Theory of two-photon emission from atomic inner shells

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We prove from quantum electrodynamics that two-photon emission in transitions between atomic inner shells can be treated in lowest-order approximation in the single-particle model. By taking many-body effects into account we show that, in the reduced single-particle model, the Pauli exclusion principle does *not* prohibit summing over *all* possible intermediate states, including core states. Some transition-rate formulas are presented that are useful for numerical calculations.

I. INTRODUCTION

Recent developments in experimental techniques have made it possible to measure two-photon transitions between atomic inner-shell vacancy states.[1] The results must be interpreted relativistically. Existing theoretical work has been based on the hydrogenic[2-4] or nonrelativistic self-consistent-field (SCF) (Refs. 4–6) models, except for the recent relativistic SCF calculation of Mu and Crasemann.[7] The latter work suggests exploration of some interesting questions: Is the Pauli exclusion principle violated if one includes occupied orbitals among the intermediate states, as done in Ref. 7? What are the theoretical similarities and differences between two-photon processes in hydrogenic versus inner-shell atomic systems? In this paper we address these questions. In Sec. II, we analyze the theory of two-photon transitions on the basis of quantum electrodynamics (QED). The possible intermediate states are identified automatically; they can be classified into two types according to time ordering. One type is advanced: The electron final state occurs when the intermediate states do, which is in advance of the disappearance of the initial state. The second type is retarded: The time order for the electron is first initial state, then intermediate states, then the final state. In this derivation, the Pauli exclusion principle is built in by commutation relations of operators. The final expression for the S matrix element shows that the problem can be formally treated as a time-independent one, with the electron moving in the field of the nucleus and the other electrons. In the reduced model, since there is no time-order difference, the original two types of intermediate states patch up a new set of intermediate states which is a complete set. The Pauli principle is thus seen to allow summation over all possible intermediate states, including occupied inner-shell states.

In Sec. III, a time-independent many-body approach is delineated. In Sec. IV, as an application of the results of earlier sections, we provide formulas for matrix elements, transition rates, and differential equations to be solved, which are formally the same as for the hydrogenic case. The only difference is the external potential acting on the electron. It is not that an inner-shell vacancy is trivially equivalent to the hydrogenic case; there are many-body effects in inner shells, particularly the restrictions imposed by the Pauli exclusion principle, while the hydrogenic system consists of just a single bound electron. Nor should the reader think that this paper's result is just a simple repetition of the conclusions of Brown *et al.*[8] regarding γ-ray scattering, which are based on a single-electron, i.e., hydrogenic model. If we consider many-body effects and the built-in Pauli exclusion principle in inner-shell γ-ray elastic scattering, the core contributions should in fact be *excluded* when summing over intermediate states (unlike the procedure in Ref. 8). By contrast, as shown here, *all* intermediate states must be included (as done in Ref. 7) when one calculates two-photon inner-shell transition probabilities, taking the Pauli exclusion principle into account.

II. QUANTUM-ELECTRODYNAMIC APPROACH AND THE QUESTION OF INTERMEDIATE STATES

The dominant part of two-photon emission is of the double electric dipole ($2E1$) type; this is the only kind considered here.

In the theory of QED, the second-order scattering operator S can be expressed in Furry's representation[9] as

$$S=-\frac{\alpha}{2}\int\int d^4x\,d^4x' T(j^{\mu}(x)j^{\nu}(x')A_{\mu}(x)A_{\nu}(x')) . \quad (1)$$

The matrix element is

$$S_{21}=\langle 0\,|\,\langle \text{core}\,|\,c_1c_2a_2^{\dagger}Sa_1\,|\,\text{core}\rangle\,|\,0\rangle , \quad (2)$$

where $|\,\text{core}\rangle$, in the lowest-order approximation, stands for an antisymmetrically filled core wave function, and $|\,0\rangle$, for a photon vacuum state. We denote photon creation and annihilation operators by $c^{\dagger}$ and c, respectively, and electron creation and annihilation operators by $a^{\dagger}$ and a. The remainder of the notation is of standard usage. Natural units ($\hbar=c=1$) are employed throughout. $\alpha=1/137.036$.

The electron field operator can be expressed as

$$\begin{aligned}\psi(t,\mathbf{r})&=\sum_a a_a\psi_a(\mathbf{r})\exp(-i\varepsilon_a t)\\&\quad+\sum_n a_n\psi_n(\mathbf{r})\exp(-i\varepsilon_n t) ,\\ \bar{\psi}(t,\mathbf{r})&=\sum_a a_a^{\dagger}\bar{\psi}_a(\mathbf{r})\exp(i\varepsilon_a t)+\sum_n a_n^{\dagger}\bar{\psi}_n(\mathbf{r})\exp(i\varepsilon_n t) .\end{aligned} \quad (3)$$

The summation index a runs over all core states, and n, over all the excited states.

After time-ordered contraction manipulation, the S matrix element takes the form

$$S_{21}=-i\alpha\int\int d^4x\,d^4x'\bar{\psi}_1(x')\gamma^\nu G(x,x')\gamma^\mu\psi_2(x) \times(A^*_{1\mu}A'^*_{2\nu}+A^*_{2\mu}A'^*_{1\nu}), \tag{4}$$

where

$$G(x,x')=-i\langle\text{core}\mid T\bar{\psi}(x)\psi(x')\mid\text{core}\rangle \tag{5}$$

is the propagator for an electron hole and $\psi_1(x)$ and $\psi_2(x)$ are bispinor wave functions. The indexes 1 and 2 denote the initial and final states of electrons, respectively.

The explicit form for the electron-hole propagator can be shown to be

$$G(x,x')=\frac{1}{2\pi}\left[\sum_{a(\in f)}\int_{-\infty}^{+\infty}d\varepsilon\frac{\psi_a(\mathbf{r}')\bar{\psi}_a(\mathbf{r})}{\varepsilon+\varepsilon_a+i\eta}e^{-i\varepsilon(t-t')}-\sum_{n(>f)}\int_{-\infty}^{+\infty}d\varepsilon\frac{\psi_n(\mathbf{r}')\bar{\psi}_n(\mathbf{r})}{\varepsilon-\varepsilon_n+i\eta}e^{-i\varepsilon(t'-t)}\right]. \tag{6}$$

The first term in the bracket on the right-hand side of Eq. (6), which involves core intermediate states, is an advanced Green's function which describes the $t>t'$ process; the second term, which involves excited intermediate states, is a retarded Green's function which describes the $t'>t$ process. This distinction is important in understanding the physical meaning of the two types of intermediate states.

The two-photon processes can be described by Feynman diagrams. We specify the initial and final hole states in the following examples. Time progresses from left to right. A solid line with arrow opposite to the time direction represents an electron hole, a line with arrow in the time direction represents a real electron. There are two equivalent ways of expressing the advanced process, as shown in Fig. 1. Figure 1(a) describes the process in terms of a hole, and Fig. 1(b), in terms of an electron. For the retarded process, there are two similar diagrams, shown in Fig. 2.

From this analysis, we obtain only two types of intermediate states. Two other types of intermediate states could in principle be constructed, but do not occur in our derivation. One of these pertains to retarded real core states; the disappearance of this type embodies the Pauli exclusion principle. The other type of intermediate that does not appear here involves advanced excited hole states; this type is excluded by the fact that an excited state cannot be annihilated from the core.

Electromagnetic waves and electron waves have the forms

$$\begin{aligned}&A_1(x)=A_1(\mathbf{r})e^{-i\omega_1 t},\quad A_2(x)=A_2(\mathbf{r})e^{-i\omega_2 t};\\&\psi_1(x)=\psi_1(\mathbf{r})e^{-i\varepsilon_1 t},\quad \psi_2(x)=\psi_2(\mathbf{r})e^{-i\varepsilon_2 t},\end{aligned} \tag{7}$$

respectively. After we integrate over time t, then t', and then energy, we find the following expression for the S matrix element:

$$\begin{aligned}S_{21}=-i2\pi\alpha\int d^3r\int d^3r'\bar{\psi}_1(\mathbf{r}')\gamma^\nu\Bigg[&\sum_a\frac{\psi_a(\mathbf{r}')\bar{\psi}_a(\mathbf{r})}{\omega_1-\varepsilon_2+\varepsilon_a+i\eta}A^*_{1\mu}(\mathbf{r})A^*_{2\nu}(\mathbf{r}')\\&-\sum_n\frac{\psi_n(\mathbf{r}')\bar{\psi}_n(\mathbf{r})}{\varepsilon_1+\omega_2-\varepsilon_n+i\eta}A^*_{1\mu}(\mathbf{r})A^*_{2\nu}(\mathbf{r}')+\sum_a\frac{\psi_a(\mathbf{r}')\bar{\psi}_a(\mathbf{r})}{\omega_2-\varepsilon_2+\varepsilon_a+i\eta}A^*_{2\mu}(\mathbf{r})A^*_{1\nu}(\mathbf{r}')\\&-\sum_n\frac{\psi_n(\mathbf{r}')\bar{\psi}_n(\mathbf{r})}{\varepsilon_1+\omega_1-\varepsilon_n+i\eta}A^*_{2\mu}(\mathbf{r})A^*_{1\nu}(\mathbf{r}')\Bigg]\gamma^\mu\psi_2(\mathbf{r})\delta(\varepsilon_2-\varepsilon_1-\omega_1-\omega_2).\end{aligned} \tag{8}$$

The δ-function factor expresses the energy conservation law

$$\varepsilon_2-\varepsilon_1=\omega_1+\omega_2. \tag{9}$$

By using this energy conservation condition, we can rewrite the energy denominators. It then becomes possible to combine the two types of Green's function into one with a complete set of wave functions as its intermediate states:

$$\begin{aligned}S_{21}=-i2\pi\alpha\int\int d^3\mathbf{r}\,d^3r'\bar{\psi}_1(\mathbf{r}')\gamma^\nu\Bigg[&\sum_i\frac{\psi_i(\mathbf{r}')\bar{\psi}_i(\mathbf{r})}{\varepsilon_i+\omega_1-\varepsilon_2}A^*_{1\mu}(\mathbf{r})A^*_{2\nu}(\mathbf{r}')\\&+\sum_i\frac{\psi_i(\mathbf{r}')\bar{\psi}_i(\mathbf{r})}{\varepsilon_i+\omega_2-\varepsilon_2}A^*_{2\mu}(\mathbf{r})A^*_{1\nu}(\mathbf{r}')\Bigg]\gamma^\mu\psi_2(\mathbf{r})\delta(\varepsilon_2-\varepsilon_1-\omega_1-\omega_2).\end{aligned} \tag{10}$$

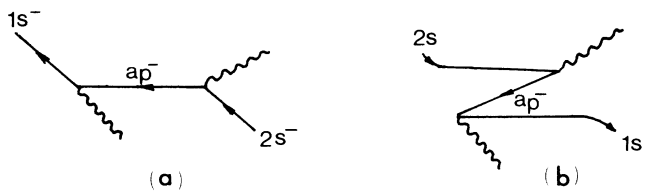

FIG. 1. Feynman diagrams for the advanced process (a) in terms of a hole, (b) in terms of an electron.

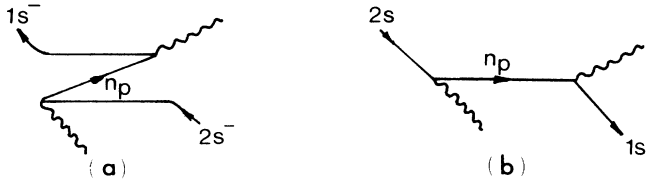

FIG. 2. Feynman diagrams for the retarded process (a) in terms of a hole, (b) in terms of an electron.

Here, the index i runs over all possible intermediate states. The expression for the S matrix element is thus reduced to a time-independent single-particle version, which is formally the same as in the hydrogenic model if we call the electron 2 the initial state, the electron 1 the final state.

The only differences between the two models are in two aspects: (1) The hydrogenic electron moves in the Coulomb field of the nucleus, while the electron displaced by the inner-shell hole moves in the combined field of the nucleus and the remaining electrons. (2) The summing and averaging over initial and final states, respectively, are opposite in the inner-shell case to that of outer shells. We have thus proved a theorem which is useful in deriving formulas for numerical calculations.

Theorem I. $E1$ photon emission from atomic inner shells, filling one vacancy, can be expressed in the lowest-order approximation in a single-particle formulation which is formally the same as in the hydrogenic case, with the electron hole being treated as a real electron. The only differences are in the potential acting on the electron and the treatment of summing and averaging over the initial and final states.

III. TIME-INDEPENDENT MANY-BODY APPROACH

In a time-independent many-body approach, the initial and final states can be written as

$$|1\rangle = a_1 |\text{core}\rangle, \quad |2\rangle = a_2 |\text{core}\rangle . \tag{11}$$

As discussed in Sec. II, we have two types of intermediate states, advanced and retarded. In time-independent many-body perturbation theory, these can be written as

$$a_a |\text{core}\rangle \tag{12}$$

and

$$a_n^\dagger a_2 a_1 |\text{core}\rangle . \tag{13}$$

The dipole operator has the form

$$Q^m = \sum_{i,j} q_{ij}^m a_i^\dagger a_j , \tag{14}$$

where m is the magnetic quantum number. The time-independent many-body expression for the emission rate is

$$\frac{dW}{d\omega_1} = \omega_1^3 \omega_2^3 \frac{8\alpha^2}{9\pi} \sum_{m_1,m_2} \left| \sum_i \frac{\langle 2|Q^{m_2}|i\rangle\langle i|Q^{m_1}|1\rangle}{E_1 - E_i - \omega_1} + \sum_i \frac{\langle 2|Q^{m_1}|i\rangle\langle i|Q^{m_2}|1\rangle}{E_1 - E_i - \omega_2} \right|^2 . \tag{15}$$

After contractions, this form is reduced to a single-body form

$$\frac{dW}{d\omega_1} = \omega_1^3 \omega_2^3 \frac{8\alpha^2}{9\pi} \sum_{m_1,m_2} \left| \sum_a \frac{q_{a2}^{m_2} q_{1a}^{m_1}}{-\varepsilon_1 + \varepsilon_a - \omega_2} + \sum_a \frac{q_{a2}^{m_1} q_{1a}^{m_2}}{-\varepsilon_1 + \varepsilon_a - \omega_2} - \sum_n \frac{q_{1n}^{m_2} q_{n2}^{m_1}}{\varepsilon_2 - \varepsilon_n - \omega_1} - \sum_n \frac{q_{1n}^{m_1} q_{n2}^{m_2}}{\varepsilon_2 - \varepsilon_n - \omega_2} \right|^2 . \tag{16}$$

The many-body forms of the two types of intermediate states are now reduced to core states and excitations in the single-body form. If we impose the energy conservation law on Eq. (9), the Green's function that involves core states and the one that involves excitations are combined into a new Green's function that involves a complete set of intermediate states. This proves Theorem I over again. We therefore have the final form for the emission rate:

$$\frac{dW}{d\omega_1} = \omega_1^3 \omega_2^3 \frac{8\alpha^2}{9\pi} \sum_{m_1,m_2} \left| \sum_i \frac{q_{1i}^{m_2} q_{i2}^{m_1}}{\varepsilon_i - \varepsilon_2 + \omega_1} + \sum_i \frac{q_{1i}^{m_1} q_{i2}^{m_2}}{\varepsilon_i - \varepsilon_2 + \omega_2} \right|^2 . \tag{17}$$

If we specify the initial state for the atom as a [1s] hole

state, the possible final states will be $[2s]$, $[3s],[3d_{3/2}],[3d_{5/2}],[4s],\ldots$, hole states. The possible intermediate states are of two types. The ones derived from retardation are of the type of single-hole p state of the core, and the ones derived from advancement are of the type of state with one excited p electron and two holes in the core. This agrees with the classification of Åberg,[5] and of Freund and Bannett.[6] Robinson,[10] and Crance and Aymar[11] had similar treatments and conclusions for core excitations in outer-shell problems.

The time-independent many-body approach leads to the same result as the QED approach. It is simpler, but involves some imposed conditions, such as those regarding the intermediate states and the energy conservation law. It cannot reveal the time-ordered process, and hence leads to less physical insight.

IV. MATRIX ELEMENTS AND EMISSION RATES

Theorem I reduces the inner-shell-vacancy two-photon-decay problem to one tractable in the single-particle model. The formulas for the hydrogenic case can all be adapted to the many-body problem, but the electron wave functions must be calculated for different potentials. A formalism for relativistic two-photon emission in the hydrogenic case has been given by Goldman and Drake.[3] We follow their approach and that of Johnson,[2] which is equivalent to the approach of Brown *et al.*[8] used in Ref. 7. After integrating over the angular distribution, averaging over all possible final states, and summing over all possible initial states, the $2E1$ decay rates per unit time in differential form are as follows. For $s\rightarrow s$,

$$\frac{dW}{d\omega_1}=\frac{8\alpha^2}{27\pi}\omega_1^3\omega_2^3\{\tfrac{1}{3}[E_1^2(\omega_1,\omega_2)+E_1^2(\omega_2,\omega_1)]+\tfrac{2}{3}[E_{-2}^2(\omega_1,\omega_2)+E_{-2}^2(\omega_2,\omega_1)]-\tfrac{2}{9}E_1(\omega_1,\omega_2)E_1(\omega_2,\omega_1)$$
$$+\tfrac{4}{9}E_{-2}(\omega_1,\omega_2)E_{-2}(\omega_2,\omega_1)+\tfrac{8}{9}[E_{-2}(\omega_1,\omega_2)E_1(\omega_2,\omega_1)+E_1(\omega_1,\omega_2)E_{-2}(\omega_2,\omega_1)]\}\ . \tag{18}$$

For $d_{3/2}\rightarrow s$,

$$\frac{dW}{d\omega_1}=\frac{8\alpha^2}{27\pi}\omega_1^3\omega_2^3\{\tfrac{2}{3}[E_1^2(\omega_1,\omega_2)+E_1^2(\omega_2,\omega_1)]+\tfrac{2}{15}[E_{-2}^2(\omega_1,\omega_2)+E_{-2}^2(\omega_2,\omega_1)]+\tfrac{8}{9}E_1(\omega_1,\omega_2)E_1(\omega_2,\omega_1)$$
$$-\tfrac{8}{45}E_{-2}(\omega_1,\omega_2)E_{-2}(\omega_2,\omega_1)+\tfrac{4}{9}[E_{-2}(\omega_1,\omega_2)E_1(\omega_2,\omega_1)+E_1(\omega_1,\omega_2)E_{-2}(\omega_2,\omega_1)]\}\ . \tag{19}$$

For $d_{5/2}\rightarrow s$,

$$\frac{dW}{d\omega_1}=\frac{8\alpha^2}{27\pi}\omega_1^3\omega_2^3\{\tfrac{6}{5}[E_{-2}^2(\omega_1,\omega_2)+E_{-2}^2(\omega_2,\omega_1)]+\tfrac{12}{5}E_{-2}(\omega_1,\omega_2)E_{-2}(\omega_2,\omega_1)\}\ . \tag{20}$$

The transition amplitude is given by

$$E_\kappa(\omega_1,\omega_2)=\frac{3}{\omega_2}\int_0^\infty dr[S_{\kappa\kappa'}(r,\omega_1)U_\kappa(r,\omega_2)+T_{\kappa\kappa'}(r,\omega_1)V_\kappa(r,\omega_2)]\ , \tag{21}$$

where $S_{\kappa\kappa'}$ and $T_{\kappa\kappa'}$ are solutions of the perturbed Dirac equations

$$[m-\varepsilon_1+\omega_1+V(r)]S_{\kappa\kappa'}(r,\omega_1)+\left[\frac{d}{dr}-\frac{\kappa}{r}\right]T_{\kappa\kappa'}(r,\omega_1)=\frac{3}{\omega_1}K_{\kappa\kappa'}(r,\omega_1)\ ,$$
$$-\left[\frac{d}{dr}+\frac{\kappa}{r}\right]S_{\kappa\kappa'}(r,\omega_1)-[m+\varepsilon_1-\omega_1-V(r)]T_{\kappa\kappa'}(r,\omega_1)=\frac{3}{\omega_1}L_{\kappa\kappa'}(r,\omega_1)\ . \tag{22}$$

The inhomogeneous terms $K_{\kappa\kappa'}$ and $L_{\kappa\kappa'}$ and the terms U_κ and V_κ in the integrand of Eq. (22) are

$$K_{\kappa\kappa'}(r,\omega_1)=\begin{cases}\dfrac{\kappa-\kappa'}{2}\left[j_1'(\omega_1 r)+\dfrac{j_1(\omega_1 r)}{\omega_1 r}\right]F_2(r)-\dfrac{j_1(\omega_1 r)}{\omega_1 r}F_2(r) & \text{(Coulomb gauge)}\\ j_1(\omega_1 r)G_2(r)-\left[\dfrac{\kappa-\kappa'}{2}+1\right]j_2(\omega_1 r)F_2(r) & \text{(length gauge)}\,,\end{cases}$$

$$L_{\kappa\kappa'}(r,\omega_1)=\begin{cases}\dfrac{\kappa-\kappa'}{2}\left[j_1'(\omega_1 r)+\dfrac{j_1(\omega_1,r)}{\omega_1 r}\right]G_2(r)-\dfrac{j_1(\omega_1 r)}{\omega_1 r}G_2(r) & \text{(Coulomb gauge)}\\ j_1(\omega_1 r)F_2(r)-\left[\dfrac{\kappa-\kappa'}{2}-1\right]j_2(\omega_1 r)G_2(r) & \text{(length gauge)}\,,\end{cases} \tag{23}$$

$$U_{\kappa}(r,\omega_2)=\begin{cases}-\dfrac{\kappa+1}{2}\left[j_1'(\omega_2 r)+\dfrac{j_1(\omega_2 r)}{\omega_2 r}\right]F_1(r)+\dfrac{j_1(\omega_2 r)}{\omega_2 r}F_1(r) & \text{(Coulomb gauge)}\\ j_1(\omega_2 r)G_1(r)+\left[\dfrac{\kappa+1}{2}+1\right]j_2(\omega_2 r)F_1(r) & \text{(length gauge)}\,,\end{cases}$$

$$V_{\kappa}(r,\omega_2)=\begin{cases}-\dfrac{\kappa+1}{2}\left[j_1'(\omega_2 r)+\dfrac{j_1(\omega_2 r)}{\omega_2 r}\right]G_1(r)-\dfrac{j_1(\omega_2 r)}{\omega_2 r}G_1(r) & \text{(Coulomb gauge)}\\ j_1(\omega_2 r)F_1(r)+\left[\dfrac{\kappa+1}{2}-1\right]j_2(\omega_2 r)G_1(r) & \text{(length gauge)}\,.\end{cases} \tag{24}$$

Here $G_a(r)$ and $F_a(r)$ are the large and small components of the electron wave function, satisfying the radial equations

$$\begin{aligned}&[m-\varepsilon_a+V(r)]G_a(r)+\left[\frac{d}{dr}-\frac{\kappa}{r}\right]F_a(r)=0\,,\\ &-\left[\frac{d}{dr}+\frac{\kappa}{r}\right]G_a(r)-[m+\varepsilon_a-V(r)]F_a(r)=0\,.\end{aligned} \tag{25}$$

These formulas are formally identical to those for the hydrogenic case. The only difference is in the potential $V(r)$. In Eqs. (22) and (25), $V(r)$ is the potential due to the nucleus and the other electrons, but we do not exclude the core contribution in the inhomogeneous equations (22). In the hydrogenic case, $V(r)$ is the pure Coulomb potential due to the nucleus. The notation used here is the same as that in the paper of Parpia and Johnson.[2] If we set $\kappa'=-1$ in Eq. (23), we obtain the $s\rightarrow s$ decay rate, which is the same as the formula of Parpia and Johnson.[2] If we set $\kappa'=2$ and -3, we find the $d_{3/2}\rightarrow s$ and $d_{5/2}\rightarrow s$ decay rates.

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