Linear response of a nonrelativistic hydrogenlike atom to a single-mode radiation field. I. Exact theory: The atomic ground state

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In the framework of semiclassical theory we investigate the influence of a low-intensity monochromatic electromagnetic plane wave on a nonrelativistic one-electron atom. The radiation field is switched on adiabatically, while the atom is assumed to be initially in its ground state. We analyze their interaction to the first order of perturbation theory, taking into account retardation effects. In the radiation gauge, the exact first-order correction to the wave function consists of separate orbital and spin contributions which are determined, respectively, by a vector and a scalar linear-response function. Starting from Hostler's integral representation of the Schrödinger Coulomb Green function in coordinate space, we have derived them, via a generating function, as closed-form contour integrals. Then they have also been written explicitly, as double power series involving linear combinations of Humbert hypergeometric functions Φ_1. From the integral representation of the linear-response wave function we have extracted the considerably simpler second-order retardation approach. We have subsequently translated it in a conveniently modified Poincaré gauge, which we call a multipolar gauge, in order to display the contributions of the genuine field-atom multipole couplings. The relevant orbital and spin multipole terms are then recovered by employing directly the generating function of the linear response. Their low- and high-frequency behavior is finally examined. [S1050-2947(97)03711-6]

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I. INTRODUCTION

The linear response of an atom to a classical radiation field is described by the first-order perturbation correction $\Psi^{(1)}$ to an initial bound-state wave function. Obviously, analytic expressions of such corrections cannot be even envisaged for complex atoms. On the contrary, obtaining in analytic form the linear response of a nonrelativistic hydrogen atom, initially in a $|nlm\rangle$ state, to a single-mode radiation field is a problem raised by Podolsky [1] soon after the advent of quantum mechanics. However, Podolsky solved this problem only for the atomic ground state and in the electric dipole approximation (EDA). In fact, he succeeded in getting the Coulomb Sturmian-function expansion of the corresponding first-order correction, by solving appropriate inhomogeneous differential equations [1]. Much later, the same correction has been found in a compact form, as an integral representation, and also explicitly [2,3].

The purpose of this paper is to evaluate the linear response of a nonrelativistic hydrogenic atom to a single-mode radiation field, when the initial atomic state is the ground state. We take full account of retardation and, in addition, consider the magnetic coupling of the electron spin. The starting point of our analytic developments is a compact integral representation of the Coulomb Green function (CGF) in coordinate space discovered by Hostler [4].

The resulting exact formulas being rather complicated and difficult to handle, we find it valuable to write them in the second-order approximation of retardation which is much simpler and also reliable in the optical regime. We have obtained approximate formulas in the radiation gauge, as well as in a modified Poincaré gauge that has the virtue of displaying the constituent multipole terms.

In Sec. II we show that the wave-function correction in the radiation gauge can be expressed in terms of a vector function $\boldsymbol{W}_{100}$ and a scalar one $\mathcal{S}_{100}$ that we call linear-response functions (LRF's). Section III is devoted to the derivation of the LRF's starting from a suitable generating function $\mathcal{F}$, determined in turn by the CGF. The functions $\boldsymbol{W}_{100}$ and $\mathcal{S}_{100}$ are calculated first as compact contour integrals and then explicitly as double series involving Humbert functions Φ_1. In Sec. IV we deal with the second-order retardation approach to the linear-response wave function. After taking it out in the radiation gauge, we write it also in the Poincaré gauge by applying the Power-Zienau-Woolley (PZW) transformation [5–7]. By means of an additional $U(1)$ transformation, we find the linear response in what we call a *multipolar gauge*. Precisely in this gauge the multipole structure of the first-order Pauli Hamiltonian, truncated to a second-order retardation approach, is laid out in Sec. V. We take advantage of the generating function $\mathcal{F}$ in Sec. VI to derive directly the relevant orbital and spin multipole terms entering the linear-response wave function in the same approximation. They are written both as integrals and in explicit form. The latter is employed in Sec. VII to evaluate their low- and high-frequency behavior. Section VIII surveys the results with a view to possible applications. In Appendix A we get the generating function $\mathcal{F}$ as a contour integral by making use of Hostler' s integral representation of the Schrödinger CGF in coordinate space. Appendix B is meant to be a brief review of some useful expressions of the Humbert hypergeometric function Φ_1. Note that the Gaussian units and Einstein's summation convention for Cartesian indices are consistently utilized.

II. FIRST-ORDER CORRECTION TO THE WAVE FUNCTION IN THE RADIATION GAUGE

We concern ourselves with a hydrogenlike atom whose nucleus is fixed at the origin of the coordinate system and has the atomic number Z. The perturbing radiation field is a monochromatic plane wave with propagation vector $\boldsymbol{\kappa}$, frequency ω, and polarization $\hat{\boldsymbol{\epsilon}}$:

$$\kappa=\frac{\omega}{c}, \quad \hat{\boldsymbol{\epsilon}}\cdot\boldsymbol{\kappa}=0. \tag{2.1}$$

Its electrodynamic potentials in the radiation gauge are

$$\Phi=0 \tag{2.2}$$

and

$$\boldsymbol{A}(\boldsymbol{r},t)=\frac{c}{\omega}\frac{1}{2i}\mathcal{E}_0\{\exp[i(\boldsymbol{\kappa}\cdot\boldsymbol{r}-\omega t)]\hat{\boldsymbol{\epsilon}}-\text{c.c.}\} \quad (\mathcal{E}_0>0), \tag{2.3}$$

with

$$\text{div}\boldsymbol{A}=0. \tag{2.4}$$

Note that the field strengths of the mode,

$$\boldsymbol{E}(\boldsymbol{r},t)=\frac{1}{2}\mathcal{E}_0\{\exp[i(\boldsymbol{\kappa}\cdot\boldsymbol{r}-\omega t)]\hat{\boldsymbol{\epsilon}}+\text{c.c.}\} \tag{2.5}$$

and

$$\boldsymbol{B}(\boldsymbol{r},t)=\frac{1}{2}\frac{c}{\omega}\mathcal{E}_0\{\exp[i(\boldsymbol{\kappa}\cdot\boldsymbol{r}-\omega t)](\boldsymbol{\kappa}\times\hat{\boldsymbol{\epsilon}})+\text{c.c.}\}, \tag{2.6}$$

have small amplitude and arbitrary frequency. As a side remark, in Eqs. (2.5) and (2.6), $\mathcal{E}_0$ is the amplitude of the oscillating field only in the particular case when its polarization is linear ($\hat{\boldsymbol{\epsilon}}^*=\hat{\boldsymbol{\epsilon}}$). However, in any case, $\frac{1}{2}\mathcal{E}_0^2$ is the time average of the squared field strength.

Dealing only with the atomic *linear* response, we are entitled to neglect the $\boldsymbol{A}^2$ term in the field-atom interaction Hamiltonian and keep only its terms that are linear in the field:

$$H^{(1)}=\frac{e}{m_e c}(\boldsymbol{A}\cdot\boldsymbol{P}+\boldsymbol{B}\cdot\boldsymbol{S}). \tag{2.7}$$

In Eq. (2.7), $-e$ is the electron charge, m_e the electron mass, while $\boldsymbol{P}$ and $\boldsymbol{S}=(\hbar/2)\boldsymbol{\sigma}$ denote the momentum and spin operators of the electron, respectively. The first term in the Hamiltonian (2.7) is due to the orbital motion of the electron, while the second one describes the magnetic coupling of the electron spin.

We shift the initial condition to $t\to-\infty$ and suppose that the field mode (2.5) and (2.6) is turned on adiabatically, with an exponential switching factor in the time interval $(-\infty,0)$. This means that in calculations, for negative times, one should multiply the interaction Hamiltonian (2.7) by a factor $\exp[(1/\hbar)\varepsilon t)]$ with $\varepsilon>0$ and eventually take $\varepsilon\to+0$. In the remote past ($t\to-\infty$), the atomic electron is assumed to be in its ground state, described by the unperturbed wave function

$$\Psi^{(0)}_{100m_s}(\boldsymbol{r},t)=\exp\left(-\frac{i}{\hbar}E_1 t\right)u_{100}(\boldsymbol{r})\zeta_{m_s}. \tag{2.8}$$

In Eq. (2.8), E_1 is the ground-state energy,

$$E_1=-\frac{Ze^2}{2a}, \tag{2.9}$$

with

$$a=\frac{\hbar^2}{m_e e^2 Z} \tag{2.10}$$

the scaled Bohr radius; $u_{100}(\boldsymbol{r})$ is the corresponding eigenfunction,

$$u_{100}(\boldsymbol{r})=(\pi a^3)^{-1/2}\exp\left(-\frac{r}{a}\right), \tag{2.11}$$

and ζ_{m_s} denotes a normalized eigenspinor of the Pauli operator σ_z:

$$\sigma_z\zeta_{m_s}=2m_s\zeta_{m_s} \quad (m_s=\pm\tfrac{1}{2}). \tag{2.12}$$

According to time-dependent perturbation theory, the first-order correction to the wave function (2.8) due to the interaction (2.7) is, for $t\geqslant 0$,

$$\begin{aligned}\Psi^{(1)}_{100m_s}(\boldsymbol{r},t)=&-\frac{e\mathcal{E}_0}{m_e\omega}\frac{1}{2i}\exp\left(-\frac{i}{\hbar}E_1 t\right)\\&\times\{e^{-i\omega t}[\hat{\boldsymbol{\epsilon}}\cdot\boldsymbol{W}_{100}(\Omega_1,\hbar\boldsymbol{\kappa};\boldsymbol{r})\\&+i\mathcal{S}_{100}(\Omega_1,\hbar\boldsymbol{\kappa};\boldsymbol{r})(\boldsymbol{\kappa}\times\hat{\boldsymbol{\epsilon}})\cdot\boldsymbol{S}]\\&-e^{i\omega t}[\hat{\boldsymbol{\epsilon}}^*\cdot\boldsymbol{W}_{100}(\Omega_2,-\hbar\boldsymbol{\kappa};\boldsymbol{r})\\&-i\mathcal{S}_{100}(\Omega_2,-\hbar\boldsymbol{\kappa};\boldsymbol{r})(\boldsymbol{\kappa}\times\hat{\boldsymbol{\epsilon}}^*)\cdot\boldsymbol{S}]\}\zeta_{m_s}.\end{aligned} \tag{2.13}$$

We have denoted

$$\Omega_1=E_1+\hbar\omega+i0, \quad \Omega_2=E_1-\hbar\omega, \tag{2.14}$$

$$\boldsymbol{W}_{100}(\Omega,\hbar\boldsymbol{\kappa};\boldsymbol{r})\equiv-\int d^3x'\,G(\Omega;\boldsymbol{r},\boldsymbol{r}')\exp(i\boldsymbol{\kappa}\cdot\boldsymbol{r}')\boldsymbol{P}'u_{100}(\boldsymbol{r}'), \tag{2.15}$$

and

$$\mathcal{S}_{100}(\Omega,\hbar\boldsymbol{\kappa};\boldsymbol{r})\equiv-\int d^3x'\,G(\Omega;\boldsymbol{r},\boldsymbol{r}')\exp(i\boldsymbol{\kappa}\cdot\mathbf{r}')u_{100}(\boldsymbol{r}'), \tag{2.16}$$

where $G(\Omega;\boldsymbol{r},\boldsymbol{r}')$ is the CGF, Eq. (A2). Notice that the linear-response correction (2.13) consists in fact of two Floquet terms, describing, respectively, one-photon absorption and emission processes. Owing to the additive structure of the Pauli Hamiltonian (2.7), each Floquet term is a sum of an orbital and a spin contribution. These contributions are characterized, respectively, by the vector LRF (2.15) and the scalar one, Eq. (2.16). It goes without saying that Eqs. (2.13)–(2.16) can be extended to an arbitrary stationary state $|Nm_s\rangle$, where N denotes the ensemble of quantum numbers associated to the orbital motion of the atomic electron.

III. EXACT EXPRESSIONS OF THE LINEAR-RESPONSE FUNCTIONS

According to Eqs. (2.11) and (A10), we find

$$\exp(i\boldsymbol{\kappa}\cdot\boldsymbol{r})\boldsymbol{P}u_{100}(\boldsymbol{r})=(\pi a^3)^{-1/2}\,\frac{\hbar^2}{a}\nabla_{\mathbf{q}}\mathcal{U}\Big|_{\{\mathbf{q}=\hbar\boldsymbol{\kappa},\ \lambda=\hbar/a\}}, \tag{3.1}$$

$$\exp(i\boldsymbol{\kappa}\cdot\boldsymbol{r})u_{100}(\boldsymbol{r})=-(\pi a^3)^{-1/2}\,\hbar\,\frac{\partial\mathcal{U}}{\partial\lambda}\Big|_{\{\mathbf{q}=\hbar\boldsymbol{\kappa},\ \lambda=\hbar/a\}}. \tag{3.2}$$

On account of the identities (3.1) and (3.2), the definitions (A9), (2.15), and (2.16) yield the similar formulas

$$\boldsymbol{W}_{100}(\Omega,\hbar\boldsymbol{\kappa};\boldsymbol{r})=(\pi a^3)^{-1/2}\,\frac{\hbar^2}{a}\nabla_{\mathbf{q}}\mathcal{F}(\boldsymbol{q},\lambda,\Omega;\boldsymbol{r})\Big|_{\{\mathbf{q}=\hbar\boldsymbol{\kappa},\ \lambda=\hbar/a\}}, \tag{3.3}$$

$$\mathcal{S}_{100}(\Omega,\hbar\boldsymbol{\kappa};\boldsymbol{r})=-(\pi a^3)^{-1/2}\,\hbar\,\frac{\partial}{\partial\lambda}\mathcal{F}(\boldsymbol{q},\lambda,\Omega;\boldsymbol{r})\Big|_{\{\mathbf{q}=\hbar\boldsymbol{\kappa},\ \lambda=\hbar/a\}}. \tag{3.4}$$

Substitution of the expressions (A14)–(A16) of the generating function $\mathcal{F}$ into Eqs. (3.3) and (3.4) gives the following closed-form representations:

$$\boldsymbol{W}_{100}(\Omega,\hbar\boldsymbol{\kappa};\boldsymbol{r})=(\pi a^3)^{-1/2}\,\frac{m_e}{X}\tau\frac{ie^{i\pi\tau}}{2\sin(\pi\tau)}\int_1^{(0+)}d\rho\rho^{-\tau}\left(\frac{2}{\mathcal{N}_{1,\tau+i\nu}}\right)^2\left(\frac{2}{\mathcal{N}_{1,\tau-i\nu}}\right)^2\Bigg\{i\rho\frac{1}{\tau}\frac{\boldsymbol{r}}{a}-2(1-\rho)\Bigg[\frac{1}{4}(1-\rho)+\frac{\rho(1/\tau)(r/a)}{\mathcal{N}_{1,\tau\mp i\nu}}\mp\rho(1-\rho)\frac{i(\kappa r\mp\boldsymbol{\kappa}\cdot\boldsymbol{r})}{\mathcal{N}_{1,\tau+i\nu}\mathcal{N}_{1,\tau-i\nu}}\Bigg]\boldsymbol{\nu}\Bigg\}\exp\left[\mp\frac{4i\rho(\kappa r\mp\boldsymbol{\kappa}\cdot\boldsymbol{r})}{\mathcal{N}_{1,\tau+i\nu}\mathcal{N}_{1,\tau-i\nu}}-\frac{\mathcal{N}_{\tau\mp i\nu,1}}{\mathcal{N}_{1,\tau\mp i\nu}}\frac{1}{\tau}\frac{r}{a}\right] \tag{3.5}$$

and

$$\mathcal{S}_{100}(\Omega,\hbar\boldsymbol{\kappa};\boldsymbol{r})=(\pi a^3)^{-1/2}\,\frac{2m_e}{X^2}\frac{ie^{i\pi\tau}}{2\sin(\pi\tau)}\int_1^{(0+)}d\rho\rho^{-\tau}\frac{4}{\mathcal{N}_{1,\tau+i\nu}\,\mathcal{N}_{1,\tau-i\nu}}\Bigg[\frac{1-\rho}{\mathcal{N}_{1,\tau\mp i\nu}}+\frac{\mp i\nu(1-\rho)^2+2\rho(1/\tau)(r/a)}{\mathcal{N}_{1,\tau-i\nu}\mathcal{N}_{1,\tau+i\nu}}+\frac{4i\rho(1-\rho)\boldsymbol{\kappa}\cdot\boldsymbol{r}}{\mathcal{N}_{1,\tau+i\nu}\,\mathcal{N}_{1,\tau-i\nu}\,\mathcal{N}_{1,\tau\mp i\nu}}-\frac{4\nu\rho(1-\rho)^2(\kappa r\mp\boldsymbol{\kappa}\cdot\boldsymbol{r})}{\mathcal{N}^2_{1,\tau+i\nu}\,\mathcal{N}^2_{1,\tau-i\nu}}\Bigg]\exp\left[\mp\frac{4i\rho(\kappa r\mp\boldsymbol{\kappa}\cdot\boldsymbol{r})}{\mathcal{N}_{1,\tau+i\nu}\,\mathcal{N}_{1,\tau-i\nu}}-\frac{\mathcal{N}_{\tau\mp i\nu,1}}{\mathcal{N}_{1,\tau\mp i\nu}}\frac{1}{\tau}\frac{r}{a}\right]. \tag{3.6}$$

In the above equations we utilize the parameters (A5) and (A6). Further, we have denoted

$$\mathcal{N}_{\tau',\tau}\equiv\tau'+\tau+(\tau'-\tau)\rho \tag{3.7}$$

and

$$\boldsymbol{\nu}\equiv\frac{\hbar\boldsymbol{\kappa}}{X},\qquad \nu\equiv\frac{\hbar\kappa}{X}, \tag{3.8}$$

Note also that one should employ consistently either upper or lower signs.

Using in Eqs. (3.5) and (3.6) the Taylor expansion of the first exponential and then the expansions of the negative powers of the ratio $\mathcal{N}_{1,\tau\pm i\nu}/\mathcal{N}_{1,\tau\mp i\nu}$, we reach the position to apply Eq. (B10). We finally get the explicit expressions of the LRF's (2.15) and (2.16) as double ascending power series, whose coefficients are linear combinations of Humbert functions Φ_1, Eq. (B5), of the dimensionless variables

$$\beta_\pm\equiv\frac{1}{2}(1-\tau\pm i\nu),\qquad \varrho_\pm\equiv 2\beta_\pm\frac{1}{\tau}\frac{r}{a}. \tag{3.9}$$

Therefore, we have found

$$\boldsymbol{W}_{100}(\Omega,\hbar\boldsymbol{\kappa};\boldsymbol{r})=(\pi a^3)^{-1/2}\ \frac{m_e}{X}\tau\left(\frac{2}{1+\tau\mp i\nu}\right)^{2+\tau}\exp\left(-\frac{1}{\tau}\frac{r}{a}\right)\sum_{\mu_1=0}^{\infty}\sum_{\mu_2=0}^{\infty}\frac{(\mu_1+\mu_2+1)!}{\mu_1!(\mu_1+1)!}\left[\mp\frac{2i(\kappa r\mp\boldsymbol{\kappa}\cdot\boldsymbol{r})}{1+\tau\mp i\nu}\right]^{\mu_1}$$
$$\times\left(\mp\frac{2i\nu}{1+\tau\mp i\nu}\right)^{\mu_2}\frac{\Gamma(\mu_1+2-\tau)}{\Gamma(\mu_1+\mu_2+3-\tau)}\left\{i\frac{1}{\tau}\frac{\boldsymbol{r}}{a}\Phi_1(\mu_1+2-\tau,-\mu_1-1-\tau,\mu_1+\mu_2+3-\tau;\beta_\pm,\varrho_\pm)\right.$$
$$-\frac{\mu_2+1}{\mu_1+\mu_2+3-\tau}\quad\frac{\boldsymbol{\nu}}{1+\tau\mp i\nu}\left[\frac{\mu_2+2}{\mu_1+1-\tau}\Phi_1(\mu_1+1-\tau,-\mu_1-\tau,\mu_1+\mu_2+4-\tau;\beta_\pm,\varrho_\pm)\right.$$
$$+2\frac{1}{\tau}\frac{r}{a}\Phi_1(\mu_1+2-\tau,-\mu_1-1-\tau,\mu_1+\mu_2+4-\tau;\beta_\pm,\varrho_\pm)\mp\frac{(\mu_2+2)(\mu_1+\mu_2+2)}{(\mu_1+2)(\mu_1+\mu_2+4-\tau)}\frac{2i(\kappa r\mp\boldsymbol{\kappa}\cdot\boldsymbol{r})}{1+\tau\mp i\nu}$$
$$\left.\left.\times\Phi_1(\mu_1+2-\tau,-\mu_1-1-\tau,\mu_1+\mu_2+5-\tau;\beta_\pm,\varrho_\pm)\right]\right\}\tag{3.10}$$

and

$$\mathcal{S}_{100}(\Omega,\hbar\boldsymbol{\kappa};\boldsymbol{r})=(\pi a^3)^{-1/2}\ \frac{m_e}{X^2}\left(\frac{2}{1+\tau\mp i\nu}\right)^{2+\tau}\exp\left(-\frac{1}{\tau}\frac{r}{a}\right)\sum_{\mu_1=0}^{\infty}\sum_{\mu_2=0}^{\infty}\frac{(\mu_1+\mu_2+1)!}{\mu_1!(\mu_1+1)!}\left[\mp\frac{2i(\kappa r\mp\boldsymbol{\kappa}\cdot\boldsymbol{r})}{1+\tau\mp i\nu}\right]^{\mu_1}$$
$$\times\left[\mp\frac{2i\nu}{1+\tau\mp i\nu}\right]^{\mu_2}\frac{(\mu_2+1)\Gamma(\mu_1+1-\tau)}{\Gamma(\mu_1+\mu_2+3-\tau)}\left\{\frac{\mu_1+1}{\mu_1+\mu_2+1}\ \Phi_1(\mu_1+1-\tau,-\mu_1-\tau,\mu_1+\mu_2+3-\tau;\beta_\pm,\varrho_\pm)\right.$$
$$+\frac{\mu_1+1-\tau}{\mu_2+1}\ \frac{1}{\tau}\ \frac{r}{a}\Phi_1(\mu_1+2-\tau,-\mu_1-1-\tau,\mu_1+\mu_2+3-\tau;\beta_\pm,\varrho_\pm)+\frac{\mu_2+2}{\mu_1+\mu_2+3-\tau}\ \frac{2}{1+\tau\mp i\nu}$$
$$\times\left[\mp\frac{i\nu}{2}\Phi_1(\mu_1+1-\tau,-\mu_1-\tau,\mu_1+\mu_2+4-\tau;\beta_\pm,\varrho_\pm)\right.$$
$$+\frac{\mu_1+1-\tau}{\mu_2+2}(i\boldsymbol{\kappa}\cdot\boldsymbol{r})\Phi_1(\mu_1+2-\tau,-\mu_1-1-\tau,\mu_1+\mu_2+4-\tau;\beta_\pm,\varrho_\pm)$$
$$\left.\left.-\nu\frac{(\mu_1+\mu_2+2)(\mu_1+1-\tau)}{(\mu_1+2)(\mu_1+\mu_2+4-\tau)}\ \frac{\kappa r\mp\boldsymbol{\kappa}\cdot\boldsymbol{r}}{1+\tau\mp i\nu}\right)\Phi_1(\mu_1+2-\tau,-\mu_1-1-\tau,\mu_1+\mu_2+5-\tau;\beta_\pm,\varrho_\pm)\right]\right\}.\tag{3.11}$$

In the EDA ($\boldsymbol{\kappa}=\boldsymbol{0}$, $\omega\neq 0$), which is valid for wavelengths considerably larger than the linear dimensions of the initial atomic state, the LRF's (2.15) and (2.16) become much simpler. First, as shown by Eq. (3.5), the orbital vector

$$\boldsymbol{W}_{100}(\Omega,\boldsymbol{0};\boldsymbol{r})\equiv\boldsymbol{w}_{100}(\Omega;\boldsymbol{r})\tag{3.12}$$

lies along the position vector $\boldsymbol{r}$:

$$\boldsymbol{w}_{100}(\Omega;\boldsymbol{r})=\frac{i}{\hbar}m_e(\pi a^3)^{-1/2}\tau\boldsymbol{r}\ \frac{ie^{i\pi\tau}}{2\sin(\pi\tau)}\int_1^{(0+)}d\rho$$
$$\times\rho^{1-\tau}\left(\frac{2}{\mathcal{N}_{1,\tau}}\right)^4\exp\left(-\frac{\mathcal{N}_{\tau,1}}{\mathcal{N}_{1,\tau}}\frac{1}{\tau}\frac{r}{a}\right).\tag{3.13}$$

According to Eq. (3.10) it has the explicit form

$$\boldsymbol{w}_{100}(\Omega;\boldsymbol{r})=\frac{i}{\hbar}m_e(\pi a^3)^{-1/2}\tau\boldsymbol{r}\left(\frac{2}{1+\tau}\right)^{2+\tau}\exp\left(-\frac{1}{\tau}\frac{r}{a}\right)$$
$$\times\frac{1}{2-\tau}\Phi_1(2-\tau,-1-\tau,3-\tau;\beta_1,\varrho_1),\tag{3.14}$$

with

$$\beta_1\equiv\frac{1}{2}(1-\tau),\qquad \varrho_1\equiv\frac{1-\tau}{\tau}\ \frac{r}{a}.\tag{3.15}$$

Second, for $\boldsymbol{\kappa}=\boldsymbol{0}$, the spin LRF (3.6) reduces, after an integration by parts, to the explicit expression

$$\mathcal{S}_{100}(\Omega,\mathbf{0};\boldsymbol{r})=-\frac{1}{\Omega-E_1}u_{100}(\boldsymbol{r}). \tag{3.16}$$

Notice that Eq. (3.16) trivially follows from the definition (2.16) by setting $\boldsymbol{\kappa}=\mathbf{0}$ therein and then substituting the eigenfunction expansion (A2) of the CGF.

We finally recall that Eqs. (3.13) and (3.14) were written in Ref. [8] as the ground-state case of our general formulas regarding the linear response from a $|nlm\rangle$ state in the EDA. It is worth adding that Eq. (3.13) invigorates the remarkable result reported by Luban, Nudler, and Freund in Ref. [2]. Indeed, for real values of τ, $0<\tau<2$, the integration path in Eq. (3.13) can be replaced by the interval $[0,1]$ on the real ρ axis:

$$\boldsymbol{w}_{100}(\Omega;\boldsymbol{r})=\frac{i}{\hbar}m_e(\pi a^3)^{-1/2}\tau\boldsymbol{r}\int_0^1 d\rho\rho^{1-\tau}\left(\frac{2}{\mathcal{N}_{1,\tau}}\right)^4 \times\exp\left\{\left[-1+\frac{2(1-\tau)\rho}{\mathcal{N}_{1,\tau}}\right]\frac{1}{\tau}\frac{r}{a}\right\} \quad (0<\tau<2). \tag{3.17}$$

Now, two changes of the variable of integration in Eq. (3.17),

$$\rho=\pm\frac{1+\tau}{1-\tau}\quad\frac{t}{1\mp t}, \tag{3.18}$$

where the upper signs must to be taken for $0<\tau<1$ and the lower ones for $1<\tau<2$, allow us to retrieve the following pair of twin compact formulas [9]:

$$\boldsymbol{w}_{100}(\Omega;\boldsymbol{r})=\frac{i}{\hbar}m_e(\pi a^3)^{-1/2}\tau\boldsymbol{r}\,\frac{2^4}{(1-\tau)^{2-\tau}(1+\tau)^{2+\tau}} \times\int_0^{(1/2)(1-\tau)}dt\ t^{1-\tau}(1-t)^{1+\tau} \times\exp\left[-(1-2t)\frac{1}{\tau}\frac{r}{a}\right] \quad (0<\tau<1) \tag{3.19}$$

and

$$\boldsymbol{w}_{100}(\Omega;\boldsymbol{r})=\frac{i}{\hbar}m_e(\pi a^3)^{-1/2}\tau\boldsymbol{r}\,\frac{2^4}{(\tau-1)^{2-\tau}(1+\tau)^{2+\tau}} \times\int_0^{(1/2)(\tau-1)}dt\ t^{1-\tau}(1+t)^{1+\tau} \times\exp\left[-(1+2t)\frac{1}{\tau}\frac{r}{a}\right] \quad (1<\tau<2). \tag{3.20}$$

IV. SECOND-ORDER RETARDATION APPROACH

A. Radiation gauge

Due to the intricacy of the explicit formulas (3.10) and (3.11), it is convenient to find the second-order retardation approach to the linear-response correction (2.13) in the radiation gauge. To this end, we write down the relevant approximations of the LRF's $\hat{\boldsymbol{\epsilon}}\cdot\boldsymbol{W}_{100}$ and $\mathcal{S}_{100}$, making use of their integral representations. Thus Eq. (3.5) provides the approximate formula

$$\hat{\boldsymbol{\epsilon}}\cdot\boldsymbol{W}_{100}(\Omega,\hbar\boldsymbol{\kappa};\boldsymbol{r})=\frac{i}{\hbar}m_e(\hat{\boldsymbol{\epsilon}}\cdot\boldsymbol{r})u_{100}(\boldsymbol{r})\tau e^{-\varrho_1}\frac{ie^{i\pi\tau}}{2\sin(\pi\tau)}\int_1^{(0+)}d\rho\rho^{1-\tau}\exp\left(\frac{2}{\mathcal{N}_{1,\tau}}\rho\varrho_1\right)\left(\frac{2}{\mathcal{N}_{1,\tau}}\right)^4 \times\left\{1+i(\boldsymbol{\kappa}\cdot\boldsymbol{r})\left(\frac{2}{\mathcal{N}_{1,\tau}}\right)^2\rho-\frac{1}{2}(\boldsymbol{\kappa}\cdot\boldsymbol{r})^2\left(\frac{2}{\mathcal{N}_{1,\tau}}\right)^4\rho^2-\frac{\nu^2}{2}\left(\frac{2}{\mathcal{N}_{1,\tau}}\right)^2(1-\rho)\left[(1-\rho)+\frac{1}{\tau}\frac{r}{a}\frac{2}{\mathcal{N}_{1,\tau}}\rho\right]+O(\nu^3)\right\}. \tag{4.1}$$

Having performed an integration by parts, we get from Eq. (3.6)

$$\mathcal{S}_{100}(\Omega,\hbar\boldsymbol{\kappa};\boldsymbol{r})=-\frac{1}{\Omega-E_1}u_{100}(\boldsymbol{r}) \times\left\{1+i\boldsymbol{\kappa}\cdot\boldsymbol{r}-i(\boldsymbol{\kappa}\cdot\boldsymbol{r})\tau\mathrm{e}^{-\varrho_1}\frac{ie^{i\pi\tau}}{2\sin(\pi\tau)} \times\int_1^{(0+)}d\rho\rho^{1-\tau}\exp\left(\frac{2}{\mathcal{N}_{1,\tau}}\rho\varrho_1\right)\left(\frac{2}{\mathcal{N}_{1,\tau}}\right)^4 +O(\nu^2)\right\}. \tag{4.2}$$

Insertion of Eqs. (4.1) and (4.2) into Eq. (2.13) yields the linear response taken in the second-order approximation of retardation:

$$\Psi_{100m_s}^{(1)}|_2(\boldsymbol{r},t)=-\frac{1}{2}e\mathcal{E}_0[e^{-i\omega t}\hat{\boldsymbol{\epsilon}}\cdot\boldsymbol{\mathcal{K}}(\Omega_1,\boldsymbol{\kappa};\boldsymbol{r}) +e^{i\omega t}\hat{\boldsymbol{\epsilon}}^*\cdot\boldsymbol{\mathcal{K}}(\Omega_2,-\boldsymbol{\kappa};\boldsymbol{r})]\Psi_{100m_s}^{(0)}(\boldsymbol{r},t). \tag{4.3}$$

In the left-hand side of Eq. (4.3) the subscript 2 denotes the order of retardation. $\boldsymbol{\mathcal{K}}$ is a 2×2 vector matrix defined as an integral representation:

$$\mathcal{K}(\Omega,\boldsymbol{\kappa};\boldsymbol{r})\equiv\frac{1}{\Omega-E_1}\Bigg\{\tau e^{-\varrho_1}\frac{ie^{i\pi\tau}}{2\sin(\pi\tau)}\int_1^{(0+)}d\rho\rho^{1-\tau}\exp\left(\frac{2}{\mathcal{N}_{1,\tau}}\rho\varrho_1\right)\left(\frac{2}{\mathcal{N}_{1,\tau}}\right)^4$$
$$\times\left[1+i(\boldsymbol{\kappa}\cdot\boldsymbol{r})\left(\frac{2}{\mathcal{N}_{1,\tau}}\right)^2\rho-\frac{1}{2}(\boldsymbol{\kappa}\cdot\boldsymbol{r})^2\left(\frac{2}{\mathcal{N}_{1,\tau}}\right)^4\rho^2-\frac{\nu}{2}\left(\frac{2}{\mathcal{N}_{1,\tau}}\right)^2(1-\rho)\left(\nu(1-\rho)+\kappa r\frac{2}{\mathcal{N}_{1,\tau}}\rho\right)\right]\boldsymbol{r}I_2$$
$$+\frac{1}{\Omega-E_1}\left[1+i\boldsymbol{\kappa}\cdot\boldsymbol{r}-i(\boldsymbol{\kappa}\cdot\boldsymbol{r})\tau e^{-\varrho_1}\frac{ie^{i\pi\tau}}{2\sin(\pi\tau)}\int_1^{(0+)}d\rho\rho^{1-\tau}\exp\left(\frac{2}{\mathcal{N}_{1,\tau}}\rho\varrho_1\right)\left(\frac{2}{\mathcal{N}_{1,\tau}}\right)^4\right]\frac{\hbar^2}{2m_e}(\boldsymbol{\kappa}\times\boldsymbol{\sigma})\Bigg\}. \quad (4.4)$$

We have denoted by I_2 the 2×2 unit matrix. By use of Eq. (B10) we get the explicit form of the matrix (4.4):

$$\mathcal{K}(\Omega,\boldsymbol{\kappa};\boldsymbol{r})=\frac{1}{\Omega-E_1}\left(\tau e^{-\varrho_1}\left(\frac{2}{1+\tau}\right)^{2+\tau}\Bigg\{\frac{1}{2-\tau}\Phi_1(2-\tau,-1-\tau,3-\tau;\beta_1,\varrho_1)+i(\boldsymbol{\kappa}\cdot\boldsymbol{r})\frac{2}{(1+\tau)(3-\tau)}\right.$$
$$\times\Phi_1(3-\tau,-2-\tau,4-\tau;\beta_1,\varrho_1)-\frac{1}{2}(\boldsymbol{\kappa}\cdot\boldsymbol{r})^2\left(\frac{2}{1+\tau}\right)^2\frac{1}{4-\tau}\Phi_1(4-\tau,-3-\tau,5-\tau;\beta_1,\varrho_1)$$
$$-\frac{\nu}{2}\left(\frac{2}{1+\tau}\right)^2\frac{1}{(3-\tau)(4-\tau)}\left[\frac{2\nu}{2-\tau}\Phi_1(2-\tau,-1-\tau,5-\tau;\beta_1,\varrho_1)+\kappa r\Phi_1(3-\tau,-2-\tau,5-\tau;\beta_1,\varrho_1)\right]\Bigg\}\boldsymbol{r}I_2$$
$$\left.+\frac{1}{\Omega-E_1}\left\{1+i(\boldsymbol{\kappa}\cdot\boldsymbol{r})\left[1-\tau e^{-\varrho_1}\left(\frac{2}{1+\tau}\right)^{2+\tau}\frac{1}{2-\tau}\Phi_1(2-\tau,-1-\tau,3-\tau;\beta_1,\varrho_1)\right]\right\}\frac{\hbar^2}{2m_e}(\boldsymbol{\kappa}\times\boldsymbol{\sigma})\right). \quad (4.5)$$

B. Multipolar gauge

We find it instructive to analyze the multipolar structure of the correction (4.3). With this aim in view, we first transform it from the radiation gauge to the Poincaré gauge by carrying out the PZW mapping [10,11],

$$\Psi'(\boldsymbol{r},t)=\exp\left[-\frac{ie}{\hbar c}\chi(\boldsymbol{r},t)\right]\Psi(\boldsymbol{r},t). \quad (4.6)$$

The generating function of the PZW transformation [12],

$$\chi(\boldsymbol{r},t)=-\int_0^1 du\ \boldsymbol{r}\cdot\boldsymbol{A}(u\boldsymbol{r},t)+c\int_0^t dt'\,\Phi(\boldsymbol{0},t'), \quad (4.7)$$

written with the potentials (2.2) and (2.3) and taken in the second-order retardation approach is

$$\chi|_2(\boldsymbol{r},t)=-\frac{c}{2i\omega}\mathcal{E}_0\left\{e^{-i\omega t}(\hat{\boldsymbol{\epsilon}}\cdot\boldsymbol{r})\left[1+\frac{1}{2}i(\boldsymbol{\kappa}\cdot\boldsymbol{r})-\frac{1}{6}(\boldsymbol{\kappa}\cdot\boldsymbol{r})^2\right]\right.$$
$$\left.-e^{i\omega t}(\hat{\boldsymbol{\epsilon}}^*\cdot\boldsymbol{r})\left[1-\frac{1}{2}i(\boldsymbol{\kappa}\cdot\boldsymbol{r})-\frac{1}{6}(\boldsymbol{\kappa}\cdot\boldsymbol{r})^2\right]\right\}. \quad (4.8)$$

In view of Eqs. (2.8), (4.6), and (4.8), the analog of the linear-response correction (4.3) in the Poincaré gauge is the sum

$$\Psi'^{(1)}_{100m_s}|_2(\boldsymbol{r},t)=\Psi^{(1)}_{100m_s}|_2(\boldsymbol{r},t)-\frac{ie}{\hbar c}\chi|_2(\boldsymbol{r},t)\Psi^{(0)}_{100m_s}(\boldsymbol{r},t). \quad (4.9)$$

According to our prescription (b) of Ref. [12], we need to perform an additional unitary transformation of the truncated wave function in the Poincaré gauge in order to get the adequate multipole contributions within a second-order approximation of retardation:

$$\Psi''|_2(\boldsymbol{r},t)=U_2\Psi'|_2(\boldsymbol{r},t). \quad (4.10)$$

In Eq. (4.10), U_2 is the phase factor

$$U_2=\exp\left[\frac{ie}{\hbar c}\frac{4\pi}{c}\frac{1}{30}r^2\boldsymbol{r}\cdot\boldsymbol{J}^d(\boldsymbol{0},t)\right], \quad (4.11)$$

where

$$\boldsymbol{J}^d(\boldsymbol{R},t)=\frac{c}{4\pi}\boldsymbol{\nabla}_{\mathbf{R}}\times\boldsymbol{B}(\boldsymbol{R},t) \quad (4.12)$$

is the displacement-current density of the given radiation field. It is worth stressing that one can regard Eq. (4.10) as a gauge transformation that is relevant *only* in the second-order approach of retardation:

$$U_2=\exp\left[-\frac{ie}{\hbar c}\tilde{\chi}_2(\boldsymbol{r},t)\right]. \quad (4.13)$$

Equations (4.11) and (4.12) specify its generating function,

$$\tilde{\chi}_2(\boldsymbol{r},t)=-\frac{1}{30}r^2\boldsymbol{r}\cdot[\boldsymbol{\nabla}_{\mathbf{R}}\times\boldsymbol{B}(\boldsymbol{R},t)]|_{\boldsymbol{R}=\boldsymbol{0}}. \quad (4.14)$$

Moreover, it can be inferred from Ref. [12] that starting from the Poincaré gauge, a sequence of new gauges is gradually generated when one increases the order of the retardation approach. Each of them is associated to a given approximation of retardation and will be called in what follows *a multipolar gauge*. The first one refers to our retardation approach and is provided by the gauge transformation (4.10). Taking into account Eq. (2.6), the generating function (4.14) reads

$$\widetilde{\chi}_2(\boldsymbol{r},t) = -\frac{c}{2i\omega}\mathcal{E}_0\frac{1}{30}(\kappa r)^2[e^{-i\omega t}(\hat{\boldsymbol{\epsilon}}\cdot\boldsymbol{r}) - e^{i\omega t}(\hat{\boldsymbol{\epsilon}}^*\cdot\boldsymbol{r})]. \tag{4.15}$$

We are left to apply the gauge transformation (4.10) to the truncated linear-response wave function in the Poincaré gauge $\Psi^{(0)}_{100m_s} + \Psi'^{(1)}_{100m_s}|_2$. On account of Eqs. (4.9) and (4.13) we get the linear-response correction in the multipolar gauge, restricted to a second-order approach of retardation:

$$\Psi''^{(1)}_{100m_s}|_2(\boldsymbol{r},t) = \Psi^{(1)}_{100m_s}(\boldsymbol{r},t) - \frac{ie}{\hbar c}[\chi|_2(\boldsymbol{r},t) + \widetilde{\chi}_2(\boldsymbol{r},t)] \times \Psi^{(0)}_{100m_s}(\boldsymbol{r},t). \tag{4.16}$$

Making use of Eqs. (4.3), (4.8), and (4.15), we derive the expression of the correction (4.16) to the unperturbed ground-state wave function (2.8):

$$\Psi''^{(1)}_{100m_s}|_2(\boldsymbol{r},t) = -\frac{1}{2}e\mathcal{E}_0[e^{-i\omega t}\hat{\boldsymbol{\epsilon}}\cdot\boldsymbol{\mathcal{L}}(\Omega_1,\boldsymbol{\kappa};\boldsymbol{r}) + e^{i\omega t}\hat{\boldsymbol{\epsilon}}^*\cdot\boldsymbol{\mathcal{L}}(\Omega_2,-\boldsymbol{\kappa};\boldsymbol{r})]\Psi^{(0)}_{100m_s}(\boldsymbol{r},t). \tag{4.17}$$

In Eq. (4.17), $\boldsymbol{\mathcal{L}}$ is a 2×2 vector matrix related to $\boldsymbol{\mathcal{K}}$ as follows:

$$\boldsymbol{\mathcal{L}}(\Omega,\boldsymbol{\kappa};\boldsymbol{r}) \equiv \boldsymbol{\mathcal{K}}(\Omega,\boldsymbol{\kappa};\boldsymbol{r}) - \frac{1}{\Omega - E_1} \times \left\{1 + \frac{1}{2}i(\boldsymbol{\kappa}\cdot\boldsymbol{r}) - \frac{1}{6}\left[(\boldsymbol{\kappa}\cdot\boldsymbol{r})^2 - \frac{1}{5}(\kappa r)^2\right]\right\}\boldsymbol{r}I_2. \tag{4.18}$$

By inserting first Eq. (4.4) and then Eq. (4.5) into the definition (4.18) we find the vector operator $\boldsymbol{\mathcal{L}}$ as an integral representation and, respectively, in explicit form.

V. STRUCTURE OF THE TRUNCATED FIRST-ORDER MULTIPOLAR PAULI HAMILTONIAN

Reference [12] provides the multipolar Pauli Hamiltonian, linear in the field and truncated to a second-order approximation of retardation,

$$H''^{(1)}|_2 = H_0^{(1)} + H_1^{(1)} + H_2^{(1)}. \tag{5.1}$$

The subscript of a term in the right-hand side of Eq. (5.1) denotes the order of retardation. We write down the three terms [13]

$$H_0^{(1)} = -\boldsymbol{Q}(x)\cdot\boldsymbol{E}(\boldsymbol{0},t), \tag{5.2a}$$

$$H_1^{(1)} = -\left\{Q_{jk}(x)\frac{\partial E_j(\boldsymbol{R},t)}{\partial X_k} + [\boldsymbol{M}(x) + \boldsymbol{M}^s]\cdot\boldsymbol{B}(\boldsymbol{R},t)\right\}\Bigg|_{\mathbf{R}=\mathbf{0}}, \tag{5.2b}$$

$$H_2^{(1)} = -\left\{Q_{jkl}(x)\frac{\partial^2 E_j(\boldsymbol{R},t)}{\partial X_k \partial X_l} + [M_{jk}(x) + M^s_{jk}(x)]\frac{\partial B_j(\boldsymbol{R},t)}{\partial X_k} + \frac{4\pi}{c}[\boldsymbol{C}(x) + \boldsymbol{C}^s(x)]\cdot\boldsymbol{J}^d(\boldsymbol{R},t)\right\}\Bigg|_{\mathbf{R}=\mathbf{0}}. \tag{5.2c}$$

Denoting by $\boldsymbol{L}$ the orbital angular momentum of the electron, we list below the observables introduced in Eqs. (5.2).

(*i*) *Electric* 2^L*-pole moments* ($L=1,2,3$): dipole (E1),

$$Q_j(x) = -ex_j, \tag{5.3a}$$

quadrupole (E2),

$$Q_{jk}(x) = -e\frac{1}{2!}\left(x_j x_k - \frac{1}{3}r^2\delta_{jk}\right), \tag{5.3b}$$

octupole (E3),

$$Q_{jkl}(x) = -e\frac{1}{3!}\left[x_j x_k x_l - \frac{1}{5}r^2(\delta_{jk}x_l + \delta_{kl}x_j + \delta_{lj}x_k)\right]. \tag{5.3c}$$

(*ii*) *Orbital magnetic* 2^L*-pole moments* ($L=1,2$): dipole (M1),

$$M_j(x) = -\frac{e}{m_e c}\frac{1}{2!}L_j, \tag{5.4a}$$

quadrupole (M2),

$$M_{jk}(x) = -\frac{e}{m_e c}\frac{1}{3!}(x_j L_k + x_k L_j). \tag{5.4b}$$

(*iii*) *Spin magnetic* 2^L*-pole moments* ($L=1,2$): dipole (M1),

$$M_j^s = -\frac{e\hbar}{2m_e c}\sigma_j, \tag{5.5a}$$

quadrupole (M2),

$$M^s_{jk}(x) = -\frac{e\hbar}{2m_e c}\frac{1}{2!}\left[x_j\sigma_k + x_k\sigma_j - \frac{2}{3}\delta_{jk}(\boldsymbol{r}\cdot\boldsymbol{\sigma})\right]. \tag{5.5b}$$

(*iv*) *Orbital displacement-current dipole moment* (C1),

$$C_j(x) = -\frac{e}{m_e c}\frac{1}{10}(\boldsymbol{r}\times\boldsymbol{L} - r^2\boldsymbol{P})_j. \tag{5.6}$$

(*v*) *Spin displacement-current dipole moment* (C1),

$$C_j^s(x) = -\frac{e}{2m_e c}\frac{1}{2!}(\boldsymbol{r}\times\boldsymbol{\sigma})_j. \tag{5.7}$$

Taking into account the expressions of the field strengths, Eqs. (2.5) and (2.6), we obtain the following form of the multipolar Hamiltonian given by Eqs. (5.1) and (5.2):

$$H''^{(1)}|_2=\frac{1}{2}[e^{-i\omega t}h(\boldsymbol{r})+e^{i\omega t}h^\dagger(\boldsymbol{r})]. \tag{5.8}$$

In Eq. (5.8) $h(\boldsymbol{r})$ is a sum of three operators,

$$h(\boldsymbol{r})=h_0(\boldsymbol{r})+h_1(\boldsymbol{r})+h_2(\boldsymbol{r}), \tag{5.9}$$

each of them having a definite order of retardation:

$$h_0(\boldsymbol{r})\equiv-\mathcal{E}_0\hat{\boldsymbol{\epsilon}}\cdot\boldsymbol{Q}(x), \tag{5.10a}$$

$$h_1(\boldsymbol{r})\equiv-\mathcal{E}_0\left\{i\epsilon_j\kappa_kQ_{jk}(x)+\frac{c}{\omega}(\boldsymbol{\kappa}\times\hat{\boldsymbol{\epsilon}})\cdot[\boldsymbol{M}(x)+\boldsymbol{M}^s]\right\}, \tag{5.10b}$$

$$h_2(\boldsymbol{r})\equiv-\mathcal{E}_0\left\{-\epsilon_j\kappa_k\kappa_lQ_{jkl}(x)+i\frac{c}{\omega}(\boldsymbol{\kappa}\times\hat{\boldsymbol{\epsilon}})_j\kappa_k[M_{jk}(x)+M^s_{jk}(x)]-i\frac{c}{\omega}\kappa^2\hat{\boldsymbol{\epsilon}}\cdot[\boldsymbol{C}(x)+\boldsymbol{C}^s(x)]\right\}. \tag{5.10c}$$

VI. MULTIPOLE TERMS IN THE LINEAR-RESPONSE WAVE FUNCTION

The truncated first-order correction (4.17) to the ground-state wave function in the multipolar gauge is expressible by means of the CGF (A2) and the two terms of the Hamiltonian (5.8) as

$$\Psi''^{(1)}_{100m_s}|_2(\boldsymbol{r},t)=\frac{1}{2}\left[e^{-i\omega t}\int d^3x'\,G(\Omega_1;\boldsymbol{r},\boldsymbol{r}')h(\boldsymbol{r}')\times\Psi^{(0)}_{100m_s}(\boldsymbol{r}',t)+e^{i\omega t}\int d^3x'\,G(\Omega_2;\boldsymbol{r},\boldsymbol{r}')\times h^\dagger(\boldsymbol{r}')\Psi^{(0)}_{100m_s}(\boldsymbol{r}',t)\right]. \tag{6.1}$$

Two relevant quantities are to be evaluated in order to get a compact form of the linear-response correction (6.1). Remark first that a 2^L-pole electric moment can be written as a harmonic homogeneous polynomial of degree L,

$$Q_{j_1\cdots j_L}(x)=C^{(L)}_{j_1\cdots j_Lj'_1\cdots j'_L}x_{j'_1}\cdots x_{j'_L}. \tag{6.2}$$

In Eq. (6.2) $C^{(L)}$ is a $2L$-rank constant tensor, which is totally symmetric and traceless with respect to Cartesian indices belonging to each of the sets $j_1,\ldots,j_L$ and $j'_1,\ldots,j'_L$, respectively. In view of Eqs. (A10) and (2.11),

$$Q_{j_1\cdots j_L}(x)u_{100}(\boldsymbol{r})=(\pi a^3)^{-1/2}C^{(L)}_{j_1\cdots j_Lj'_1\cdots j'_L}(-\hbar)\times\left(\frac{\hbar}{i}\right)^L\frac{\partial^L}{\partial q_{j'_1}\ldots\partial q_{j'_L}}\frac{\partial\mathcal{U}}{\partial\lambda}\bigg|_{(\mathbf{q=0},\ \lambda=\hbar/a)}. \tag{6.3}$$

Hence, according to Eq. (A9),

$$-\int d^3x'\,G(\Omega;\boldsymbol{r},\boldsymbol{r}')Q_{j_1\cdots j_L}(x')u_{100}(\boldsymbol{r}')=(\pi a^3)^{-1/2}(-\hbar)\left(\frac{\hbar}{i}\right)^LC^{(L)}_{j_1\cdots j_Lj'_1\cdots j'_L}\times\frac{\partial^L}{\partial q_{j'_1}\cdots\partial q_{j'_L}}\frac{\partial\mathcal{F}}{\partial\lambda}\bigg|_{(\mathbf{q=0},\ \lambda=\hbar/a)}. \tag{6.4}$$

Making use of Eqs. (A14)–(A16), we get

$$-\int d^3x'\,G(\Omega;\boldsymbol{r},\boldsymbol{r}')Q_{j_1\cdots j_L}(x')u_{100}(\boldsymbol{r}')=-\frac{m_e}{X}(\pi a^3)^{-1/2}Q_{j_1\cdots j_L}(x)\frac{ie^{i\pi\tau}}{2\sin(\pi\tau)}\times\int_1^{(0+)}d\rho\rho^{-1-\tau}\left\{\left[(L+1)\frac{\partial f}{\partial\lambda}-f\frac{\partial g}{\partial\lambda}\frac{1}{\tau}\frac{r}{a}\right]f^L\times\exp\left(-g\frac{1}{\tau}\frac{r}{a}\right)\right\}\bigg|_{(\mathbf{q=0},\ \lambda=\hbar/a)}\quad(L=0,1,2,3,\ldots). \tag{6.5}$$

On the other hand, there is no contribution of the orbital magnetic moments in the ground state:

$$M_{j_1\cdots j_L}(x)u_{100}(\boldsymbol{r})=0\quad(L=1,2,3,\ldots). \tag{6.6}$$

By contrast, the orbital displacement-current dipole moment has a nonvanishing effect:

$$C_j(x)u_{100}(\boldsymbol{r})=-(\pi a^3)^{-1/2}\frac{e}{m_ec}\frac{1}{10a}\left(\frac{\hbar}{i}\right)^4\times\frac{\partial^3\mathcal{U}}{\partial q_j\partial q_l\partial q_l}\bigg|_{(\mathbf{q=0},\ \lambda=\hbar/a)}. \tag{6.7}$$

Equation (A9) yields the parallel relationship

$$-\int d^3x'\,G(\Omega;\boldsymbol{r},\boldsymbol{r}')C_j(x')u_{100}(\boldsymbol{r}')=-(\pi a^3)^{-1/2}\frac{e}{m_ec}\frac{1}{10a}\left(\frac{\hbar}{i}\right)^4\frac{\partial^3\mathcal{F}}{\partial q_j\partial q_l\partial q_l}\bigg|_{(\mathbf{q=0},\ \lambda=\hbar/a)}, \tag{6.8}$$

which becomes, upon using Eqs. (A14)–(A16),

$$-\int d^3x'\,G(\Omega;\boldsymbol{r},\boldsymbol{r}')C_j(x')u_{100}(\boldsymbol{r}')$$
$$=\frac{1}{2\Omega}(\pi a^3)^{-1/2}\frac{e}{m_e c}\,\frac{1}{10}\,\frac{1}{\tau a^2}\,\frac{\hbar}{i}x_j\frac{ie^{i\pi\tau}}{2\sin(\pi\tau)}$$
$$\times\int_1^{(0+)}d\rho\,\rho^{-1-\tau}\exp\left(-g\frac{1}{\tau}\frac{r}{a}\right)$$
$$\times\left\{f^4r^2-10\hbar^2\left[2\frac{\partial f}{\partial(q^2)}-f\frac{\partial g}{\partial(q^2)}\frac{1}{\tau}\frac{r}{a}\right]\right\}\Bigg|_{(\mathbf{q}=\mathbf{0},\ \lambda=\hbar/a)}. \tag{6.9}$$

Substitution of Eqs. (A15) and (A16) into Eq. (6.5) leads to the following integral representation:

$$-\int d^3x'\,G(\Omega;\boldsymbol{r},\boldsymbol{r}')Q_{j_1\cdots j_L}(x')u_{100}(\boldsymbol{r}')$$
$$=-\frac{1}{2\Omega}Q_{j_1\cdots j_L}(x)u_{100}(\boldsymbol{r})e^{-\varrho_1}\frac{ie^{i\pi\tau}}{2\sin(\pi\tau)}$$
$$\times\int_1^{(0+)}d\rho\,\rho^{L-\tau}\exp\left(\frac{2}{\mathcal{N}_{1,\tau}}\rho\varrho_1\right)\left(\frac{2}{\mathcal{N}_{1,\tau}}\right)^{2L+3}$$
$$\times\left[(L+1)(1-\rho)+\frac{2}{\mathcal{N}_{1,\tau}}\rho\frac{1}{\tau}\frac{r}{a}\right]\quad(L=0,1,2,3,\ldots). \tag{6.10a}$$

Integration by parts in Eq. (6.10a) allows us to write the equivalent formula

$$-\int d^3x'\,G(\Omega;\boldsymbol{r},\boldsymbol{r}')Q_{j_1\cdots j_L}(x')u_{100}(\boldsymbol{r}')$$
$$=-\frac{1}{\Omega-E_1}Q_{j_1\cdots j_L}(x)u_{100}(\boldsymbol{r})$$
$$\times\left[1-L\tau e^{-\varrho_1}\frac{ie^{i\pi\tau}}{2\sin(\pi\tau)}\int_1^{(0+)}d\rho\,\rho^{L-\tau}\right.$$
$$\left.\times\exp\left(\frac{2}{\mathcal{N}_{1,\tau}}\rho\varrho_1\right)\left(\frac{2}{\mathcal{N}_{1,\tau}}\right)^{2L+2}\right]\quad(L=0,1,2,3,\ldots). \tag{6.10b}$$

Similarly, using Eq. (6.9), one gets directly the integral representation

$$-\int d^3x'\,G(\Omega;\boldsymbol{r},\boldsymbol{r}')C_j(x')u_{100}(\boldsymbol{r}')$$
$$=-\frac{1}{2\Omega}[C_j(x)u_{100}(\boldsymbol{r})]\tau\frac{a}{r}e^{-\varrho_1}\frac{ie^{i\pi\tau}}{2\sin(\pi\tau)}$$
$$\times\int_1^{(0+)}d\rho\,\rho^{1-\tau}\exp\left(\frac{2}{\mathcal{N}_{1,\tau}}\rho\varrho_1\right)\left(\frac{2}{\mathcal{N}_{1,\tau}}\right)^6\left[5(1-\rho)^2\right.$$
$$\left.+5\frac{1}{\tau}\frac{r}{a}\rho(1-\rho)\frac{2}{\mathcal{N}_{1,\tau}}+\left(\frac{1}{\tau}\frac{r}{a}\right)^2\rho^2\left(\frac{2}{\mathcal{N}_{1,\tau}}\right)^2\right]. \tag{6.11a}$$

After performing a suitable integration by parts in Eq. (6.11a), we find alternatively

$$-\int d^3x'\,G(\Omega;\boldsymbol{r},\boldsymbol{r}')C_j(x')u_{100}(\boldsymbol{r}')$$
$$=-\frac{1}{\Omega-E_1}[C_j(x)u_{100}(\boldsymbol{r})]\left(1-\tau\frac{a}{r}e^{-\varrho_1}\frac{ie^{i\pi\tau}}{2\sin(\pi\tau)}\right.$$
$$\times\int_1^{(0+)}d\rho\,\rho^{1-\tau}\exp\left(\frac{2}{\mathcal{N}_{1,\tau}}\rho\varrho_1\right)\left(\frac{2}{\mathcal{N}_{1,\tau}}\right)^6$$
$$\times\left\{-5\rho+5(1-\rho)\left[\frac{1}{2}\mathcal{N}_{1,\tau}-(1-\tau)\rho\right]\right.$$
$$\left.\left.+\frac{1}{\tau}\frac{r}{a}\rho\left[3-\tau-3(1-\tau)\frac{2}{\mathcal{N}_{1,\tau}}\rho\right]\right\}\right). \tag{6.11b}$$

Now, by applying Eq. (B10), we obtain from the integral representations (6.10) and (6.11) two pairs of equivalent explicit formulas:

$$-\int d^3x'\,G(\Omega;\boldsymbol{r},\boldsymbol{r}')Q_{j_1\cdots j_L}(x')u_{100}(\boldsymbol{r}')$$
$$=-\frac{1}{2\Omega}Q_{j_1\cdots j_L}(x)u_{100}(\boldsymbol{r})e^{-\varrho_1}\left(\frac{2}{1+\tau}\right)^{L+2+\tau}$$
$$\times\left[\frac{L+1}{(L+1-\tau)(L+2-\tau)}\right.$$
$$\times\Phi_1(L+1-\tau,-L-\tau,L+3-\tau;\beta_1,\varrho_1)$$
$$+\frac{1}{\tau}\frac{r}{a}\frac{1}{L+2-\tau}$$
$$\left.\times\Phi_1(L+2-\tau,-L-1-\tau,L+3-\tau;\beta_1,\varrho_1)\right]$$
$$(L=0,1,2,3,\ldots), \tag{6.12a}$$

or

$$-\int d^3x'\,G(\Omega;\boldsymbol{r},\boldsymbol{r}')Q_{j_1\cdots j_L}(x')u_{100}(\boldsymbol{r}')$$
$$=-\frac{1}{\Omega-E_1}Q_{j_1\cdots j_L}(x)u_{100}(\boldsymbol{r})$$
$$\times\left[1-\left(\frac{2}{1+\tau}\right)^{L+1+\tau}e^{-\varrho_1}\frac{L\tau}{L+1-\tau}\right.$$
$$\left.\times\Phi_1(L+1-\tau,-L-\tau,L+2-\tau;\beta_1,\varrho_1)\right]$$
$$(L=0,1,2,3,\ldots). \tag{6.12b}$$

Then,

$$-\int d^3x' G(\Omega;\boldsymbol{r},\boldsymbol{r}')C_j(x')u_{100}(\boldsymbol{r}')$$
$$=-\frac{1}{2\Omega}[C_j(x)u_{100}(\boldsymbol{r})]\tau\frac{a}{r}e^{-\varrho_1}\left(\frac{2}{1+\tau}\right)^{4+\tau}$$
$$\times\left[\frac{10}{(2-\tau)(3-\tau)(4-\tau)}\right.$$
$$\times\Phi_1(2-\tau,-1-\tau,5-\tau;\beta_1,\varrho_1)$$
$$+\frac{1}{\tau}\frac{r}{a}\frac{5}{(3-\tau)(4-\tau)}\Phi_1(3-\tau,-2-\tau,5-\tau;\beta_1,\varrho_1)$$
$$\left.+\left(\frac{1}{\tau}\frac{r}{a}\right)^2\frac{1}{4-\tau}\Phi_1(4-\tau,-3-\tau,5-\tau;\beta_1,\varrho_1)\right] \tag{6.13a}$$

or

$$-\int d^3x' G(\Omega;\boldsymbol{r},\boldsymbol{r}')C_j(x')u_{100}(\boldsymbol{r}')$$
$$=-\frac{1}{\Omega-E_1}[C_j(x)u_{100}(\boldsymbol{r})]\left(1-e^{-\varrho_1}\left(\frac{2}{1+\tau}\right)^{3+\tau}\right.$$
$$\times\left\{\tau\frac{a}{r}\left[-\frac{5}{3-\tau}\Phi_1(3-\tau,-2-\tau,4-\tau;\beta_1,\varrho_1)\right.\right.$$
$$+\frac{5}{(2-\tau)(3-\tau)}\Phi_1(2-\tau,-1-\tau,4-\tau;\beta_1,\varrho_1)$$
$$\left.-\frac{5(1-\tau)}{(3-\tau)(4-\tau)}\Phi_1(3-\tau,-1-\tau,5-\tau;\beta_1,\varrho_1)\right]$$
$$+\Phi_1(3-\tau,-2-\tau,4-\tau;\beta_1,\varrho_1)$$
$$\left.\left.-3\frac{1-\tau}{4-\tau}\Phi_1(4-\tau,-2-\tau,5-\tau;\beta_1,\varrho_1)\right\}\right). \tag{6.13b}$$

Substitution of Eqs. (6.10b), (6.6), and (6.11a) into Eq. (6.1) allows us to retrieve the structure (4.17), where each operator $\mathcal{L}$ is written as an integral representation which is precisely that obtained by plugging Eq. (4.4) into Eq. (4.18). Finally, insertion of the explicit formulas (6.12b), (6.6), and (6.13a) into Eq. (6.1) leads again to Eq. (4.17) with each operator $\mathcal{L}$ given this time explicitly by Eqs. (4.18) and (4.5).

VII. LOW- AND HIGH-FREQUENCY BEHAVIOR OF THE MULTIPOLE TERMS

A. Low-frequency case

As we have seen in Sec. VI, the linear-response correction (4.16), truncated to the second-order retardation approach in the multipolar gauge, has an orbital part which consists of electric 2^L-pole contributions (6.12) with $L=1,2,3$, and displacement-current dipole terms (6.13). Besides, its spin terms are linear combinations of the electric dipole components, Eq. (6.12), with $L=1$. The low-frequency regime is characterized by values Ω in the vicinity of the ground-state Bohr level E_1,

$$\Omega=E_1+\delta\Omega \quad (|\delta\Omega|\ll|E_1|). \tag{7.1}$$

Consequently, we set in Eqs. (6.12) and (6.13)

$$\tau\equiv 1+\delta\tau \quad (|\delta\tau|\ll 1), \tag{7.2}$$

keeping all terms of orders not exceeding the order $(\delta\tau)^2$, which corresponds to $(\delta\Omega/2|E_1|)^2$. From Eq. (6.12a) we have derived the following approximate formula valid for an arbitrary multipole rank L:

$$-\int d^3x' G(E_1+\delta\Omega;\boldsymbol{r},\boldsymbol{r}')Q_{j_1\cdots j_L}(x')u_{100}(\boldsymbol{r}')$$
$$=\frac{1}{2|E_1|}Q_{j_1\cdots j_L}(x)u_{100}(\boldsymbol{r})\left(\frac{1}{L}+\frac{1}{L+1}\frac{r}{a}+\frac{\delta\Omega}{2|E_1|}\left[\left(\frac{1}{L}+\frac{1}{L+1}+\frac{1}{L+2}\right)\left(\frac{1}{L}+\frac{1}{L+1}\frac{r}{a}\right)+\frac{1}{(L+1)(L+2)}\left(\frac{r}{a}\right)^2\right]\right.$$
$$+\left(\frac{\delta\Omega}{2|E_1|}\right)^2\left\{\left[\frac{1}{L^2}+\frac{1}{(L+1)^2}+\frac{1}{(L+2)^2}+\frac{3}{L(L+1)}+\frac{9}{(L+1)(L+3)}\right]\left(\frac{1}{L}+\frac{1}{L+1}\frac{r}{a}\right)\right.$$
$$\left.\left.+\frac{1}{(L+1)(L+2)}\left(\frac{1}{L}+\frac{1}{L+1}+\frac{1}{L+2}+\frac{3}{L+3}\right)\left(\frac{r}{a}\right)^2+\frac{1}{(L+1)(L+2)(L+3)}\left(\frac{r}{a}\right)^3\right\}+O\left[\left(\frac{\delta\Omega}{2|E_1|}\right)^3\right]\right) \quad (L=1,2,3,\dots). \tag{7.3}$$

Equation (6.12b) was used to check the low-frequency expansion (7.3). Then we have found with Eq. (6.13a) and checked by means of Eq. (6.13b) the similar expansion of the displacement-current dipole term:

$$-\int d^3x'\, G(E_1+\delta\Omega;\boldsymbol{r},\boldsymbol{r}')C_j(x')u_{100}(\boldsymbol{r}')$$
$$=\frac{1}{2|E_1|}[C_j(x)u_{100}(\boldsymbol{r})]\frac{a}{r}\Bigg\{\frac{5}{3}\left(1+\frac{1}{2}\frac{r}{a}\right)+\frac{1}{3}\left(\frac{r}{a}\right)^2+\frac{\delta\Omega}{2|E_1|}\left[\frac{155}{36}\left(1+\frac{1}{2}\frac{r}{a}\right)+\frac{19}{36}\left(\frac{r}{a}\right)^2+\frac{1}{12}\left(\frac{r}{a}\right)^3\right]$$
$$+\left(\frac{\delta\Omega}{2|E_1|}\right)^2\left[\frac{4643}{432}\left(1+\frac{1}{2}\frac{r}{a}\right)+\frac{2783}{2160}\left(\frac{r}{a}\right)^2+\frac{137}{720}\left(\frac{r}{a}\right)^3+\frac{1}{60}\left(\frac{r}{a}\right)^4\right]+O\left[\left(\frac{\delta\Omega}{2|E_1|}\right)^3\right]\Bigg\}. \tag{7.4}$$

When employing the expansions (7.3) and (7.4) in Eq. (6.1), we get the truncated linear-response correction (4.17) to the wave function in the range of low frequencies, which is accurate to the order $(\hbar\omega/2|E_1|)^2$.

B. High-frequency case

We analyze the relevant multipole terms (6.12a) and (6.13a) in the first-order correction (6.1) to the ground-state wave function for frequencies which are high in comparison with the characteristic atomic ones. Applying the asymptotic formula (B12) of the Humbert function Φ_1, we obtain for $\tau\rightarrow 0$

$$-\int d^3x'\, G(\Omega;\boldsymbol{r},\boldsymbol{r}')Q_{j_1,\ldots,j_L}(x')u_{100}(\boldsymbol{r}')$$
$$=-\frac{1}{\Omega}Q_{j_1,\ldots,j_L}(x)u_{100}(\boldsymbol{r})[1+O(|\tau|)]$$
$$(L=1,2,3,\ldots) \tag{7.5}$$

and

$$-\int d^3x'\, G(\Omega;\boldsymbol{r},\boldsymbol{r}')C_j(x')u_{100}(\boldsymbol{r}')$$
$$=-\frac{1}{\Omega}[C_j(x)u_{100}(\boldsymbol{r})][1+O(|\tau|)]. \tag{7.6}$$

Note that Eqs. (7.5) and (7.6) are direct consequences of the asymptotic behavior of the CGF, Eq. (A3). By inserting Eqs. (7.5) and (7.6) into Eq. (6.1), we find the high-frequency limit of the truncated linear-response correction in the multipolar gauge:

$$\Psi''^{(1)}_{100m_s}|_2(\boldsymbol{r},t)\underset{\omega\rightarrow\infty}{\sim}\frac{1}{2\hbar\omega}[e^{-i\omega t}h(\boldsymbol{r})-e^{i\omega t}h^{\dagger}(\boldsymbol{r})]$$
$$\times\Psi^{(0)}_{100m_s}(\boldsymbol{r},t). \tag{7.7}$$

In addition, we use Eq. (A3) to get the high-frequency behavior of the exact first-order correction (2.13) in the radiation gauge:

$$\Psi^{(1)}_{100m_s}(\boldsymbol{r},t)\underset{\omega\rightarrow\infty}{\sim}\frac{e\mathcal{E}_0}{m_e\omega}\frac{1}{2\hbar\omega}\Bigg\{e^{i(\boldsymbol{\kappa}\cdot\mathbf{r}-\omega t)}$$
$$\times\left[\frac{1}{i}\hat{\boldsymbol{\epsilon}}\cdot\boldsymbol{P}+(\boldsymbol{\kappa}\times\hat{\boldsymbol{\epsilon}})\cdot\boldsymbol{S}\right]+e^{-i(\boldsymbol{\kappa}\cdot\mathbf{r}-\omega t)}$$
$$\times\left[\frac{1}{i}\hat{\boldsymbol{\epsilon}}^*\cdot\boldsymbol{P}-(\boldsymbol{\kappa}\times\hat{\boldsymbol{\epsilon}}^*)\cdot\boldsymbol{S}\right]\Bigg\}\Psi^{(0)}_{100m_s}(\boldsymbol{r},t). \tag{7.8}$$

VIII. CONCLUSIONS

We have obtained an *exact* linear-response wave function of a hydrogenlike atom perturbed by a single-mode radiation field. In the remote past ($t\rightarrow-\infty$), the atom is in its unperturbed ground state, and for negative times the field evolves adiabatically. The first-order correction (2.13) to the wave function in the radiation gauge, with the closed-form expressions (3.5) and (3.6) inserted, is the analytic solution of Podolsky's problem complemented with the first-order contribution due to the magnetic interaction of the electron spin. The basic mathematical tool is the generating function (A9) of the linear response, which has been evaluated using Hostler's compact integral representation (A4) of the nonrelativistic CGF in coordinate space. It will also be employed in the following paper [14] to derive in the EDA the LRF's associated to any excited stationary state.

The closed-form contour integrals (3.5) and (3.6) are by far more important than the explicit expressions (3.10) and (3.11) of the LRF's as double ascending power series. We have started out with them to get the linear response in the second-order retardation approach in the radiation gauge, Eq. (4.3), as well as in the multipolar gauge, Eq. (4.17). It is also worth adding that the exact nonrelativistic Rayleigh scattering amplitude from the atomic ground state [15] can be retrieved such as to include the corrections due to the intrinsic magnetic moment of the electron [16].

We have analyzed the first- and second-order corrections of retardation in the multipolar gauge, split as they are into orbital and spin contributions. The orbital part of the first-order correction arises from the electric quadrupole coupling, while that of the second-order correction consists of electric octupole and displacement-current dipole terms. The spin second-order corrections can be described by means of the electric dipole coupling. For low and high frequencies of the applied field, the behavior of all the relevant quantities has been finally established.

APPENDIX A: GENERATING FUNCTION OF THE LINEAR RESPONSE

We recall that the Schrödinger CGF in coordinate space $G(\Omega;\boldsymbol{r},\boldsymbol{r}')$ is a solution of the inhomogeneous differential equation

$$(H^{(0)}-\Omega)G(\Omega;\boldsymbol{r},\boldsymbol{r}')=-\delta(\boldsymbol{r}-\boldsymbol{r}'), \tag{A1}$$

whose eigenfunction expansion is

$$G(\Omega;\boldsymbol{r},\boldsymbol{r}')=\sum_{N'}\frac{u_{N'}(\boldsymbol{r})u_{N'}^{*}(\boldsymbol{r}')}{\Omega-E_{n'}}. \tag{A2}$$

In Eq. (A1), $H^{(0)}$ is the field-free Coulomb Hamiltonian, while in Eq. (A2), $E_{n'}$ are its eigenvalues and $\{u_{N'}(\boldsymbol{r})\}$ denotes a complete orthonormal set of Coulomb energy eigenfunctions. The CGF is defined for any complex value Ω not belonging to the energy spectrum. Note also its asymptotic behavior:

$$G(\Omega;\boldsymbol{r},\boldsymbol{r}')\widetilde{_{|\Omega|\to\infty}}\frac{1}{\Omega}\delta(\boldsymbol{r}-\boldsymbol{r}'). \tag{A3}$$

In what follows we employ Hostler's integral representation of the CGF [4]:

$$G(\Omega;\boldsymbol{r},\boldsymbol{r}')=-\frac{2m_e}{\hbar^2}\frac{1}{4\pi}\frac{X}{\hbar}\frac{ie^{i\pi\tau}}{2\sin(\pi\tau)}\int_{+\infty}^{(+1)}d\zeta\left(\frac{\zeta+1}{\zeta-1}\right)^{\tau}\times\exp\left[-\frac{X}{\hbar}(r+r')\zeta\right]\times I_0\left(2\frac{X}{\hbar}(rr')^{1/2}(\zeta^2-1)^{1/2}\cos\frac{\Theta}{2}\right). \tag{A4}$$

In Eq. (A4), the contour of integration in the complex ζ plane starts at $+\infty+i0$, runs down to the point $\zeta=1$ on the upper side of the cut $(1,+\infty)$, with $\arg\zeta=0$, encircles the point $\zeta=1$ in the counterclockwise sense, and then goes to $+\infty-i0$ along the cut $(1,+\infty)$, below it. We have denoted

$$X\equiv(-2m_e\Omega)^{1/2}\quad(\mathrm{Re}X>0), \tag{A5}$$

$$\tau\equiv\frac{\hbar}{aX}, \tag{A6}$$

where a is the scaled Bohr radius (2.10), and

$$\cos\Theta\equiv\hat{\boldsymbol{r}}\cdot\hat{\boldsymbol{r}}'. \tag{A7}$$

$I_0(z)$ is the modified Bessel function of zeroth order, which can be written as a contour integral [17],

$$I_0(z)=\frac{1}{2\pi i}\int_{c-i\infty}^{c+i\infty}ds\frac{1}{s}\exp\left(s+\frac{z^2}{4s}\right), \tag{A8}$$

where c is a positive number.

We define the generating function

$$\mathcal{F}(\Omega,\boldsymbol{q},\lambda;\boldsymbol{r})\equiv-\int d^3x'\,G(\Omega;\boldsymbol{r},\boldsymbol{r}')\mathcal{U}(\boldsymbol{q},\lambda;\boldsymbol{r}'), \tag{A9}$$

with

$$\mathcal{U}(\boldsymbol{q},\lambda;\boldsymbol{r})\equiv\frac{1}{r}\exp\left[\frac{1}{\hbar}(i\boldsymbol{q}\cdot\boldsymbol{r}-\lambda r)\right]. \tag{A10}$$

In Eq. (A10), $\boldsymbol{q}$ is a complex vector and λ is a positive real scalar. In evaluating the integral (A9) we follow a technique put forward by Hostler [18] in his derivation of the momentum-space CGF, and subsequently adapted by Klarsfeld [19] to the study of two-photon transitions between hydrogenic s states. After inserting into Eq. (A4) the integral representation (A8) of the Bessel function I_0, substitution of Eq. (A4) into Eq. (A9) leads to a space integral of the type

$$\int d^3x\frac{1}{r}\exp(-Ar-\boldsymbol{B}\cdot\boldsymbol{r})=4\pi(A^2-\boldsymbol{B}\cdot\boldsymbol{B})^{-1}\quad(\mathrm{Re}A>|\mathrm{Re}\boldsymbol{B}|). \tag{A11}$$

The integral (A11) is immediate if $\boldsymbol{B}$ is a real vector; for a complex $\boldsymbol{B}$ the result is then obtained by analytic continuation. In our specific case, a sufficient condition for the convergence of the space integral is

$$\lambda>|\mathrm{Im}\boldsymbol{q}|. \tag{A12}$$

After computing successively the space integral with Eq. (A11) and a closed-contour integral over the complex variable s from Eq. (A8) by means of the residue theorem, we change the variable of integration,

$$\rho=\frac{\zeta-1}{\zeta+1}, \tag{A13}$$

and find

$$\mathcal{F}(\Omega,\boldsymbol{q},\lambda;\boldsymbol{r})=\frac{m_e}{\hbar X}\frac{ie^{i\pi\tau}}{2\sin(\pi\tau)}\int_1^{(0+)}d\rho\rho^{-1-\tau}f\times\exp\left(\frac{i}{\hbar}f\boldsymbol{q}\cdot\boldsymbol{r}-g\frac{X}{\hbar}r\right), \tag{A14}$$

with

$$f(X,\boldsymbol{q}^2,\lambda;\rho)\equiv\frac{4X^2\rho}{[X+\lambda+(X-\lambda)\rho]^2+\boldsymbol{q}^2(1-\rho)^2}, \tag{A15}$$

and

$$g(X,\boldsymbol{q}^2,\lambda;\rho)\equiv\frac{(X+\lambda)^2-(X-\lambda)^2\rho^2+\boldsymbol{q}^2(1-\rho^2)}{[X+\lambda+(X-\lambda)\rho]^2+\boldsymbol{q}^2(1-\rho)^2}. \tag{A16}$$

In Eq. (A11) the integration path in the complex ρ plane begins at the point $\rho=1+i0$, where the argument of ρ is zero, then encircles the origin $\rho=0$ in the positive sense within the unit circle, and terminates at the point $\rho=1-i0$.

To the best of our knowledge, the result (A14)–(A16) has been first obtained by Kelsey and Macek [20]. It is worth emphasizing that we have previously utilized Schwinger's integral representation of the GCF in momentum space [21] in order to derive Eqs. (A14)–(A16) [22]. However, the above-presented proof is conveniently straightforward.

APPENDIX B: THE HUMBERT HYPERGEOMETRIC FUNCTION Φ_1

The function Φ_1 is obtained by confluence from the Appell hypergeometric function F_1:

$$\Phi_1(a,b,c;x,y)=\lim_{b'\to\infty} F_1\left(a;b,b';c;x,\frac{y}{b'}\right). \quad \text{(B1)}$$

We recall the double ascending power series of the Appell function F_1 [23],

$$F_1(a;b,b';c;x,x')=\sum_{\nu=0}^{\infty}\sum_{\nu'=0}^{\infty}\frac{(a)_{\nu+\nu'}(b)_\nu(b')_{\nu'}}{(c)_{\nu+\nu'}\nu!\,\nu'!}x^\nu(x')^{\nu'} \quad (|x|<1,\ |x'|<1), \quad \text{(B2)}$$

and its integral representation [24]

$$F_1(a;b,b';c;x,x') = \frac{\Gamma(c)}{\Gamma(a)\Gamma(c-a)}\frac{-ie^{-i\pi a}}{2\sin(\pi a)}\int_1^{(0+)} d\rho\rho^{a-1} \times(1-\rho)^{c-a-1}(1-x\rho)^{-b}(1-x'\rho)^{-b'}$$
$$[\mathrm{Re}(c-a)>0,\quad a\neq 1,2,3,\ldots, \quad |\arg(-x)|\leqslant\pi, \quad |\arg(-x')|\leqslant\pi]. \quad \text{(B3)}$$

In Eq. (B2), as well as throughout this paper and the following one [14], Pochhammer's symbol

$$(a)_n\equiv\frac{\Gamma(a+n)}{\Gamma(a)} \quad \text{(B4)}$$

is utilized. Taking the limit (B1), Eqs. (B2) and (B3) become, respectively [25],

$$\Phi_1(a,b,c;x,y)=\sum_{\nu=0}^{\infty}\sum_{\nu'=0}^{\infty}\frac{(a)_{\nu+\nu'}(b)_\nu}{(c)_{\nu+\nu'}\nu!\,\nu'!}x^\nu y^{\nu'} \quad (|x|<1) \quad \text{(B5)}$$

and

$$\Phi_1(a,b,c;x,y) = \frac{\Gamma(c)}{\Gamma(a)\Gamma(c-a)}\frac{-ie^{-i\pi a}}{2\sin(\pi a)} \times\int_1^{(0+)} d\rho\rho^{a-1}(1-\rho)^{c-a-1}(1-x\rho)^{-b}e^{y\rho}$$
$$[\mathrm{Re}(c-a)>0,\quad a\neq 1,2,3,\ldots,\quad |\arg(-x)|\leqslant\pi]. \quad \text{(B6)}$$

Note that Eq. (B5) can be written out as a simple series either of Kummer hypergeometric functions ${}_1F_1$,

$$\Phi_1(a,b,c;x,y)=\sum_{\nu=0}^{\infty}\frac{(a)_\nu(b)_\nu}{(c)_\nu}\frac{x^\nu}{\nu!} \times{}_1F_1(a+\nu;c+\nu;y) \quad (|x|<1), \quad \text{(B7)}$$

or of Gauss hypergeometric functions ${}_2F_1$,

$$\Phi_1(a,b,c;x,y)=\sum_{\nu'=0}^{\infty}\frac{(a)_{\nu'}}{(c)_{\nu'}}\frac{y^{\nu'}}{\nu'!} \times{}_2F_1(a+\nu',b+\nu';c+\nu';x). \quad \text{(B8)}$$

With the change of the variable of integration

$$\rho=\frac{t}{1-x+xt} \quad (x\neq 1), \quad \text{(B9)}$$

Eq. (B6) yields another useful integral representation:

$$\Phi_1(a,b,c;x,y) =(1-x)^{-a}\frac{\Gamma(c)}{\Gamma(a)\Gamma(c-a)}\frac{-ie^{-i\pi a}}{2\sin(\pi a)} \times\int_1^{(0+)} dt t^{a-1}(1-t)^{c-a-1} \times\left(1-\frac{x}{x-1}t\right)^{b-c}\exp\left(\frac{yt}{1-x+xt}\right)$$
$$\left(\mathrm{Re}(c-a)>0,\ a\neq 1,2,3,\ldots,\ x\neq 1,\ \arg\left|\frac{x}{x-1}\right|<\pi\right). \quad \text{(B10)}$$

We finally remark that the dominant asymptotic behavior of the Kummer function [26],

$${}_1F_1(a;c;z)=\frac{\Gamma(c)}{\Gamma(c-a)}(-z)^{-a}\left[1+O\left(\frac{1}{|z|}\right)\right] +\frac{\Gamma(c)}{\Gamma(a)}e^z z^{a-c}\left[1+O\left(\frac{1}{|z|}\right)\right]$$
$$[|z|\to\infty,\quad -\pi<\arg(z)\leqslant 0], \quad \text{(B11)}$$

leads via Eq. (B7) to the following asymptotically dominant terms of the Humbert function Φ_1:

$$\Phi_1(a,b,c;x,y)=\frac{\Gamma(c)}{\Gamma(c-a)}(-y)^{-a}\left[1+O\left(\frac{1}{|y|}\right)\right] +\frac{\Gamma(c)}{\Gamma(a)}e^y y^{a-c}(1-x)^{-b}\left[1+O\left(\frac{1}{|y|}\right)\right]$$
$$[|x|<1,\ |y|\to\infty,\quad -\pi<\arg(y)\leqslant 0]. \quad \text{(B12)}$$

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