \mathbf{r}_4\end{matrix}\right\rangle d\mathbf{r}_3\,d\mathbf{r}_4\left\langle \begin{matrix}\mathbf{r}_3\\ \mathbf{r}_4\end{matrix}\middle|\frac{R}{k-\mathscr{H}}\middle|\begin{matrix}\mathbf{r}_5\\ \mathbf{r}_6\end{matrix}\right\rangle d\mathbf{r}_5\,d\mathbf{r}_6\left\langle \begin{matrix}\mathbf{r}_5\\ \mathbf{r}_6\end{matrix}\middle|\tilde{\Lambda}_k\right\rangle\ .$$

The $\lambda_k^{(\mathrm{form})}$ term on the right-hand side of (A17) is given by (A7), the $\mathscr{U}$ matrix element by (A8). In order to complete the translation into the forms of M_{pq} which appear in Eqs. (8) and (9) of Sec. II, we illustrate a part of the $R/(k-\mathscr{H})$ matrix element in (A17):

$$\left\langle \begin{matrix}\mathbf{r}_1\\ \mathbf{r}_2\end{matrix}\middle|\frac{P_U Q_L}{k-\mathscr{H}}\middle|\begin{matrix}\mathbf{r}_3\\ \mathbf{r}_4\end{matrix}\right\rangle=\sum\!\!\!\!\!\!\int_{\alpha,j}\frac{\langle\mathbf{r}_1|P_\alpha|\mathbf{r}_3\rangle\langle\mathbf{r}_4|Q_j|\mathbf{r}_2\rangle}{k-e_\alpha+e_j}\ . \tag{A18}$$

Equations (8) or (9) can now be obtained without any difficulty.

In order to arrive at Eqs. (13)–(17), it is preferable to carry out our manipulations on the abstract forms, and translate only our final results into conventional forms. To obtain new relationships, we imagine expanding to all orders in $\mathscr{U}$, reordering as suits our convenience, and resumming the infinite series of terms. Thus, (A6) in iterated form is

$$|\tilde{\Lambda}_k\rangle=\left[1+\mathscr{U}\frac{R}{k-\mathscr{H}}+\mathscr{U}\frac{R}{k-\mathscr{H}}\mathscr{U}\frac{R}{k-\mathscr{H}}+\cdots\right]\times|\lambda_k^{(\mathrm{form})}\rangle\ . \tag{A19}$$

Use of (A19) in (A1), with $p=f$, $q=i$ gives

$$M_{fi}=m_{fi}^{(\mathrm{form})}+(2k)^{-1/2}\langle\mathscr{F}^{fi}|\lambda_k^{(\mathrm{form})}\rangle\ , \tag{A20}$$

where

$$\langle\mathscr{F}^{fi}|=\left\langle\begin{matrix}f\\ i\end{matrix}\middle|\left[\mathscr{U}\frac{R}{k-\mathscr{H}}+\mathscr{U}\frac{R}{k-\mathscr{H}}\mathscr{U}\frac{R}{k-\mathscr{H}}+\cdots\right]\right. . \tag{A21}$$

Resumming and going over to the ket from the bra form gives the basic inhomogeneous integro-differential equation for $|\mathscr{F}^{fi}\rangle$:

$$(k-\mathscr{H})|\mathscr{F}^{fi}\rangle=R\,\mathscr{U}\left[\left|\begin{matrix}f\\ i\end{matrix}\right\rangle+|\mathscr{F}^{fi}\rangle\right]\ . \tag{A22}$$

Equation (13), on the one hand, and Eqs. (14)–(17), on the other hand, follow from Eqs. (A20) and (A22), respectively, upon translation into conventional notation. We will next indicate some of the key steps in this process.

Because of the presence of R in (A22), it follows immediately that

$$\left\langle\begin{matrix}a\\ b\end{matrix}\middle|\mathscr{F}^{fi}\right\rangle=0,\quad\left\langle\begin{matrix}j\\ k\end{matrix}\middle|\mathscr{F}^{fi}\right\rangle=0\ , \tag{A23}$$

where a,b refer to core states α,β and positron states. If we define the functions $F_{\alpha\pm}(\mathbf{r})$, which make their appearance in (13) and (14), in terms of $|\mathscr{F}^{fi}\rangle$ by[16]

$$F_{\alpha+}(\mathbf{r}_1)\equiv\langle\mathbf{r}_1|F_{\alpha+}\rangle\equiv\left\langle\begin{matrix}\mathbf{r}_1\\ \alpha\end{matrix}\middle|\mathscr{F}^{fi}\right\rangle\ , \tag{A24}$$

$$F_{\alpha-}(\mathbf{r}_2)\equiv\langle\mathbf{r}_2|F_{\alpha-}\rangle\equiv\left\langle\mathscr{F}^{fi}\middle|\begin{matrix}\alpha\\ \mathbf{r}_2\end{matrix}\right\rangle\ . \tag{A25}$$

Equations (20) are just translations of the first of Eqs. (A23). The various forms, $\langle\begin{smallmatrix}\mathbf{r}_1\\ \mathbf{r}_2\end{smallmatrix}|\mathscr{F}^{fi}\rangle$, $\langle\begin{smallmatrix}\mathbf{r}_1\\ p\end{smallmatrix}|\mathscr{F}^{fi}\rangle$, $\langle\begin{smallmatrix}p\\ \mathbf{r}_2\end{smallmatrix}|\mathscr{F}^{fi}\rangle$, $\langle\begin{smallmatrix}p\\ q\end{smallmatrix}|\mathscr{F}^{fi}\rangle$ are merely representations of $|\mathscr{F}^{fi}\rangle$ in different bases,[3] $(\mathrm{coordinate})_U$-$(\mathrm{coordinate})_L$, $(\mathrm{coordinate})_U$-$(\mathrm{particle})_L$, etc. We shall take advantage of the freedom of choosing any basis system which suits our convenience. In particular, by adding or subtracting zero [in the form (A23)], as needed, we obtain the identity

$$\left\langle\mathscr{F}^{fi}\middle|\begin{matrix}\mathbf{r}_1\\ \mathbf{r}_2\end{matrix}\right\rangle=\sum\!\!\!\!\!\!\int_a\left[\left\langle\mathscr{F}^{fi}\middle|\begin{matrix}\mathbf{r}_1\\ a\end{matrix}\right\rangle\langle a|\mathbf{r}_2\rangle+\left\langle\mathscr{F}^{fi}\middle|\begin{matrix}a\\ \mathbf{r}_2\end{matrix}\right\rangle\langle a|\mathbf{r}_1\rangle\right]\ , \tag{A26}$$

the use of which in (A20) [see the definitions (A24) and (25)] leads to

$$M_{fi}=m_{fi}^{(\mathrm{form})}+(2k)^{-1/2}\sum\!\!\!\!\!\!\int_a\langle a|\lambda_k^{(\mathrm{form})}|F_{a-}\rangle+\langle F_{a+}|\lambda_k^{(\mathrm{form})}|\alpha\rangle\ . \tag{A27}$$

If positron states are neglected, $\sum\!\!\!\!\!\!\int_a\to\sum_\alpha$ and (A27), written in conventional notation, is Eq. (13).

Equation (A26), with positron states neglected, is also used in (A22). If one successively takes the $\langle\begin{smallmatrix}\mathbf{r}_1\\ \alpha\end{smallmatrix}|$ matrix element of (A22) and the $|\begin{smallmatrix}\alpha\\ \mathbf{r}_2\end{smallmatrix}\rangle$ matrix elements of the adjoint of (A22), and uses the translation technique elaborated earlier in this appendix, one quickly obtains Eqs. (14)–(17).

APPENDIX B: REDUCTION OF EQUATIONS TO RADIAL FORM

To aid in the numerical evaluation of the transition matrix element M_{fi} in Eq. (13), we express the perturbed core orbitals $F_{\alpha\pm}(\mathbf{r})$ occurring in Eq. (14) in terms of angular momentum eigenstates and evaluate the angular parts of the integrals analytically. The radial differential equations resulting from the angular momentum decomposition of the perturbed orbitals $F_{\alpha\pm}(\mathbf{r})$, as well as the final radial integrals in Eq. (14), can then be evaluated numerically. In carrying out the angular momentum analysis, we follow closely the corresponding analysis of the RRPA equations described in Ref. 11. In the paragraphs below we summarize the results of this analysis, and write down the radial equations used in our numerical studies.

To facilitate comparison with previous work on atomic transition probabilities, we factor the photon polarization vector ϵ_k from the transition matrix element M_{fi} and from the local dipole currents $\lambda_k^{(\mathrm{form})}(\mathbf{r}_1)$ in Eqs. (4) and (5). We can then rewrite Eq. (13) as the matrix element of a nonlocal dipole operator $\mathbf{D}(\mathbf{r}_1,\mathbf{r}_2)$:

$$M_{fi}=i(k/2)^{1/2}\epsilon_k\cdot(\mathbf{D})_{fi} \ . \tag{B1}$$

The local dipole operators, $\mathbf{d}^{(\mathrm{form})}(\mathbf{r})$ corresponding to Eqs. (4) and (5) are

$$\mathbf{d}^{(\mathrm{velocity})}(\mathbf{r})=-i\boldsymbol{\alpha}/k \ , \tag{B2}$$

$$\mathbf{d}^{(\mathrm{length})}(\mathbf{r})=\mathbf{r} \ . \tag{B3}$$

Equation (13) becomes

$$(\mathbf{D})_{fi}=(\mathbf{d}^{(\mathrm{form})})_{fi}+\sum_{\alpha}\int[\,u_{\alpha}^{\dagger}(\mathbf{r})\mathbf{d}^{(\mathrm{form})}F_{\alpha-}(\mathbf{r})+F_{\alpha+}^{\dagger}(\mathbf{r})d^{(\mathrm{form})}u_{\alpha}(\mathbf{r})]d^3r \ , \tag{B4}$$

where the perturbed core orbitals $F_{\alpha\pm}(\mathbf{r})$ satisfy Eq. (14).

Expressing the vector operators $\mathbf{D}$ and $\mathbf{d}$ in a spherical basis and employing the Wigner-Eckart theorem, one may replace Eq. (B4) by an equation for the reduced matrix element $\langle f||D||i\rangle$ of the dipole operator

$$(D_{\mu})_{fi}=(-1)^{j_f-m_f}\begin{pmatrix} j_f & 1 & j_i \\ -m_f & \mu & m_i \end{pmatrix}\langle f||D||i\rangle \ , \tag{B5}$$

where j_i, m_i, j_f, and m_f are angular-momentum quantum numbers of the initial and final states, and where μ designates a spherical component of the dipole operator $\mathbf{D}$. The reduced matrix element satisfies the equation

$$\langle f||D||i\rangle=\langle f||d^{(\mathrm{form})}||i\rangle+\sum_{\alpha,q}[\langle\alpha||d^{(\mathrm{form})}||q-\rangle+(-1)^{j_q-j_\alpha}\langle q+||d^{(\mathrm{form})}||\alpha\rangle] \ . \tag{B6}$$

In Eq. (B6) the summation index α refers to occupied core subshells (n_α,κ_α) where n_α is the subshell principal quantum number and κ_α is the relativistic angular-momentum quantum number: $\kappa_\alpha=\mp(j_\alpha+\frac{1}{2})$ for $j_\alpha=(l_\alpha\pm\frac{1}{2})$; l_α being the orbital angular momentum of the core orbital, α. The states $||q\pm\rangle$ in Eq. (B6) refer to angular momentum components of the perturbed core orbitals $F_{\alpha\pm}(\mathbf{r})$. Only those components of the perturbed orbital $F_{\alpha\pm}(\mathbf{r})$ which satisfy the angular momentum selection rules $|j_\alpha-1|\le j_q\le j_\alpha+1$, and the parity selection rule, $l_\alpha+l_q$ equals an odd integer, contribute to the sum in (B6).

Each of the terms on the right-hand side of Eq. (B6) can be expressed as a product of an angular factor and a radial integral. The angular factors are conveniently written in terms of reduced matrix elements of a normalized spherical harmonic $C_{LM}(\hat{\mathbf{r}})=\sqrt{4\pi/(2L+1)}Y_{LM}(\hat{\mathbf{r}})$:

$$C_L(a,b)=\langle\kappa_a||C_L||\kappa_b\rangle=(-1)^{j_a+1/2}\sqrt{[a][b]}\begin{pmatrix} j_a & j_b & L \\ -\frac{1}{2} & \frac{1}{2} & 0 \end{pmatrix}\pi(l_a+l_b+L) \ , \tag{B7}$$

where $\pi(x)=1$ if x is even, and 0 if x is odd, and where $[a]=2j_a+1$. We write the length form matrix elements of $\mathbf{d}$ as

$$\langle a||d^{(\mathrm{length})}||b\rangle=C_1(a,b)\int_0^{\infty}[P_a(\mathbf{r})P_b(r)+Q_a(r)Q_b(r)]dr \ , \tag{B8}$$

and the velocity form of $\mathbf{d}$ as

$$\langle a||d^{(\mathrm{velocity})}||b\rangle=C_1(a,b)\int_0^{\infty}[(1-\kappa_b+\kappa_a)Q_a(r)P_b(r)-(1+\kappa_b-\kappa_a)P_a(r)Q_b(r)]dr \ . \tag{B9}$$

The functions $P_a(r)$ and $Q_a(r)$ in Eqs. (B8) and (B9) are large and small component radial Dirac functions. For the core orbitals, α, and for the final and initial orbitals, f and i, the radial functions are solutions to frozen-core radial DF equations. For the components $q\pm$ of the perturbed core orbitals $F_{\alpha\pm}(\mathbf{r})$, the radial functions satisfy inhomogeneous equations following from Eq. (14).

The radial orbitals $P_a(r)$ and $Q_a(r)$ are collected into a two-component radial orbital, $R_a(r)$, and the radial DF equations are written

$$(H_a+V_{\mathrm{DF}})R_a(r)=e_aR_a(r) \ . \tag{B10}$$

The term H_a is the kinetic energy and nuclear potential contribution to the radial Dirac Hamiltonian and V_{DF} is the frozen-core Hartree-Fock potential:

$$V_{\rm DF}R_a(r)=\sum_\alpha [\alpha]\left[v_0(\alpha,\alpha,r)R_a(r) + \sum_L \Lambda_L(\alpha,a)v_L(\alpha,a,r)R_\alpha(r)\right], \tag{B11}$$

where we have introduced the exchange angular-momentum factor

$$\Lambda_L(a,b)=\frac{C_L(a,b)^2}{[a][b]}, \tag{B12}$$

and the multipole components of the Coulomb potential

$$v_L(a,b,r)=\int \frac{r_<^L}{r_>^{L+1}}R_a^\dagger(r')R_b(r')dr' . \tag{B13}$$

In Eq. (B13) $r_<$ and $r_>$ designate the minimum and maximum of r and r', respectively.

Before writing down the equations satisfied by the perturbed radial functions $R_{q\pm}(r)$ it is convenient to introduce one further angular coupling factor:

$$A(a,b,c,d,L,J)=(-1)^{J+L-j_b-j_c}C_L(a,b)C_L(c,d)\begin{Bmatrix} j_a & j_b & L \\ j_d & j_c & J \end{Bmatrix} . \tag{B14}$$

The perturbed orbital equations become

$$\begin{aligned}(H_q+V_{\rm HF}-e_\alpha-\omega)R_{q+}(r)=&-\tfrac{1}{3}C_1(\alpha,q)C_1(i,f)v_1(i,f,r)R_\alpha(r)+\sum_L A(\alpha,i,q,f,L,1)v_L(i,\alpha,r)R_f(r)\\ &-\sum_{p,\beta}\tfrac{1}{3}C_1(\alpha,q)C_1(\beta,p)[v_1(p-,\beta,r)+v_1(\beta,p+,r)]R_\alpha(r)\\ &+\sum_{p,\beta}\sum_L A(\alpha,\beta,q,p,L,1)v_L(\beta,\alpha,r)R_{p+}(r)\\ &+\sum_{p,\beta}\sum_L (-1)^{j_p-j_\beta}A(\alpha,p,q,\beta,L,1)v_L(p-,\alpha,r)R_\beta(r) ,\end{aligned} \tag{B15}$$

$$\begin{aligned}(H_q+V_{\rm HF}-e_\alpha+\omega)R_{q-}(r)=&-\tfrac{1}{3}C_1(\alpha,q)C_1(i,f)v_1(f,i,r)R_\alpha(r)+\sum_L (-1)^{j_f-j_i}A(\alpha,f,q,i,L,1)v_L(f,\alpha,r)R_i(r)\\ &-\sum_{p,\beta}\tfrac{1}{3}C_1(\alpha,q)C_1(\beta,p)[v_1(p+,\beta,r)+v_1(\beta,p-,r)]R_\alpha(r)\\ &+\sum_{p,\beta}\sum_L A(\alpha,\beta,q,p,L,1)v_L(\beta,\alpha,r)R_{p-}(r)\\ &+\sum_{p,\beta}\sum_L (-1)^{j_p-j_\beta}A(\alpha,p,q,\beta,L,1)v_L(p+,\alpha,r)R_\beta(r) .\end{aligned} \tag{B16}$$

In Eqs. (B15) and (B16) we set $\omega=e_f-e_i$ and we let $p\pm$ represent the angular-momentum components of the perturbed core orbital $F_{\beta\pm}(\mathbf{r})$. The coupled equations are solved numerically and the solutions are used to evaluate the reduced matrix elements in Eq. (B6). For the example of Li I considered in the present paper, the system (B15) and (B16) consists of four coupled equations corresponding to the two angular momenta, $\kappa_q=1,-2$, of the perturbed $1s_{1/2}$ core orbital permitted by dipole selection rules.

[1]T. Fulton, Nucl. Phys. **A443**, 77 (1985).

[2]G. Feldman and T. Fulton, Nucl. Phys. **A362**, 361 (1981).

[3]G. Feldman and T. Fulton, Ann. Phys. (N.Y.) **152**, 367 (1984).

[4]G. Feldman and T. Fulton, Ann. Phys. (to be published).

[5]Equation (2) corresponds to Eq. (2.3) of Ref. 1. The function Λ is the reduced vertex of field theory; $\overline{\Lambda}$ is the instantaneous relative time version of Λ; $\widetilde{\Lambda}$ is the form appropriate to the dipole approximation [see Eq. (2.5) of Ref. 1, where $\widetilde{\Lambda}$ is designated as λ].

[6]W. E. Lamb, Jr. and R. C. Retherford, Phys. Rev. **79**, 549 (1950); **81**, 222 (1951); W. E. Lamb, Jr., *ibid.* **85**, 259 (1952).

[7]D. H. Kobe, Phys. Rev. A **19**, 205 (1979); K.-H. Yang, Ann. Phys. (N.Y.) **101**, 62 (1976).

[8]W. R. Johnson, in *Atomic Theory Workshop on Relativistic and QED Effects in Heavy Atoms, Gaithersburg, MD, 1985,* AIP Conf. Proc. No. 136, edited by H. P. Kelly and Y.-K. Kim (AIP, New York, 1985), p. 150.

[9]W. R. Johnson, Dong-Sheng Guo, M. Idrees, and J. Sapirstein, Phys. Rev. A **34**, 1043 (1986).

[10]As a practical criterion for the exclusion of such low-lying core states, one can study the stabilization of the value of M_{fi} on the one hand and the convergence of the M_{fi}'s based on $m_{fi}^{(\rm length)}$ and $m_{fi}^{(\rm velocity)}$ on the other hand. One must continue to add core states until such stability and convergence is achieved (see Ref. 9). Of course, for Li I, both $1s$ core states must be included.

[11]W. R. Johnson, C. D. Lin, K. T. Cheng, and C. M. Lee, Phys. Scr. **21**, 409 (1980).

[12]This point was not stressed, but was implied, in the discussion following Eq. (3.58) of Ref. 3. In any event, as the opening paragraph of section 2.2.2 of Ref. 11 indicates, the orthogonality conditions are taken into account there, as they are in the present work.

[13]A. Gaupp, P. Kuske, and H. J. Andrä, Phys. Rev. A **26**, 3351 (1982).

[14]Compare in particular, Eqs. (3.28) and (3.38) of Ref. 3 or, alternatively, (3.48) and (3.53) of Ref. 4 [noting also (3.50) and (3.37) of Ref. 4].

[15]M. Ya. Amus'ya and N. A. Cherepkov, Case Stud. At. Phys. **5**, 47 (1975).

[16]The corresponding definitions of the RRPA functions of Ref. 11, as they *should* appear in Ref. 3 are $w_{\alpha+}(\mathbf{r}_1)\equiv\langle {}^{\mathbf{r}_1}_{\alpha}|\bar{\chi}_n\rangle^N$, $w^*_{\alpha-}(\mathbf{r}_1)\equiv\langle {}^{\alpha}_{\mathbf{r}_2}|\bar{\chi}_n\rangle^N$, where $|\bar{\chi}_n\rangle^N$ is defined in that reference. Unfortunately, there is an inadvertent interchange of $w_{\alpha+}$ and $w^*_{\alpha-}$ in Eqs. (3.56) and (3.57) of Ref. 3.

[17]Lloyd Armstrong Jr., W. R. Fielder, and Dong L. Lin, Phys. Rev. A **14**, 1114 (1976).

[18]S. Garpman, Phys. Scr. **12**, 295 (1975).

[19]A. W. Weiss, Astrophys. J. **138**, 1262 (1963).

[20]D. W. Norcross, J. Phys. B **4**, 1458 (1971).

[21]R. K. Morita and P. K. Mukerjee, Int. J. Quantum Chem. **6**, 211 (1972).

[22]M. Ya. Amusya, N. A. Cherepkov, Dj. Zivanovic, and V. Radojevic, Phys. Rev. A **13**, 1466 (1976).