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Two-photon transitions in atomic hydrogen: An alternative approach

Ramesh Babu Thayyullathil and K. J. Prasanna*
Department of Physics, Cochin University of Science and Technology, Cochin 682-022, India

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We present an efficient analytic approach for the evaluation of two-photon transition rates in a hydrogen atom. The method is a variation of the Dalgarno and Lewis method [Proc. R. Soc. London Ser. A **233**, 70 (1955)] for the evaluation of the second-order Stark effect in a hydrogen atom.

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The problem of gauge invariance in multiphoton transitions was illustrated by calculating the two-photon transition rates in hydrogen atoms in length and velocity gauges [1]. These explicit calculations demonstrated that the gauge invariance can be maintained only by using a complete set of intermediate states. This clearly indicates the need for an accurate evaluation of the sum of the contributions from various intermediate states.

Different methods [1,2] can be used to evaluate the sum over the intermediate states very accurately. In the present work we repeat the calculations of Ref. [1] using a differential-equation approach. This method is an extension of Dalgarno and Lewis [3] treatment of second-order Stark effect in a hydrogen atom. This procedure has already been used for the evaluation of logarithmic mean excitation energies for hydrogen and helium [4]. In this approach the sum over the intermediate states is completely absent. It is also an elegant method and can be generalized to other systems.

The two-photon transition rate W_i^f from an initial state $|i\rangle$ to a final state $|f\rangle$ in the dipole approximation can be given as

$$W_i^f \propto |\mathcal{D}_i^f|^2 \delta(\hbar\omega_1+\hbar\omega_2-E_{fi}) \ , \tag{1}$$

where

$$\mathcal{D}_i^f=\frac{e^2}{2a}\sum_I\left\{\frac{\left\langle f\left|\frac{z}{a}\right|I\right\rangle\left\langle I\left|\frac{z}{a}\right|i\right\rangle}{E_I-E_i-\hbar\omega_1}+\frac{\left\langle f\left|\frac{z}{a}\right|I\right\rangle\left\langle I\left|\frac{z}{a}\right|i\right\rangle}{E_I-E_i-\hbar\omega_2}\right\} . \tag{2}$$

where a is the Bohr radius and for simplicity we take both the beams polarized along the z axis. The factor $e^2/2a$ is introduced to make $\mathcal{D}_i^f$ dimensionless. The rate calculation involves the evaluation of the discrete and the continuum intermediate sum.

Our method depends on finding operators $F^{(1)}$ and $F^{(2)}$ such that

$$\frac{z}{a}|i\rangle=\frac{2a}{e^2}[H_0F^{(1)}-F^{(1)}H_0-\hbar\omega_1F^{(1)}]|i\rangle \ , \tag{3}$$

$$\frac{z}{a}|i\rangle=\frac{2a}{e^2}[H_0F^{(2)}-F^{(2)}H_0-\hbar\omega_2F^{(2)}]|i\rangle \ , \tag{4}$$

where $H_0=\mathbf{p}^2/2m-e^2/r$ is the unperturbed hydrogen-atom Hamiltonian. From Eq. (3) we have

$$\left\langle I\left|\frac{z}{a}\right|i\right\rangle=\frac{2a}{e^2}[E_I+E_i-\hbar\omega_1]\langle I|F^{(1)}|i\rangle \ . \tag{5}$$

Using a similar equation for $F^{(2)}$ we get

$$\mathcal{D}_i^f=\left\langle f\left|\frac{z}{a}F^{(1)}\right|i\right\rangle+\left\langle f\left|\frac{z}{a}F^{(2)}\right|i\right\rangle \ . \tag{6}$$

Using $|i\rangle=\psi_i(\mathbf{r})$, $F^{(1)}=F^{(1)}(\mathbf{r})$ satisfies the equation

$$\frac{2a}{e^2}[H_0(F^{(1)}\psi_i(\mathbf{r}))-F^{(1)}H_0\psi_i(\mathbf{r})-\hbar\omega_1F^{(1)}\psi_i(\mathbf{r})]=\frac{z}{a}\psi_i(\mathbf{r}) \ . \tag{7}$$

Further considering the transitions from ground state $\psi_i(\mathbf{r})=\psi_{1s}(\mathbf{r})$ and using $F^{(1)}(\mathbf{r})=f^{(1)}(r)z/a$ and using Eq. (7), we find that $f^{(1)}(r)$ satisfies the equation

$$\rho\frac{d^2f^{(1)}(\rho)}{d\rho^2}+(4-2\rho)\frac{df^{(1)}(\rho)}{d\rho}+(\nu_1\rho-2)f^{(1)}(\rho)=-\rho \ , \tag{8}$$

where $\rho=r/a$ and $(e^2/2a)\nu_1=\hbar\omega_1$. A similar equation can be obtained for $f^{(2)}(r)$ where $F^{(2)}(\mathbf{r})=f^{(2)}(r)z/a$ with ν_1 replaced by ν_2 in Eq. (8). Thus in general we need a solution for an equation of the form

$$\rho f''(\rho)+(4-2\rho)f'(\rho)+(\nu\rho-2)f(\rho)=-\rho \ . \tag{9}$$

This equation and the solution to this equation have been extensively used in Ref. [4] for the evaluation of logarithmic mean excitation energies in hydrogen atoms. But we use a different integral representation for the solution. With this representation we can easily exhibit the singularities of the solution as a function of ν. The solution to Eq. (9), which is finite at $\rho=0$ and does not go to infinity faster than e^{ρ} as $\rho\rightarrow\infty$, can be given as

$$f(\rho)=\frac{1}{\nu}\left\{\frac{2}{\nu}B(p,q,\rho)-1\right\} \tag{10}$$

with

$$B(p,q,\rho)=e^{\rho}\int_\lambda^1 dz\, e^{-\rho z}\left[\frac{z+\lambda}{1+\lambda}\right]^{p-1}\left[\frac{z-\lambda}{1-\lambda}\right]^{q-1} \ , \tag{11}$$

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*Present address: Physics Department, S. N. College, Chertalla 688-582, India.

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where $\lambda=\sqrt{1-\nu}$, $p=2+1/\lambda$, and $q=2-1/\lambda$. The function $B(p,q,\rho)$ defined by the equation exists only for $q>0$. That this is a solution can be explicitly verified by substituting Eqs. (11) and (10) in Eq. (9) and assuming $q>0$. Similar to the definition of Γ function using the contour integral, a contour with branch point at $z=\lambda$ can be envisaged for this function also. This can be used to define this function for $q<0$ also. But we approach this problem from a practical computational point of view. Assuming $q>0$ from Eq. (11) after performing a partial integration we get

$$B(p,q,\rho)=\frac{1-\lambda}{q}\left\{1+\rho B(p,q+1,\rho)-\frac{p-1}{1+\lambda}B(p-1,q+1,\rho)\right\} . \tag{12}$$

Just as the relation $\Gamma(q)=\Gamma(q+1)/q$ can be used to analytically continue the Γ function for $q<0$, we use the recurrence relation given in Eq. (12) to define $B(p,q,\rho)$ for $q<0$. By repeatedly using the recurrence relation given in Eq. (12) we see that the solution has simple poles at $q=0,\ -1,\ -2,\dots,-\infty$. These values of q corresponds to $\nu=(n^2-1)/n^2$, where $n=2,3,\dots,\infty$. In other words at these values of ν the incident photon energies are in resonance with the intermediate states. This is what we expect and our solution has got this important property. At these values of ν, p, and q are integers and the integral in Eq. (11) has a closed expression. Thus one can obtain an expression for the residues in a closed form. Another important property of our solution is the form of the limiting solution when $\nu\rightarrow 0$. After using the recurrence relation in Eq. (12) twice this limiting solution $f(\rho)$ is found to be $(1+\rho/2)/2$. This is exactly the solution for the problem of second-order stark effect in a hydrogen atom [3].

From Eq. (6), using $\psi_f(\mathbf{r})=R_{nl}(r)Y_{lm}(\theta,\phi)$ for transitions to the $l=0$ state, we get

$$\mathcal{D}_{1s}^{ns}=(2/3)d_{1s}^{ns} , \tag{13}$$

where

$$d_{1s}^{ns}=\left\{\frac{4}{n^3}\right\}^{1/2}\int_0^\infty d\rho\, e^{-\rho(1+1/n)}\rho^4\{f^{(1)}(\rho)+f^{(2)}(\rho)\}F(1-n,2,\rho/n) . \tag{14}$$

Using Eq. (11) in Eq. (14) we obtain

$$d_{1s}^{ns}=\left\{\frac{4}{n^3}\right\}^{1/2}\left[\frac{1}{\nu_1}\left[\frac{2}{\nu_1}\mathcal{B}(p_1,q_1,\nu_1)-\frac{\Gamma(5)}{(1+1/n)^5}F(1-n,5,2,2/(1+n))\right]+\frac{1}{\nu_2}\left[\frac{2}{\nu_2}\mathcal{B}(p_2,q_2,\nu_2)-\frac{\Gamma(5)}{(1+1/n)^5}F(1-n,5,2,2/(1+n))\right]\right] , \tag{15}$$

where $\lambda_1=\sqrt{1-\nu_1}$, $p_1=2+1/\lambda_1$, $q_1=2-1/\lambda_1$, and $\mathcal{B}(p_1,q_1,\nu_1)$ is given by

$$\mathcal{B}(p_1,q_1,\nu_1)=\Gamma(5)\int_{\lambda_1}^{1}dz\left\{\frac{z+\lambda_1}{1+\lambda_1}\right\}^{p_1-1}\left\{\frac{z-\lambda_1}{1-\lambda_1}\right\}^{q_1-1}\frac{F(1-n,5,2,2/(1+nz))}{(z+1/n)^5} , \tag{16}$$

TABLE I. Dimensionless two-photon transition-rate amplitudes for $1s$-$3s$ transition in a hydrogen atom for a number of values of ν_1.

ν_1	d_{1s}^{3s}
0.3750	−3.2354
0.6750	−1.6693
0.6875	−0.69628
0.7000	0.98470
0.7125	4.15829
0.7250	11.2162
0.7375	34.2263
0.7475	226.8138
0.7650	−58.2000
0.8000	−38.3099
0.8250	−46.57966
0.8500	−74.4204
0.8750	−219.9846
0.8860	−1117.238

TABLE II. Dimensionless two-photon transition-rate amplitudes for $1s$-$3d$ transition in a hydrogen atom for a number of values of ν_1.

ν_1	d_{1s}^{3d}
0.3750	0.7020
0.6750	0.6353
0.6875	0.5687
0.7000	0.4515
0.7125	0.2295
0.7250	−0.2604
0.7375	−1.8352
0.7475	−14.8543
0.7650	4.2968
0.8000	2.5551
0.8250	2.2368
0.8500	1.1822
0.8750	−8.5601
0.8860	−84.9263

and similar expressions can be obtained for $\mathcal{B}(p_2,q_2,\nu_2)$ by replacing ν_1 with ν_2. Because of energy conservation $\nu_2=1-1/n^2-\nu_1$. As we mentioned earlier the integral in Eq. (16) exists only for $q_1>0$. For $q_1<0$ we have to use the relation given in Eq. (12). For transitions to $l=2$ states we have

$$\mathcal{D}_{1s}^{nd}=(16/45)^{1/2}d_{1s}^{nd}\,, \tag{17}$$

where

$$d_{1s}^{nd}=\frac{1}{15n^4}\left[\frac{(n+2)!}{(n-3)!}\right]^{1/2}\int_0^\infty dp\, e^{-p(1+1/n)}p^6\{f^{(1)}(p)+f^{(2)}(p)\}F(3-n,6,2p/n) \tag{18}$$

In Eqs. (13) and (17) we have factored out some numerical factors, which is twice the contribution of the angular integral in Eq. (6). With this our results can be directly compared with that of Ref. [1]. The integration of Eq. (16) has to be performed numerically. Similar results can be obtained for transitions to $l=2$ states also.

In Table I, we present the values of d_{1s}^{3s} for the 1s-3s transitions. These values are exactly the same as in Ref. [1]. We also report in Table II the values of d_{1s}^{3d} for the 1s-3d transitions for the same values of ν_1 as in the 1s-3s transitions.

The conclusions are the same as those in Ref. [1]. But the calculations here involve a simple one-dimensional numerical integration. This can be generalized to other complex systems also. Given an unperturbed Hamiltonian we need not solve for a complete set of eigenstates to calculate the transition rates. What we require is the initial and the final states. With this Eq. (7) can be solved numerically to obtain F. Using this F the transition rate can be calculated using Eq. (6).

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