# Radiative decay of hydrogenlike atoms in an electric field

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Photon emission by an atom in the metastable 2S state in a weak static electric field is examined within the context of bound-state quantum electrodynamics. Low's method is applied to obtain the expansion in terms of Coulomb wave functions of the propagation function for an electron interacting with the nuclear Coulomb field, the applied electric field, and the quantized radiation field. For a weak applied field, and to lowest relative order in  $\alpha/\pi$ , the 2S-state resonant contribution to the propagation function can be factorized to provide the basis for a calculation of the angular distribution of single-photon radiation and lifetime of the excited state. A numerical evaluation is given, which takes into account effects of electric dipole, magnetic dipole, and magnetic quadrupole emission, relativistic corrections, and finite level widths.

#### I. INTRODUCTION

The Lamb shift in high-Z hydrogenlike atoms may be determined experimentally by observing decays of the  $2S_{1/2}$  state in a uniform constant electric field.<sup>1</sup> The experiments measure either the lifetime of the excited state<sup>2-6</sup> or the angular distribution of radiation from the single-photon decay of the excited state.<sup>7,8</sup> In either case, the interpretation of the experiments is based on the theoretical connection between the observed property of the decay and the corresponding value for the Lamb shift. Methods for calculating the decay properties of a hydrogenlike atom in a constant electric field have been discussed by Lamb and Retherford,<sup>9</sup> Fan et al.,<sup>2</sup> Fontana and Lynch,<sup>10</sup> Holt and Sellin,<sup>11</sup> Grisaru et al.,<sup>12</sup> Drake et al.,<sup>13</sup> and Kelsey and Macek.14

The purpose of this paper is to examine radiative decay of an atom in an applied electric field within the framework of the bound interaction picture of quantum electrodynamics, and to give a detailed evaluation of the relevant properties of the decays as an aid in the interpretation of the Lamb-shift experiments.

Radiative decay of the 2S state in an electric field is strongly influenced by the electric field mixing of the 2S and 2P states and by the complex level shifts due to the mass operator. In order to take these effects into account within the S-matrix formalism, we examine the scattering process in which an atom is excited to the narrow 2S-state resonance and decays by single-photon emission. This is the approach employed by Low to study the field-free resonant line shape for an atom.<sup>15</sup> The decay properties are determined mainly by the propagation function for the bound electron, with radiative corrections and the electric field taken into account. We write an approximate expression for the full propagator which for weak electric fields is valid to lowest relative order in  $\alpha/\pi$  for the 2S-state resonant contribution to the scattering. The expressions for the 2S-state lifetime based on the resonance width, and for the angular distribution of radiation based on the resonant scattering amplitude are evaluated relativistically, taking into account effects of electric dipole, magnetic dipole, and magnetic quadrupole decay and complex radiative level shifts. Hyperfine-structure effects are not considered.

## **II. SCATTERING FORMULATION**

We employ the Feynman-Dyson S-matrix formalism. The nuclear Coulomb field is taken into account by working in the Furry bound-interaction picture and the external constant electric field is included as a perturbation along with the radiative corrections. We assume that the appropriate renormalization is applied so that infinite operators are replaced by their finite parts.

In order to be definite, we consider a beam-foil experiment in which an atom emerges from a foil in an excited state and emits a photon of energy  $k \approx E_{2S} - E_{1S}$ . An effective Feynman diagram for this process is shown in Fig. 1. In that figure, the cross labeled W represents an external potential that accounts for the excitation of the atom, the single line corresponds to the electron in the Coulomb field of the nucleus, and the double line corresponds to the electron in the Coulomb field of the nucleus including the effects of radiative corrections and the external electric field. The associated crossed diagram does not contribute

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FIG. 1. Feynman diagram for excitation of an atom by an effective potential W followed by emission of a photon with momentum  $\vec{k}$  and polarization vector  $\hat{\epsilon}_{\lambda}$ .

to the scattering resonance. The effect of the electric field in the external legs, which represent the electron in the 1S state, is negligible as shown by a simple perturbation theory estimate. Radiative corrections in the external legs and vertex corrections to the diagram in Fig. 1 contribute in relative order  $\alpha/\pi$  and are not included here. In related work, Lin and Feinberg<sup>16</sup> and Barbieri and Sucher<sup>17</sup> have examined radiative corrections to the decay rate for 2S - 1S + one photon in hydrogenlike ions with no applied field. Their calculations show that these corrections are negligible for the present work.

The complete propagation function  $S'_F$ , corresponding to the double line in Fig. 1, which includes radiative corrections and the external electric field, can be written in terms of the Coulomb propagation function  $S_F$  as the solution of

$$S'_{F} = S_{F} - iS_{F} (\Sigma + V + V^{(1)} + V^{(2)} + \cdots)S'_{F}, \qquad (1)$$

where  $\Sigma$  is the finite part of the operator corresponding to the sum over proper self-energy and vacuum-polarization corrections, V is the operator corresponding to interaction with the external electric field, and  $V^{(n)}$  corresponds to the renormalized sum over radiative corrections which contain n interactions with the external electric field. Feynman diagrams associated with these operators are shown in Fig. 2. Feynman diagrams for the perturbation expansion of Eq. (1) are shown in Fig. 3. In coordinate space, Eq. (1) takes the form



FIG. 2. Feynman diagrams for the mass operator  $\Sigma$ , the applied-field operator V, and the operator  $V^{(1)}$ .



FIG. 3. Feynman diagrams for the perturbation expansion of Eq. (1).

$$S'_{F}(x_{2}, x_{1}) = S_{F}(x_{2}, x_{1}) - i \int d^{4}x_{4} \int d^{4}x_{3}$$
$$\times S_{F}(x_{2}, x_{4})U(x_{4}, x_{3})S'_{F}(x_{3}, x_{1}), \qquad (2)$$

where  $U(x_4, x_3) = U(\mathbf{\tilde{x}}_4, \mathbf{\tilde{x}}_3; t_4 - t_3)$  is the kernel for the operator

$$U = \Sigma + V + V^{(1)} + V^{(2)} + \cdots$$
 (3)

The Coulomb-field propagation function  $S_F$  is written as

$$S_F(x_2, x_1) = \frac{1}{2\pi i} \int_{-\infty}^{\infty} d\omega \sum_n \frac{\psi_n(\mathbf{\bar{x}}_2)\overline{\psi}_n(\mathbf{\bar{x}}_1)}{E_n - \omega(1 + i\epsilon)} e^{-i\omega(t_2 - t_1)}, \quad (4)$$

where  $\psi_n$  is a bound or continuum Dirac wave function, with energy  $E_n$ , for an electron in the nuclear Coulomb field. In Eq. (4), the summation symbol is understood to mean summation over all bound states and integration over the continuous spectrum. Following Low,<sup>15</sup> we write

$$S'_{F}(x_{2}, x_{1}) = \frac{1}{2\pi i} \int_{-\infty}^{\infty} d\boldsymbol{\omega} \sum_{nm} f_{nm}(\boldsymbol{\omega}) \psi_{n}(\mathbf{\bar{x}}_{2}) \overline{\psi}_{m}(\mathbf{\bar{x}}_{1}) e^{-i \, \boldsymbol{\omega} (t_{2} - t_{1})} ,$$
(5)

where the coefficients  $f_{nm}(\omega)$  are to be determined so that Eq. (2) is satisfied. Let

$$U_{nm}(\omega) = \int_{-\infty}^{\infty} dt \, e^{i\,\omega t} \int d^3 \vec{\mathbf{x}}_2 \int d^3 \vec{\mathbf{x}}_1 \, \vec{\psi}_n(\vec{\mathbf{x}}_2) \\ \times U(\vec{\mathbf{x}}_2, \vec{\mathbf{x}}_1; t) \psi_m(\vec{\mathbf{x}}_1) ; \qquad (6)$$

then

$$U(x_{2}, x_{1}) = \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega \sum_{nm} U_{nm}(\omega) \gamma^{0} \psi_{n}(\vec{\mathbf{x}}_{2})$$
$$\times \overline{\psi}_{m}(\vec{\mathbf{x}}_{1}) \gamma^{0} e^{-i\omega(t_{2}-t_{1})} . \tag{7}$$

Substitution of Eqs. (4), (5), and (7) into (2) yields equations<sup>15</sup> which determine the  $f_{nm}(\omega)$ :

$$f_{nm}(\omega) = \frac{1}{E_n - \omega(1 + i\epsilon)} \left( \delta_{nm} - \sum_{l} U_{nl}(\omega) f_{lm}(\omega) \right) .$$
(8)

The matrix element corresponding to the Feynman diagram in Fig. 1 is (in natural units:  $m_e = c = \hbar = 1$ )

$$M = e \left(\frac{2\pi}{k}\right)^{1/2} \int d^4x_2 \int d^4x_1 \overline{\psi}_{1S}^{\mu}(x_2) \overline{\gamma} \cdot \hat{\epsilon}_{\lambda} e^{ik \cdot x_2} \\ \times S'_F(x_2, x_1) \gamma^0 W(x_1) \psi_{1S}^{\mu}(x_1) , \qquad (9)$$

where  $\bar{k}$  and  $\hat{\epsilon}_{\lambda}$  are the momentum and polarization vectors for the emitted photon, and  $\mu_i$  and  $\mu_f$  are the eigenvalues of  $J_z$  for the initial and final electron states. Writing

$$W(\mathbf{x},t) = \int d\boldsymbol{\omega} \, e^{-i\,\boldsymbol{\omega}\,t} \, \mathbf{\mathfrak{W}}(\mathbf{x},\boldsymbol{\omega}) \tag{10}$$

for the effective foil potential and employing the expression (5) for  $S'_F(x_2, x_1)$ , we have

$$M = -2\pi i e \left(\frac{2\pi}{k}\right)^{1/2} \int d^{3}\vec{\mathbf{x}}_{2} \int d^{3}\vec{\mathbf{x}}_{1} \psi_{1S}^{\mu_{f}^{\dagger}}(\vec{\mathbf{x}}_{2}) \vec{\alpha} \cdot \hat{\boldsymbol{\epsilon}}_{\lambda} e^{-i\vec{\mathbf{k}}\cdot\vec{\mathbf{x}}_{2}} \sum_{nm} f_{nm}(E_{1S}+k)\psi_{n}(\vec{\mathbf{x}}_{2})\psi_{m}^{\dagger}(\vec{\mathbf{x}}_{1}) \cdot \boldsymbol{w}(\vec{\mathbf{x}}_{1},k)\psi_{1S}^{\mu_{i}}(\vec{\mathbf{x}}_{1}) .$$
(11)

The relevant portion of the scattering amplitude is the contribution from the narrow resonance near  $k \approx E_{2S} - E_{1S}$ . An approximate solution of Eq. (8) for the  $f_{nm}(\omega)$  which gives the leading contribution for resonant scattering is considered in Sec. III.

# III. APPROXIMATION FOR $S'_F$

The propagation function  $S'_F$  is determined, in principle, exactly by Eq. (8). In order to carry out this calculation, we make two approximations in Eq. (8) which are expected to be valid for the resonance scattering.

First, we omit the higher-order corrections  $V^{(1)}, V^{(2)}, \ldots$  in Eq. (1). In Eq. (8), this means replacing  $U_{nm}(\omega)$  by  $\sum_{nm}(\omega) + V_{nm}$ , where the latter matrix elements are given by the right-hand side of Eq. (6) with U replaced by  $\Sigma$  and V, respectively. The fact that  $V_{nm}$  is independent of  $\omega$  follows from the form of  $V(x_2, x_1) = \delta^4(x_2 - x_1)\gamma^0 e \vec{E} \cdot \vec{x}_2$ , in which -e is the electron charge and  $\vec{\mathbf{E}}$  is the applied field. The sum  $\Sigma + V^{(1)} + V^{(2)} + \cdots$  is formally equivalent to the sum of the mass- and vacuum-polarization operators in both the Coulomb and applied electric fields. For a weak applied electric field, the main contribution comes from the leading terms, in analogy with radiative corrections in the presence of hyperfine structure. By weak field, we mean  $E(V/cm) \ll 10^2 Z^5$ , where E is the magnitude of the applied field, and Z is the nuclear charge of the hydrogenlike atom. This condition insures that the  $2S_{1/2}$ - $2P_{1/2}$  matrix element of the electric field perturbation is small compared to the zero-field radiative level separation (Lamb shift). The condition is met in current Lamb-shift experiments with Z = 18 and  $Z = 8.^{6,7}$  Despite the weak-field condition the effect of V is large, because it mixes states of opposite parity and allows strong electric dipole transitions which would otherwise be forbidden for the 2S state. The term  $V^{(1)}$  is the sum of the

vertex and vacuum-polarization corrections to V of relative order  $\alpha/\pi$ , and is therefore neglected.

The second approximation is made by including only the mixing among states in the subset  $S = \{2S_{1/2}, 2P_{1/2}, 2P_{3/2}\}$ . The corrections to this approximation are terms of relative order  $\Sigma_{nm}(\omega)/(E_r - \omega)$  and  $V_{nm}/(E_r - \omega)$ , where  $r \notin S$ , which are small in the resonance region  $\omega \approx E_{2S}$ . The lowestorder correction is given in Appendix A. To implement this approximation in Eq. (8), we replace  $\Sigma_{nm}(\omega)$  and  $V_{nm}$  by  $\bar{\Sigma}_{nm}(\omega)$  and  $\bar{V}_{nm}$ , respectively, where

$$\tilde{\Sigma}_{nm}(\omega) = \begin{cases} \Sigma_{nm}(\omega), & \text{for } n \text{ and } m \in \mathcal{S}, \\ 0, & \text{for } n \text{ or } m \notin \mathcal{S}, \end{cases}$$
(12)

and

$$\tilde{V}_{nm} = \begin{cases} V_{nm}, & \text{for } n \text{ and } m \in S \\ 0, & \text{for } n \text{ or } m \in S \end{cases}$$
(13)

The above-mentioned replacements lead to a modified version of Eq. (8) with solutions  $g_{nm}(\omega)$  given by

$$g_{nm}(\omega)$$

$$=\frac{1}{E_{n}-\omega(1+i\epsilon)}\left(\delta_{nm}-\sum_{l}\left[\tilde{\Sigma}_{nl}(\omega)+\tilde{V}_{nl}\right]g_{lm}(\omega)\right)$$
(14)

 $\mathbf{or}$ 

$$\sum_{l} \{ [E_{n} - \omega (1 + i\epsilon)] \delta_{nl} + \tilde{\Sigma}_{nl} (\omega) + \tilde{V}_{nl} \} g_{lm} (\omega) = \delta_{nm} .$$
(15)

The solutions  $g_{nm}(\omega)$  are obtained by inverting the infinite-dimensional matrix in the curly brackets in Eq. (15). In view of Eqs. (12) and (13), the matrix is diagonal except for the matrix elements between states in the subset \$. Hence

$$g_{nm}(\omega) = \delta_{nm} / [E_n - \omega(1 + i\epsilon)]$$
 for  $n$  or  $m \in S$ . (16)

In the submatrix for the states in §, the matrix elements  $\tilde{\Sigma}_{nm}(\omega)$  are nonzero only if n=m, because the operator for radiative corrections in an external Coulomb field connects only states with the same eigenvalue for  $J^2, J_z$ , and parity. We thus have

$$\sum_{l \in S} \{ [E_n + \Sigma_{nn}(\omega) - \omega] \delta_{nl} + V_{nl} \} g_{lm}(\omega)$$
$$= \delta_{nm} \text{ for } n \text{ and } m \in S.$$
(17)

For convenience, we may assume that the applied

electric field is in the z direction and the Coulomb states are labeled by  $\mu$ , the eigenvalue of  $J_z$ . Then the matrix elements  $V_{nm}$  are nonzero only if the states n and m have the same eigenvalue for  $J_z$ , and Eq. (17) separates into four uncoupled equations corresponding to  $\mu = -\frac{3}{2}, -\frac{1}{2}, \frac{1}{2}, \frac{3}{2}$ . [Note that  $\sum_{nn}(\omega)$  is independent of  $\mu$  for a given state.] Denoting the states  $2S_{1/2}, 2P_{1/2}, \text{ and } 2P_{3/2}$  by s, p, and q, respectively, and displaying explicitly the values for  $\mu$ , we can express the solutions  $g_{nm}(\omega)$ of Eq. (17) in the factorized form

Here

$$E'_n = E_n + \Sigma_{nn}(\omega) \tag{19}$$

and the  $E''_n$  are the roots of

$$(E'_{s} - x)(E'_{p} - x)(E'_{q} - x) - (E'_{q} - x)|V^{(1/2)}_{ps}|^{2} - (E'_{p} - x)|V^{(1/2)}_{qs}|^{2} = 0,$$
(20)

with  $E''_n \to E'_n$  as  $V \to 0$ . The solutions  $g_{nm}(\omega)$  in Eqs. (16) and (18) define an approximate propagation function  $S_F^{(0)}$  given by

$$S_{F}^{(0)}(x_{2}, x_{1}) = \frac{1}{2\pi i} \int_{-\infty}^{\infty} d\omega \sum_{n \, m} g_{nm}(\omega) \psi_{n}(\vec{\mathbf{x}}_{2}) \overline{\psi}_{n}(\vec{\mathbf{x}}_{1}) \, e^{-i \, \omega \, (t_{2} - t_{1})} \,.$$
(21)

## IV. RESONANCE SCATTERING

The leading resonance contribution to M in Eq. (11) may be evaluated by replacing  $f_{nm}(E_{1S}+k)$  by the  $g_{nm}(E_{1S}+k)$  given in Eq. (18). The resonances in the amplitude M are determined by the first energy denominator in each of the four terms on the right-hand side in Eq. (18) for a weak applied electric field. This follows from the fact that all other energy differences in Eq. (18), such as  $E''_{g}$  $-E''_{p}$ , are slowly varying as  $\omega$  varies over the resonance region. The electric field shifts in the energy,  $E''_{n} - E'_{n}$ , are small compared to the energy separations  $E'_{n} - E'_{m}$ , which include radiative corrections; the radiative corrections  $E'_{n} - E_{n}$  are slowly varying, since the scale for the dependence of  $\sum_{nm}(\omega)$  on  $\omega$  is determined by the Coulomb energy-level separations, i.e.,

$$\Sigma_{nn}(E_n + \delta E) = [1 + O(\delta E/E_c)]\Sigma_{nn}(E_n),$$

where  $E_c = O(Z\alpha)^2$ . In addition,  $\mathfrak{W}(\mathbf{\bar{x}}, k)$  in (11) is taken to be slowly varying over the resonance region  $k \approx E_{2S} - E_{1S}$ .

The narrow resonance corresponding to the longlived 2S state is thus given by the first term in Eq. (18) and

$$M_{2S} \approx -2\pi i e \left(\frac{2\pi}{k}\right)^{1/2} \frac{1}{E_s'' - E_{1S} - k} \frac{(E_s'' - E_p')(E_s'' - E_q')}{(E_s'' - E_p'')(E_s'' - E_q'')} \times \sum_{\mu=\pm 1/2} A_{\mu_f \mu} B_{\mu \mu_i}, \qquad (22)$$

where

$$A_{\mu_{f}\mu} = \int d^{3}\vec{\mathbf{x}}\psi_{1S}^{\mu}^{\dagger}(\vec{\mathbf{x}})\vec{\alpha}\cdot\hat{\epsilon}_{\lambda}e^{-i\vec{\mathbf{k}}\cdot\vec{\mathbf{x}}}$$

$$\times \left(\psi_{s}^{\mu}(\vec{\mathbf{x}}) + \frac{V_{ps}^{\mu}}{E_{s}^{\prime\prime} - E_{p}^{\prime\prime}}\psi_{p}^{\mu}(\vec{\mathbf{x}}) + \frac{V_{qs}^{\mu}}{E_{s}^{\prime\prime} - E_{q}^{\prime\prime}}\psi_{q}^{\mu}(\vec{\mathbf{x}})\right)$$
(23)

and

$$B_{\mu\mu}{}_{i} = \int d^{3}\mathbf{\hat{x}} \left( \psi_{s}^{\mu} \dagger(\mathbf{\hat{x}}) + \frac{V_{sp}^{\mu}}{E_{s}'' - E_{p}'} \psi_{p}^{\mu\dagger}(\mathbf{\hat{x}}) + \frac{V_{sq}^{\mu}}{E_{s}'' - E_{q}'} \psi_{q}^{\mu\dagger}(\mathbf{\hat{x}}) \right) \\ \times \mathbf{\hat{w}}(\mathbf{\hat{x}}, k) \psi_{1s}^{\mu}(\mathbf{\hat{x}}) .$$

$$(24)$$

The emission line shape is determined primarily by the factor

$$|1/(E_{s}'' - E_{1s} - k)|^{2} = 1/[(\operatorname{Re}E_{s}'' - E_{1s} - k)^{2} + (\operatorname{Im}E_{s}'')^{2}]$$
(25)

in  $|M_{2S}|^2$ . The decay rate R for the resonance is related to the imaginary part of the pole in k by

$$R = -2 \operatorname{Im} E_s'' \tag{26}$$

evaluated at  $k = E''_s - E_{1s}$ . Equation (20) may be solved perturbatively in V for  $E''_s$ , and yields

$$E_{s}^{\prime\prime} = E_{s}^{\prime} + \frac{|V_{ps}^{(1/2)}|^{2}}{E_{s}^{\prime} - E_{p}^{\prime}} + \frac{|V_{qs}^{(1/2)}|^{2}}{E_{s}^{\prime} - E_{q}^{\prime}} + O\left(\frac{|V_{ps}^{(1/2)}|^{4}}{(E_{s}^{\prime} - E_{p}^{\prime})^{3}}\right) .$$
(27)

In Eq. (27), we may approximate  $E'_n$  by

$$E'_{n} \approx E_{n} + \sum_{nn} (E_{n}) = E_{n} + \Delta E_{n} - \frac{1}{2} i \Gamma_{n} , \qquad (28)$$

where  $\Delta E_n$  and  $-\frac{1}{2}\Gamma_n$  are the real and imaginary parts of  $\Sigma_{nn}(E_n)$ . The lowest-order decay rate Ris thus given by

$$R = \Gamma_s + \frac{|V_{ps}^{(1/2)}|^2}{S^2 + \frac{1}{4}\Gamma_p^2} \Gamma_p + \frac{|V_{qs}^{(1/2)}|^2}{(\Delta E - S)^2 + \frac{1}{4}\Gamma_q^2} \Gamma_q , \qquad (29)$$

where  $S = \Delta E_s - \Delta E_p$ , and  $\Delta E = E_q + \Delta E_q - E_p - \Delta E_p$ , and  $\Gamma_s$  is neglected compared to  $\Gamma_p$  or  $\Gamma_q$  in the last two terms. The width  $\Gamma_s$  includes the twophoton decay rate of the 2S state, which is contained in the fourth-order ( $\alpha^2$ ) contributions to  $\Sigma_{nn}(E_n)$ . Equation (29) agrees with the decay rate obtained by Lamb's method of derivation,<sup>9</sup> which is based on the Weisskopf-Wigner approach.<sup>18</sup>

The angular distribution of resonance radiation is determined by the transition probability  $|M_{2S}|^2$ averaged over angular momentum projections of the initial atomic 1S state (for an unpolarized incident beam), and summed over final atomic angular momentum projections and photon polarizations. The relevant factor from Eq. (22) is

$$\sum_{\lambda \mu_{f} \mu_{i}} \left| \sum_{\mu} A_{\mu_{f} \mu} B_{\mu \mu_{i}} \right|^{2} = \sum_{\substack{\lambda \mu_{f} \mu_{i} \\ \mu \mu'}} A_{\mu_{f} \mu} A_{\mu_{f} \mu'} B_{\mu \mu_{i}} B_{\mu' \mu_{i}}^{*} .$$
(30)

The term  $\sum_{\mu_i} B_{\mu \mu_i} B^*_{\mu' \mu_i}$  describes the excitation process. We consider here only the case in which no polarization of the excited state is introduced by the foil, i.e., when

$$\sum_{\mu_i} B_{\mu\mu_i} B^*_{\mu'\mu_i} \propto \delta_{\mu\mu'} . \qquad (31)$$

This is expected to apply to ordinary beam-foil experiments. Consequences of a polarized excited state are discussed in Appendix B. From Eqs. (30) and (31) the angular distribution of radiation is determined by

$$\sum_{\lambda \mu_{f} \mu} |A_{\mu_{f} \mu}|^{2}, \qquad (32)$$

where to lowest order we have

$$A_{\mu_{f}\mu} = \int d^{3}\mathbf{\tilde{x}} \psi_{1S}^{\mu_{f}\dagger}(\mathbf{\tilde{x}}) \mathbf{\alpha} \cdot \hat{\epsilon}_{\lambda} e^{-i\mathbf{\tilde{k}} \cdot \mathbf{x}}$$

$$\times \left( \psi_{s}^{\mu}(\mathbf{\tilde{x}}) + \frac{V_{\rho_{s}}^{\mu}}{S + \frac{1}{2}i\Gamma_{\rho}} \psi_{\rho}^{\mu}(\mathbf{\tilde{x}}) - \frac{V_{q_{s}}^{\mu}}{\Delta E - S - \frac{1}{2}i\Gamma_{q}} \psi_{q}^{\mu}(\mathbf{\tilde{x}}) \right), \qquad (33)$$

with  $E''_s - E'_p$  and  $E'_q - E''_s$  approximated by  $S + \frac{1}{2}i\Gamma_p$ and  $\Delta E - S - \frac{1}{2}i\Gamma_q$ , respectively, and  $|\vec{\mathbf{k}}|$  evaluated at  $E_{2S} - E_{1S}$ .

# V. ANGULAR DISTRIBUTION AND DECAY RATE

To evaluate the angular distribution of radiation given by Eq. (32), the Coulomb bound-state wave functions are written in the form<sup>19</sup>

$$\psi_{n}(\mathbf{\hat{x}}) = \begin{pmatrix} g_{n}(x)\chi_{\kappa}^{\mu}(\hat{x}) \\ if_{n}(x)\chi_{-\kappa}^{\mu}(\hat{x}) \end{pmatrix},$$
(34)

where  $g_n(x)$  and  $f_n(x)$  are the large and small radial wave functions and  $\chi^{\mu}_{\kappa}(\hat{x})$  is the two-component spin-angle function. The matrix element in Eq. (33) is the sum

$$A_{\mu_{f}\mu} = M^{a}_{\mu_{f}\mu} + \eta M^{b}_{\mu_{f}\mu} - \rho M^{c}_{\mu_{f}\mu} , \qquad (35)$$

in which

$$M^{a}_{\mu_{f}\mu} = \int d^{3}\mathbf{\tilde{x}} \psi^{\mu_{f}\dagger}_{1S}(\mathbf{\tilde{x}}) \mathbf{\tilde{\alpha}} \cdot \hat{\epsilon}_{\lambda} e^{-i\mathbf{\tilde{k}} \cdot \mathbf{\tilde{x}}} \psi^{\mu}_{S}(\mathbf{\tilde{x}}) , \qquad (36a)$$

$$M^{b}_{\mu_{f}\mu} = \int d^{3}\vec{\mathbf{x}} \psi^{\mu_{f}\dagger}_{1S}(\vec{\mathbf{x}})\vec{\alpha} \cdot \hat{\epsilon}_{\lambda} e^{-i\vec{\mathbf{k}}\cdot\vec{\mathbf{x}}}\psi^{\mu}_{\mu}(\vec{\mathbf{x}}) V^{\mu}_{\rho_{S}}, \quad (36b)$$

$$M^{c}_{\mu_{f}\mu} = \int d^{3}\mathbf{\hat{x}} \,\psi^{\mu_{f}\dagger}_{\mathbf{1}s}(\mathbf{\hat{x}}) \mathbf{\hat{\alpha}} \cdot \hat{\epsilon}_{\lambda} \,e^{-i\mathbf{\hat{k}}\cdot\mathbf{\hat{x}}} \psi^{\mu}_{q}(\mathbf{\hat{x}}) \,V^{\mu}_{qs} \,, \quad (36c)$$

with  $\eta = (S + \frac{1}{2}i\Gamma_p)^{-1}$  and  $\rho = (\Delta E - S - \frac{1}{2}i\Gamma_q)^{-1}$ . The matrix elements  $V_{\rho s}^{\mu}$  and  $V_{q s}^{\mu}$  are given by

$$V_{\rho s}^{\mu} = \int d^{3} \vec{\mathbf{x}} \psi_{\rho}^{\mu \dagger} (\vec{\mathbf{x}}) e \vec{\mathbf{E}} \cdot \vec{\mathbf{x}} \psi_{s}^{\mu} (\vec{\mathbf{x}}) , \qquad (37a)$$

$$V_{qs}^{\mu} = \int d^{3}\mathbf{\hat{x}} \,\psi_{q}^{\mu\,\dagger}(\mathbf{\hat{x}}) e\,\mathbf{\vec{E}}\cdot\mathbf{\hat{x}} \,\psi_{s}^{\mu}(\mathbf{\hat{x}}) \,. \tag{37b}$$

Integration over coordinate angles in Eqs. (36) and (37) yields

$$M^{a}_{\mu_{f}\mu} = -\langle \mu_{f} | \vec{\sigma} \cdot \hat{\epsilon}_{\lambda} \vec{\sigma} \cdot \hat{k} I_{M1}(k) | \mu \rangle , \qquad (38a)$$

$$M^{b}_{\mu f \mu} = i \langle \mu_{f} | \vec{\sigma} \cdot \hat{\epsilon}_{\lambda} \vec{\sigma} \cdot \hat{z} J_{E1}(k) | \mu \rangle V^{(1/2)}_{ps}, \qquad (38b)$$

$$M^{c}_{\mu f \mu} = -(i/\sqrt{2}) \langle \mu_{f} | (\vec{\sigma} \cdot \hat{\epsilon}_{\lambda} \vec{\sigma} \cdot \hat{z} - 3\hat{\epsilon}_{\lambda} \cdot \hat{z}) K_{E1}(k)$$

$$+ (\vec{\sigma} \cdot \hat{k} \vec{\sigma} \cdot \hat{\epsilon}_{\lambda} \vec{\sigma} \cdot \hat{z} \vec{\sigma} \cdot \hat{k} - \hat{\epsilon}_{\lambda} \cdot \hat{z}) \sqrt{3} K_{M2}(k) | \mu \rangle V^{(1/2)}_{qs} \qquad (38c)$$

where  $\sigma_x$ ,  $\sigma_y$ , and  $\sigma_z$  are the Pauli matrices and  $|\mu\rangle$  is a two-component spinor with upper and lower components  $\frac{1}{2} + \mu$  and  $\frac{1}{2} - \mu$ , respectively, with  $\mu = \pm \frac{1}{2}$ . The functions  $I_{M1}$ ,  $J_{E1}$ ,  $K_{E1}$ , and  $K_{M2}$  in Eq. (38), associated with magnetic dipole (M1), electric dipole (E1), and magnetic quadrupole (M2) radiation,<sup>20</sup> are

$$I_{M1}(k) = \int_{0}^{\infty} dx \, x^{2} [f_{1s}(x)g_{s}(x) + g_{1s}(x)f_{s}(x)]j_{1}(kx) , \qquad (39a)$$

$$J_{E1}(k) = \int_{0}^{\infty} dx \, x^{2} [f_{1s}(x)g_{p}(x) \\ \times [\frac{1}{3}j_{0}(kx) - \frac{2}{3}j_{2}(kx)] + g_{1s}(x)f_{p}(x)j_{0}(kx)] ,$$
(39b)

$$K_{E1}(k) = \int_{0}^{\infty} dx \, x^{2} [f_{1S}(x)g_{q}(x) \\ \times [\frac{2}{3}j_{0}(kx) + \frac{1}{6}j_{2}(kx)] - g_{1S}(x)f_{q}(x)\frac{1}{2}j_{2}(kx)] ,$$
(39c)

 $K_{M2}(k)$ 

$$=\frac{\sqrt{3}}{2}\int_{0}^{\infty}dx\,x^{2}[f_{1s}(x)g_{q}(x)+g_{1s}(x)f_{q}(x)]j_{2}(kx)\,.$$
(39d)

In Eq. (39),  $j_0$ ,  $j_1$ , and  $j_2$  are the spherical Bessel

functions. The radial integrals in Eq. (39) are the same as those which appear in the lowestorder (in  $\alpha$ ) field-free one-photon transition rates to the 1S state<sup>21</sup>:

$$A_{M1}(2S_{1/2}) = 4\alpha k_1 I_{M1}^2(k_1) , \qquad (40a)$$

$$A_{E1}(2P_{1/2}) = 4\alpha k_1 J_{E1}^2(k_1) , \qquad (40b)$$

$$A_{E1}(2P_{3/2}) + A_{M2}(2P_{3/2})$$
  
=  $4\alpha k_2 [K_{E1}^2(k_2) + K_{M2}^2(k_2)],$  (40c)

with  $k_1 = E(2S_{1/2}) - E(1S_{1/2}) = E(2P_{1/2}) - E(1S_{1/2})$  and  $k_2 = E(2P_{3/2}) - E(1S_{1/2})$ . The electric field perturbation matrix elements  $V_{ps}^{(1/2)}$  and  $V_{qs}^{(1/2)}$  are

$$V_{ps}^{(1/2)} = -eE\frac{1}{3} \int_0^\infty dx \, x^{3}[g_p(x)g_s(x) + f_p(x)f_s(x)] \,, \quad (41a)$$

$$V_{qs}^{(1/2)} = eE\frac{\sqrt{2}}{3} \int_0^\infty dx \, x^3 [g_q(x)g_s(x) + f_q(x)f_s(x)] \,. \tag{41b}$$

Numerical evaluation of the radial integrals in Eqs. (39) and (41), at  $k = k_1$ , yields

$$I_{M1} = [(Z\alpha)^4/3\sqrt[3]{2}][1+0.4193(Z\alpha)^2F_1(Z\alpha)],$$

(42a)

$$J_{E1} = -(\frac{2}{3})^{9/2} (Z\alpha) [1 - 0.0557 (Z\alpha)^2 F_2(Z\alpha)], \quad (42b)$$
  

$$K_{E1} = -(\frac{2}{3})^{9/2} (Z\alpha) [1 - 0.2716 (Z\alpha)^2 F_3(Z\alpha)], \quad (42c)$$

$$K_{M2} = -[(Z\alpha)^3/3\sqrt{2}][1 - 0.1821(Z\alpha)^2F_4(Z\alpha)],$$

(42d)

$$V_{ps}^{(1/2)} = -eE[\sqrt{3}/(Z\alpha)][1 - 0.4167(Z\alpha)^2F_5(Z\alpha)],$$
(42e)

$$V_{qs}^{(1/2)} = -eE[\sqrt{6}/(Z\alpha)][1 - 0.1667(Z\alpha)^2F_6(Z\alpha)],$$
(42f)

where the functions  $F_i(\mathbb{Z}\alpha)$ , i=1 to 6, are shown in Figs. 4 and 5. Carrying out the indicated summations in Eq. (32), we obtain

$$\frac{1}{4} \sum_{\lambda \mu_{f} \mu} |A_{\mu_{f} \mu}|^{2} = I_{M1}^{2} + |\eta|^{2} J_{E1}^{2} (V_{\rho s}^{(1/2)})^{2} + |\rho|^{2} (K_{E1}^{2} + K_{M2}^{2}) (V_{q s}^{(1/2)})^{2} \\ + [2 \operatorname{Im}(\eta) I_{M1} J_{E1} V_{\rho s}^{(1/2)} + \sqrt{2} \operatorname{Im}(\rho) I_{M1} (K_{E1} - \sqrt{3} K_{M2}) V_{q s}^{(1/2)}] P_{1}(\hat{k} \cdot \hat{z}) \\ + [\sqrt{2} \operatorname{Re}(\eta * \rho) J_{E1} (K_{E1} - \sqrt{3} K_{M2}) V_{\rho s}^{(1/2)} V_{q s}^{(1/2)} - \frac{1}{2} |\rho|^{2} (K_{E1}^{2} + 2\sqrt{3} K_{E1} K_{M2} - K_{M2}^{2}) (V_{q s}^{(1/2)})^{2}] P_{2}(\hat{k} \cdot \hat{z}) ,$$
(43)

where  $P_1$  and  $P_2$  are Legendre polynomials. The one-photon differential decay rate is

$$\frac{dR_{1\gamma}}{d\Omega_{k}} = \frac{\alpha k_{1}}{4\pi} \sum_{\lambda,\mu_{f}\mu} |A_{\mu_{f}\mu}|^{2}$$
(44)

and the integrated decay rate is

$$R_{1\gamma} = \int d\Omega_k \left( \frac{dR_{1\gamma}}{d\Omega_k} \right)$$
  
=  $4\alpha k_1 [I_{M1}^2 + |\eta|^2 J_{E1}^2 (V_{\rho s}^{(1/2)})^2$   
+  $|\rho|^2 (K_{E1}^2 + K_{M2}^2) (V_{\sigma s}^{(1/2)})^2],$  (45)



FIG. 4. Graph of the functions  $F_1(Z\alpha)$ ,  $F_2(Z\alpha)$ , and  $F_6(Z\alpha)$  which appear in Eqs. (42a), (42b), and (42f).

where the normalization has been determined by comparison of Eqs. (45) and (29) via (40). The one-photon decay rate in Eq. (45) differs slightly from the one-photon portion of Eq. (29), because the energy k and radial integals  $K_{E1}$  and  $K_{M2}$  in Eq. (45) are evaluated at  $k = k_1$ , whereas the definition of  $\Gamma_q$  in Eq. (29) corresponds to evaluation at  $k = k_2$ . The expression in Eq. (45) should be more accurate, but the difference is negligible for most applications.

The lifetime of the 2S state is the inverse of the sum of the field-free two-photon decay rate  $R_{2\gamma}$ and the one-photon decay rate  $R_{1\gamma}$  in Eq. (45):  $\tau_{2S} = (R_{2\gamma} + R_{1\gamma})^{-1}$ . The field-free decay rate  $R_{2\gamma}$ +  $4\alpha k_1 I_{M1}^2$  has been accurately evaluated by John- $\operatorname{son}^{22}$  over a wide range of Z; our numerical values for the one-photon portion  $4\alpha k_1 I_{M1}^2$  are in complete agreement with the corresponding values of Johnson. Drake and Lin have calculated the anisotropy for the electric field induced radiation from the 2S state including relativistic and hyperfine-structure corrections<sup>23</sup> for Z up to 16. They consider E1 radiation which, for zero-spin nuclei, corresponds to the terms  $J_{E1}$  and  $K_{E1}$  in Eq. (43). For these terms, i.e., with  $I_{M1}$  and  $K_{M2}$  replaced by zero, Eq. (43) is in reasonable agreement with their calculated anisotropies. The magnetic dipole term  $I_{M1}$  can be expected to have a measurable effect on the angular distribution of radiation at high Z, as was indicated previously.<sup>24</sup> The magnetic quadrupole term  $K_{M2}$  reduces the predicted anisotropy by a factor of relative order  $(Z\alpha)^2$ . Both  $I_{M1}$  and  $K_{M2}$  have a negligible effect at the current level of experimental accuracy at  $Z = 1.^{8,25}$ The dominant relativistic correction to the elec-



FIG. 5. Graph of the functions  $F_3(Z\alpha)$ ,  $F_4(Z\alpha)$ , and  $F_5(Z\alpha)$  which appear in Eqs. (42c)-(42e).

tric field induced decay rate in Eq. (45), a 1.4% reduction in  $(V_{p_s}^{(1/2)})^2$ , has been included in the analysis of the recent Lamb-shift measurement at Z = 18 by Gould and Marrus.<sup>6</sup> The relativistic correction in the dipole decay rate  $4\alpha k_1 J_{E1}^2$  is 0.2% at Z = 18.

The one-photon differential decay rate in Eq. (44) coincides with the result obtained by a golden rule calculation in which the effect of the electric field on the initial 2S state is taken into account in first-order perturbation theory with complex radiative level shifts included in the energy denominators. This prescription is modified for photon absorption, because the energy denominators in the right-hand factors in Eq. (18) are not complex conjugates of the energy denominators in the lefthand factors. The connection between the form of the electron propagation function and the angular distribution in resonant photon scattering from an atom in an electric field is illustrated in Appendix C.

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# APPENDIX A

In this appendix, we calculate the first-order correction to  $S_{\mathbf{r}}^{(0)}$  from the perturbation

 $\delta U_{nm}(\omega) = \sum_{nm}(\omega) + V_{nm} - \tilde{\sum}_{nm}(\omega) - \tilde{V}_{nm}.$ The function  $S'_{F}$  is the solution of

 $S'_F = S_F - iS_F US'_F . \tag{A1}$ 

If we write  $U = U^{(0)} + \Delta U$ , then  $S'_F$  is given by

$$S'_{F} = S_{F}^{(0)} - iS_{F}^{(0)} \Delta US'_{F} , \qquad (A2)$$

where

. .

$$S_F^{(0)} = S_F - iS_F U^{(0)} S_F^{(0)}.$$
(A3)

This is readily verified by employing the relation  $S_F^{(0)} = (1 + iS_F U^{(0)})^{-1}S_F$  from Eq. (A3) to eliminate  $S_F^{(0)}$  in Eq. (A2).

Let  $U_{nm}^{(0)}(\omega)$  and  $\Delta U_{nm}(\omega)$  be the functions corresponding to  $U^{(0)}$  and  $\Delta U$  according to Eq. (6), and let  $U_{nm}^{(0)}(\omega) = \tilde{\Sigma}_{nm}(\omega) + \tilde{V}_{nm}$ . Then Eq. (A3) is equivalent to Eq. (14), which has known solutions  $g_{nm}(\omega)$ , and the equation for the functions  $f_{nm}(\omega)$ , which follows from (A2), is

$$f_{nm}(\omega) = g_{nm}(\omega) - \sum_{ij} g_{ni}(\omega) \Delta U_{ij}(\omega) f_{jm}(\omega) .$$
 (A4)

The first-order correction  $f'_{nm}(\omega)$  to  $g_{nm}(\omega)$ , due to the perturbation  $\delta U_{nm}(\omega)$ , is thus

$$f'_{nm}(\omega) = -\sum_{ij} g_{ni}(\omega) \delta U_{ij}(\omega) g_{jm}(\omega) , \qquad (A5)$$

which yields

$f'_{nm}(\omega)=0,$		for $n, m \in S$ ,
$f'_{nm}(\omega) = -\frac{1}{E_m - \omega} \sum_{i \in S}$	$g_{ni}(\omega)\delta U_{im}(\omega),$	for $n \in S$ , $m \notin S$ ,
$f'_{nm}(\omega) = -\frac{1}{E_n - \omega} \sum_{j \in S}$	$\delta U_{nj}(\boldsymbol{\omega})g_{jm}(\boldsymbol{\omega}),$	for $n \in S$ , $m \in S$
$f_{nm}'(\omega) = -\delta U_{nm}(\omega)/(E_{nm})$	$(E_m - \omega)$ ,	for $n, m \notin S$ .
		(A6)

For the 2S resonance, these corrections are smaller than the leading terms by a factor of order  $\delta U_{ii}(\omega)/(E_r-\omega)$ , with  $r \in \$$ .

#### APPENDIX B

If the mechanism which excites the metastable 2S state introduces a spin polarization, there is, of course, additional angular variation in the intensity of the decay radiation. This effect may be useful in certain experiments with hydrogenlike atoms,  $^{26}$  so an evaluation of the angular distribution is given here.

Without specifying the details of the excitation mechanism, we can account for spin effects by employing a density matrix

$$\sum_{\mu_i} B_{\mu\mu_i} B_{\mu'\mu_i}^* = b \langle \mu | \frac{1}{2} (1 + \vec{\sigma} \cdot \vec{\mathbf{P}}) | \mu' \rangle , \qquad (B1)$$

where b is a positive real number and  $\vec{P}$  is the polarization vector, with  $|\vec{P}| \leq 1$ , for the 2S resonance. The magnitude of  $\vec{P}$  corresponds to the degree of polarization with  $|\vec{P}|=0$  for no polarization and  $|\vec{P}|=1$  for complete polarization. The angular distribution of the decay radiation is then

$$\sum_{\lambda \mu_{f} \mu_{i}} \left| \sum_{\mu} A_{\mu_{f} \mu} B_{\mu \mu_{i}} \right|^{2}$$
$$= \frac{1}{2} b \left( \sum_{\lambda \mu_{f} \mu} |A_{\mu_{f} \mu}|^{2} + \sum_{\substack{\lambda \mu_{f} \\ \mu \mu'}} A_{\mu_{f} \mu} A_{\mu_{f} \mu}^{*} |A_{\mu_{f} \mu'} \rangle \right).$$
(B2)

The first term on the right-hand side of Eq. (B2) is given by Eq. (43), and the additional term is

$$\sum_{\substack{\lambda \mu \\ \mu \mu^{l}}} A_{\mu_{f}\mu} A_{\mu_{f}\mu'}^{*} \langle \mu | \vec{\sigma} \cdot \vec{\mathbf{p}} | \mu' \rangle = [8 \operatorname{Re}(\eta) I_{M1} J_{E1} V_{Ps}^{(1/2)} - 2\sqrt{2} \operatorname{Re}(\rho) I_{M1} (K_{E1} - \sqrt{3} K_{M2}) V_{qs}^{(1/2)}] \vec{\mathbf{p}} \cdot \hat{k} \times \hat{z} + 6\sqrt{2} \operatorname{Im}(\eta * \rho) J_{E1} (K_{E1} - \sqrt{3} K_{M2}) V_{Ps}^{(1/2)} V_{qs}^{(1/2)} \hat{k} \cdot \hat{z} \, \vec{\mathbf{p}} \cdot \hat{k} \times \hat{z}$$
(B3)

The term proportional to  $\hat{k} \cdot \hat{z} \vec{P} \cdot \hat{k} \times \hat{z}$  should be measurable at low Z.

#### APPENDIX C

Here we examine elastic resonant scattering of a photon by a hydrogenlike atom in the ground state in an electric field. We simplify the calculation, while still retaining the essential features, by restricting our attention to the narrow 2S resonant contribution and considering only mixing of the  $2S_{1/2}$  and  $2P_{1/2}$  states.

The matrix element for photon scattering is

$$\begin{split} M' &= \frac{2\pi e^2}{(k_i k_f)^{1/2}} \int d^4 x_2 \int d^4 x_1 \, \overline{\psi} \, {}^{\mu}_{1S}(x_2) \, \overline{\gamma} \cdot \widehat{\epsilon}^f_{\lambda} \\ &\times e^{i k_f \cdot x_2} S'_F(x_2, x_1) \overline{\gamma} \cdot \widehat{\epsilon}^i_v \, e^{-i k_i \cdot x_1} \psi^{\mu}_{1S}(x_1) \,, \end{split}$$

(C1)

with the resonant contribution

(C6)

$$M'_{2S} \approx -i \frac{(2\pi e)^2}{(k_i k_f)^{1/2}} \delta(k_f - k_i) \frac{1}{E''_s - E_{1S} - k_i} \frac{E''_s - E'_p}{E''_s - E''_p}$$
$$\times \sum_{\mu = \pm 1/2} C_{\mu_f \mu} D_{\mu \mu_i} , \qquad (C2)$$

where

$$\begin{split} C_{\mu_{f}\mu} &= \int d^{3} \mathbf{\tilde{x}} \, \psi_{1s}^{\mu_{f}\dagger} \left( \mathbf{\tilde{x}} \right) \mathbf{\tilde{\alpha}} \cdot \hat{\epsilon}_{\lambda}^{f} \\ &\times e^{-i \mathbf{\tilde{k}}_{f} \cdot \mathbf{\tilde{x}}} \left( \psi_{s}^{\mu} \left( \mathbf{\tilde{x}} \right) + \frac{V_{ps}^{\mu}}{E_{s}^{\prime\prime} - E_{p}^{\prime\prime}} \psi_{p}^{\mu} \left( \mathbf{\tilde{x}} \right) \right) \\ &\approx M_{\mu_{f}\mu}^{af} + \eta M_{\mu_{f}\mu}^{bf} \end{split} \tag{C3}$$

and

$$D_{\mu \mu_{i}} = \int d^{3} \mathbf{\hat{x}} \left( \psi_{s}^{\mu \dagger} (\mathbf{\hat{x}}) + \frac{V_{sp}^{\mu}}{E_{s}'' - E_{p}'} \psi_{p}^{\mu \dagger} (\mathbf{\hat{x}}) \right) \mathbf{\hat{\alpha}} \cdot \hat{\epsilon}_{v}^{i}$$

$$\times e^{i \mathbf{\hat{k}}_{i}} \cdot \mathbf{\hat{x}} \psi_{is}^{\mu} (\mathbf{\hat{x}})$$

$$\approx M_{\mu_{i},\mu}^{ai} + \eta M_{\mu_{i},\mu}^{bi}. \qquad (C4)$$

Here we have approximated  $(E''_s - E'_p)^{-1}$  by  $\eta = (S + \frac{1}{2}i\Gamma_p)^{-1}$ . In Eqs. (C3) and (C4), the *M*'s are those given in Eqs. (36a) and (36b) with the appropriate labels added to the vectors  $\hat{k}$  and  $\hat{\epsilon}$ . The matrix element squared  $|M'_{2S}|^2$ , averaged over initial and summed over final atomic angular momentum projections and photon polarizations, is proportional to

$$\sum_{\lambda\nu\mu_{f}}\sum_{\mu_{i}}\left|\sum_{\mu}C_{\mu_{f}\mu}D_{\mu\mu_{i}}\right|^{2} = \sum_{\mu\mu'}\left(\sum_{\lambda\mu_{f}}C_{\mu_{f}\mu}C_{\mu_{f}\mu'}\right)\left(\sum_{\nu\mu_{i}}D_{\mu\mu_{i}}D_{\mu'\mu_{i}}^{*}\right).$$
(C5)

**Evaluation yields** 

$$\begin{split} \sum_{\lambda \mu_{f}} C_{\mu_{f} \ \mu} C_{\mu_{f} \mu}^{*} C_{\mu_{f} \mu'} \\ &= 2 \langle \mu' | I_{M1}^{2} + | \eta |^{2} J_{E1}^{2} (V_{ps}^{(1/2)})^{2} \\ &+ 2 I_{M1} J_{E1} V_{ps}^{(1/2)} [\operatorname{Re}(\eta) \widehat{\sigma} \cdot \widehat{k}_{f} \times \widehat{z} + \operatorname{Im}(\eta) \widehat{k}_{f} \cdot \widehat{z} ] | \mu \rangle \end{split}$$

and  

$$\sum_{\nu \mu_{i}} D_{\mu \mu_{i}} D_{\mu' \mu_{i}}^{*} D_{\mu' \mu_{i}}^{*}$$

$$= 2 \langle \mu | I_{M1}^{2} + |\eta|^{2} J_{E1}^{2} (V_{ps}^{(1/2)})^{2}$$

$$+ 2 I_{M1} J_{E1} V_{ps}^{(1/2)} [\operatorname{Re}(\eta) \hat{\sigma} \cdot \hat{k}_{i} \times \hat{z} - \operatorname{Im}(\eta) \hat{k}_{i} \cdot \hat{z}] | \mu' \rangle ,$$
(C7)

with  $I_{M_1}$  and  $J_{E_1}$  evaluated at  $k_f = k_i$ . Hence,

$$\sum_{\lambda\nu\mu_{f}\mu_{i}} \left| \sum_{\mu} C_{\mu_{f}\mu} D_{\mu\mu_{i}} \right|^{2} \\ = 8 [I_{M1}^{2} + \left| \eta \right|^{2} J_{E1}^{2} (V_{ps}^{(1/2)})^{2} + 2I_{M1} J_{E1} V_{ps}^{(1/2)} \operatorname{Im}(\eta) \hat{k}_{f} \cdot \hat{z} ] [I_{M1}^{2} + \left| \eta \right|^{2} J_{E1}^{2} (V_{ps}^{(1/2)})^{2} - 2I_{M1} J_{E1} V_{ps}^{(1/2)} \operatorname{Im}(\eta) \hat{k}_{i} \cdot \hat{z} ] \\ + 32 [I_{M1} J_{E1} V_{ps}^{(1/2)} \operatorname{Re}(\eta) f^{2} (\hat{k}_{f} \times \hat{z}) \cdot (\hat{k}_{i} \times \hat{z}) .$$
(C8)

The right-hand side of Eq. (C8) is clearly invariant under time reversal, i.e., under the replacement  $\vec{k}_i - \vec{k}_f$  and  $\vec{k}_f - \vec{k}_i$ .

The angular distribution for photon absorption is obtained by summing over directions for the emitted photon:

$$\int d\Omega_{k_{f}} \sum_{\lambda \nu \mu_{f} \mu_{i}} \left| \sum_{\mu} C_{\mu_{f} \mu} D_{\mu \mu_{i}} \right|^{2} = 32 \pi [I_{M1}^{2} + |\eta|^{2} J_{E1}^{2} (V_{ps}^{(1/2)})^{2}] [I_{M1}^{2} + |\eta|^{2} J_{E1}^{2} (V_{ps}^{(1/2)})^{2} - 2I_{M1} J_{E1} V_{ps}^{(1/2)} \operatorname{Im}(\eta) \hat{k}_{i} \cdot \hat{z}].$$
(C9)

The analogous angular distribution for photon emission, averaged over directions of the absorbed photon, is

$$\frac{1}{4\pi} \int d\Omega_{k_{i}} \sum_{\lambda \nu \mu_{f} \mu_{i}} \left| \sum_{\mu} C_{\mu_{f} \mu} D_{\mu \mu_{i}} \right|^{2} = 8 [I_{M1}^{2} + |\eta|^{2} J_{E1}^{2} (V_{\rho s}^{(1/2)})^{2}] [I_{M1}^{2} + |\eta|^{2} J_{E1}^{2} (V_{\rho s}^{(1/2)})^{2} + 2 I_{M1} J_{E1} V_{\rho s}^{(1/2)} \operatorname{Im}(\eta) \hat{k}_{f} \cdot \hat{z}].$$
(C10)

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 $(\vec{\sigma} \cdot \hat{k} \vec{\sigma} \cdot \hat{\epsilon}_{\lambda} \vec{\sigma} \cdot \hat{z} \vec{\sigma} \cdot \hat{k} - \hat{\epsilon}_{\lambda} \cdot \hat{z}) = i \vec{\sigma} \cdot (\hat{k} \times \hat{\epsilon}_{\lambda} \hat{k} + \hat{k} \hat{k} \times \hat{\epsilon}_{\lambda}) \cdot \hat{z}$ 

for the coefficient in Eq. (38c).

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