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Two-photon E1M1 and E1E2 transitions between 2p and 1s levels in hydrogen

L. Labzowsky[1,2], D. Solovyev[1], G. Plunien[3,a], and G. Soff[3,b]

[1] Department of Physics, St. Petersburg State University, 198504 Petrodvorets, St. Petersburg, Russia
[2] Petersburg Nuclear Physics Institute, 188350 Gatchina, St. Petersburg, Russia
[3] Institut für Theoretische Physik, Technische Universität Dresden, Mommsenstrasse 13, 01062 Dresden, Germany

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Abstract. Two-photon transitions in the hydrogen atom are analytically evaluated within the nonrelativistic limit utilizing the Coulomb Green function method. The two-photon emission probability for the transition process $2s \rightarrow 2\gamma(E1) + 1s$ serves as a test for the other calculations and was compared with the results of previous analytical and numerical calculations. The two-photon emission probabilities for the processes $2p \rightarrow \gamma(E1) + \gamma(M1) + 1s$ and $2p \rightarrow \gamma(E1) + \gamma(E2) + 1s$ are also evaluated and compared with previous numerical calculations. Different nonrelativistic "forms" for the decay probabilities in combination with different gauge choices are considered.

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1 Introduction

The probabilities for the spontaneous two-photon decay in hydrogen atoms and hydrogenlike ions are under investigation since the theoretical formalism has been introduced by Göppert-Mayer [1] and a first estimate for the two-photon E1E1 transition $2s \rightarrow 2\gamma(E1) + 1s$ has been presented by Breit and Teller [2]. A highly accurate calculation of the E1E1-transition probability has been performed by Klarsfeld [3]. Relativistic corrections to his result have been obtained by Drake and Goldman [4] and by Parpia and Johnson [5], while the recoil corrections have been provided in the papers by Fried and Martin [6] and by Bacher [7]. An accurate fully relativistic calculation of $2s$–$1s$ transition in H-like ions with the partial inclusion of QED corrections, as well as nuclear size and polarization corrections was performed in [8]. Karshenboim and Ivanov [9–11] evaluated the leading logarithmic contribution to radiative corrections to this decay. Recently Jentschura [12] performed a complete evaluation of these radiative corrections. The two-photon decay probabilities for processes $ns \rightarrow 2\gamma(E1) + 1s$ with $n = 3$–6 in hydrogen have been calculated in [13]. The present paper follows these calculations and is devoted to the evaluation of the probabilities for two-photon decays $2p \rightarrow \gamma(E1) + \gamma(M1) + 1s$ and $2p \rightarrow \gamma(E1) + \gamma(E2) + 1s$, respectively. Evaluations of the two latter transitions have been first accomplished in references [14,15] for hydrogenlike systems with nuclear charge numbers Z within the range $1 \leq Z \leq 100$ by pure numerical methods. Here we present analytic calculations in the nonrelativistic limit and compare them with corresponding numerical results. For performing the summations over intermediate states (i.e. over the complete set of solutions of the Schrödinger equation describing electrons in the Coulomb field of the nucleus) we employ the Coulomb Green function [16]. The Green function method has been first applied for deriving the general expression for the two-photon decay probability in H atom and H-like ions in references [17,18]. An alternative approach applicable for arbitrary states based on Schwinger's analytical representation of the Coulomb Green function has been presented in [19,20].

One motivation for calculating the E1M1 two-photon decay is provided by the fact that in He-like ions this decay channel dominates in the absence of hyperfine-quenching effects as it has been first stated in reference [21]. Accordingly, the selection rules resulting from the angular-momentum coupling for $0 \rightarrow 0$ transitions allow for the emission of two photons with equal angular momenta only. The probability for the two-photon E1M1-decay process $2^3P_0 \rightarrow \gamma(E1)+\gamma(M1)+1^1S_0$ has been evaluated within a fully relativistic approach for $Z = 92$ in [22], for $50 < Z < 94$ in [23] and for $30 < Z < 100$ in [24]. As it has been indicated in [22], the nonrelativistic behavior of E1M1 transitions as a function of Z with the neglect of the interelectron interaction should be $W^{\mathrm{E1M1}} \sim (8/9\pi)(\alpha Z)^{12}/100$.

[a] e-mail: plunien@physik.tu-dresden.de
[b] Deseased

This implies that the same order of magnitude can be expected for the E1M1-decay probability for the process $2p_{1/2} \to \gamma(E1) + \gamma(M1) + 1s$ in H-like ions.

According to [22], this very small value arises due to the cancellation of contributions of the leading terms $2p_{1/2}$ and $2p_{3/2}$ in the summation over intermediate np-states. However, we should note that this result yields only a minor contribution to the total two-photon E1M1 $2p$–$1s$ decay rate for small nuclear charges Z when it is evaluated within the "velocity" gauge [22]. In this case a major contribution arises from the negative-energy intermediate states and scales like $\sim (\alpha Z)^8$ [14,15].

For the $2p_{1/2} \to 1s$ transition in hydrogen and hydrogen-like ions Schmieder's rule [19] does not apply and the two-photon transition $2p_{1/2} \to \gamma(E1)+\gamma(E2)+1s$ is allowed. As to our knowledge this transition probability has been evaluated for the first time in references [13,14]. It turned out that the major contribution is proportional to $(\alpha Z)^8$. Thus the two-photon E1M1 and E1E2 decay rates for $2p$–$1s$ transitions represent higher-order corrections to the life time of the $2p_{1/2}$-level when compared to the lowest-order $(\alpha Z)^3 \log(\alpha Z)$ radiative corrections derived in references [21,22]. A direct observation of the influence of these corrections in the H atom does not look feasible due to the huge background arising from the one-photon transition $2p \to \gamma(E1) + 1s$. However, two-photon decays of the $2p$-level could be observed in coincidence experiments.

In references [25,26] the parity-violation effect in the H atom has been described. Accordingly, the two-photon E1M1 transition $1s+2\gamma \to 2p_{1/2}$ plays the role of the basic transition while the parity-violating E1E1-transition becomes admixed by the parity-nonconserving (PNC) electroweak interaction. It was assumed that a coincidence experiment should be performed in oder to avoid the huge background from the one-photon process $1s + \gamma \to 2p_{1/2}$. Recent experimental and theoretical investigations of the PNC effects in neutral atoms indicate a possible disagreement with the predictions of the Standard Model [27]. At the earlier stage of the theoretical calculations of this effect the radiative corrections were fully disregarded and the agreement with the Standard Model predictions seemed to exist. However, as it was found in the work by Johnson, Bednyakov and Soff [28], the vacuum polarization correction violates this agreement. This result triggered a series of works (see [27]) where the electron self-energy correction was evaluated within different approximations. This correction compensated the vacuum polarization contribution. The most accurate result for the self-energy was recently obtained in [29]. Still, there is a number of radiative corrections to the PNC effects which are formally of the same order as the calculated corrections [30,31]. Before all these corrections are included, it is too early to make a conclusive statement about the agreement of the Cs experiments with the Standard Model predictions. The evaluation of radiative corrections to the PNC effects in neutral many-electron atoms presents a rather delicate problem. Therefore, experiments with simpler systems such as H-like ions are highly desirable. Our calculations of E1M1 and E1E2 transitions can be used directly for the evaluation of the "degree" of the PNC effect in experiments, similar to [25,26]. The knowledge of E1M1 and E1E2 transition rates in the hydrogen atom is also necessary for providing a more accurate estimate of the nonresonant correction to the two-photon resonance $1s$–$2s$ [32].

In this paper at first we repeated the calculation of the E1E1 decay of the $2s$-state in hydrogen. The result obtained is in satisfactory agreement with that of previous calculations and serves as a control for the further E1E2, E1M1 calculations and serves as control for the further E1E2, E1M1 calculations. The other parts of this paper are devoted to explore analytical evaluation methods of E1M1 and E1E2 contributions to the two-photon emission processes $2p_{1/2} \to 2\gamma + 1s$. The calculations are performed within different gauges and employing different forms for the expression of the transition probability that can be derived in the nonrelativistic limit (see [33]). Atomic units (a.u.) are used throughout the paper.

2 Transition probabilities in different forms and gauges

The transition probability for the emission of a photon with definite angular momentum and parity can be described in first-order of QED-perturbation theory within arbitrary gauges as

$$W_{A\to A'}(\omega) = \sum_{kq} \left[\left| \langle A'| \left(\vec{\alpha} \cdot {}_{\mathrm{e}}\vec{A}_{\omega kq}(\vec{r}) \right) + \Phi_{\omega kq}(\vec{r}) \,|A\rangle \right|^2 + \left| \langle A'| \vec{\alpha} \cdot {}_{\mathrm{m}}\vec{A}_{\omega kq}(\vec{r}) \,|A\rangle \right|^2 \right], \quad (1)$$

where ${}_{\mathrm{e}}\vec{A}_{\omega kq}$ and ${}_{\mathrm{m}}\vec{A}_{\omega kq}$ denote the spherical components of the transverse electric and transverse magnetic vector potential and $\Phi_{\omega kq}$ those of the scalar potential. The brakets $|A\rangle$ and $\langle A'|$ are stationary Dirac states (wave functions) with energies E_A and $E_{A'}$, and $\vec{\alpha}$ is the vector of the Dirac matrices. In the momentum representation these potentials take the form

$${}_{\mathrm{e}}\vec{A}_{\omega kq}(\vec{æ}) = \frac{4\pi^2 c^{3/2}}{\omega^{3/2}} \left(\delta\left(|\vec{æ}| - \frac{\omega}{c} \right) {}_{\mathrm{e}}\vec{Y}_{kq} + K\vec{n}Y_q^{(k)} \right), \quad (2)$$

$${}_{\mathrm{m}}\vec{A}_{\omega kq}(\vec{æ}) = \frac{4\pi^2 c^{3/2}}{\omega^{3/2}} \delta\left(|\vec{æ}| - \frac{\omega}{c} \right) {}_{\mathrm{m}}\vec{Y}_{kq}, \quad (3)$$

$$\Phi_{\omega kq}(|\vec{æ}|) = \frac{4\pi^2 c^{3/2}}{\omega^{3/2}} \delta\left(|\vec{æ}| - \frac{\omega}{c} \right) K Y_q^{(k)}. \quad (4)$$

Here $\vec{æ}$ denotes the photon momentum (with direction $\vec{n} = \vec{æ}/|æ|$), which is related to the frequency $\omega = E_A - E_{A'}$ of the emitted photon via $|\vec{æ}| = \omega/c$. ${}_{\mathrm{e}}\vec{Y}_{kq}$ and ${}_{\mathrm{m}}\vec{Y}_{kq}$ denote the vector spherical harmonics of electric and magnetic type, respectively, $Y_q^{(k)}$ is an ordinary spherical harmonic, c is the speed of light, K is the gauge constant and finally kq denote the angular momentum and projections of the photon, respectively.

The spherical components of the transversal electric ${}_{\rm e}A^{(1\lambda)}$ and the longitudinal ${}_{\rm l}A^{(1\lambda)}$ parts of the electromagnetic vector potential (the superscript (1λ) denotes the rank of a spherical tensor and labels the components) are

$$ {}_{\rm e}A^{(1\lambda)}_{\omega kq} = \sqrt{\frac{\omega}{\pi c(2k+1)}}\left[\sqrt{k(2k+3)}\begin{pmatrix}1 & k+1 & k\\ \lambda & q-\lambda & q\end{pmatrix} g_{k+1}(\omega r)C^{(k+1)}_{-q+\lambda} + \sqrt{(k+1)(2k-1)}\begin{pmatrix}1 & k-1 & k\\ \lambda & q-\lambda & q\end{pmatrix} g_{k-1}(\omega r)C^{(k-1)}_{-q+\lambda}\right] \times i^{-k-1}(-1)^{k+q-\lambda}, \quad (5) $$

$$ {}_{\rm l}A^{(1\lambda)}_{\omega kq} = \sqrt{\frac{\omega}{\pi c(2k+1)}}\left[\sqrt{(k+1)(2k+3)}\begin{pmatrix}1 & k+1 & k\\ \lambda & q-\lambda & q\end{pmatrix} \times g_{k+1}(\omega r)C^{(k+1)}_{-q+\lambda} + \sqrt{k(2k-1)}\begin{pmatrix}1 & k-1 & k\\ \lambda & q-\lambda & q\end{pmatrix} g_{k-1}(\omega r)C^{(k-1)}_{-q+\lambda}\right] \times i^{-k-1}(-1)^{k+q+\lambda+1}, \quad (6) $$

where $C^{(k)}_{-q} = \sqrt{4\pi/(2k+1)}Y^{(k)}_{-q}$ and usual notations for 3j-symbols are employed. The spherical components of the transverse magnetic vector potential read

$$ {}_{\rm m}A^{(1\lambda)}_{\omega qk} = (-1)^{\lambda+k+q} i^{-k}\sqrt{\frac{\omega(2k+1)}{\pi c}} g_k(\omega r) \times \begin{pmatrix}1 & k & k\\ -\lambda & -q+\lambda & -q\end{pmatrix} C^{(k)}_{-q+\lambda}, \quad (7) $$

while the spherical components of the scalar potential are given by

$$ \Phi_{\omega kq} = i^{-k}(-1)^{k+q}2\sqrt{\frac{\omega}{c}}g_k(\omega r)Y^{(k)}_{-q}. \quad (8) $$

The radial functions $g_k(\omega r)$ are related to Bessel functions J_μ via $g_k(z) = (2\pi)^{3/2}(1/\sqrt{z})J_{k+\frac{1}{2}}(z)$.

Usually two gauges are frequently used: the so-called Coulomb gauge is characterized by the choice for the gauge parameter $K = 0$ and by vanishing longitudinal part of the vector potential and scalar potential (i.e. $\vec{\nabla}\cdot\vec{A} = \vec{\nabla}\cdot{}_{\rm e}\vec{A} = 0$ and $\Phi = 0$), respectively. Another convenient choice is characterized by the parameter $K = -\sqrt{(k+1)/k}$. Within this gauge, as it can be seen from equations (5) and (6), the terms containing spherical functions $C^{(k)}_{-q}$ of the order $k-1$, vanish in the expression for the transition probability (1).

After some manipulations the expression for the emission probability of an electric photon with the angular momentum k can be cast into the form [27]

$$ W^{{\rm E}k}_{A\to A'} = \frac{2(k+1)\omega}{k(2k+1)c} \times \sum_{q=-k}^{k}\left|\langle A'|\left[{}_{\rm e}O'^{(k)}_{-q} + K\sqrt{\frac{k}{k+1}}\left({}_{\rm l}O^{(k)}_{-q} + {}_{\Phi}O^{(k)}_{-q}\right)\right]|A\rangle\right|^2 \quad (9) $$

Here

$$ {}_{\rm e}O'^{(k)}_{-q} = -i\left[k\sqrt{\frac{2k+3}{k+1}}g_{k+1}(\omega r)\left[C^{(k+1)}\times\alpha^{(1)}\right]^{(k)}_{-q} + \sqrt{k(2k-1)}g_{k-1}(\omega r)\left[C^{(k-1)}\times\alpha^{(1)}\right]^{(k)}_{-q}\right], \quad (10) $$

$$ {}_{\rm l}O^{(k)}_{-q} = i\left[\sqrt{(k+1)(2k+3)}g_{k+1}(\omega r)\left[C^{(k+1)}\times\alpha^{(1)}\right]^{(k)}_{-q} - \sqrt{k(2k-1)}g_{k-1}(\omega r)\left[C^{(k-1)}\times\alpha^{(1)}\right]^{(k)}_{-q}\right], \quad (11) $$

$$ {}_{\Phi}O^{(k)}_{-q} = \sqrt{2k+1}g_k(\omega r)C^{(k)}_{-q} \quad (12) $$

and $[a^{(s_1)}\times b^{(s_2)}]^{(s)}_q$ represents the tensor product of two irreducible tensors of rang s_1 and s_2 coupled to a tensor of rang s with components q.

Using the following integral relation for the Dirac wave functions [34]

$$ i\int \psi^*_{A'}\left(\vec{\alpha}\cdot\vec{\nabla}\chi\right)\psi_A\, d^3\tau = \frac{\omega}{c}\int \psi^*_{A'}\chi\psi_A\, d^3\tau, \quad (13) $$

where χ is an arbitrary function, one can establish another form for the Ek-transition probability (see [33])

$$ W^{{\rm E}k}_{A\to A'} = \frac{2(k+1)\omega^3}{k(2k+1)c^3}\sum_{q=-k}^{k}\left|\langle A'|{}_{\rm e}O^{(k)}_{-q} + K\frac{c}{\omega}\sqrt{\frac{k}{k+1}}\left[{}_{\rm l}O^{(k)}_{-q} + {}_{\Phi}O^{(k)}_{-q}\right]|A\rangle\right|^2, \quad (14) $$

where

$$ {}_{\rm e}O^{(k)}_{-q} = -rg_k(\omega r)C^{(k)}_{-q} - i\frac{r}{k+1}g_k(\omega r)\left[\sqrt{k(2k-1)}\left[C^{(k-1)}\times\alpha^{(1)}\right]^{(k)}_{-q} + \sqrt{(k+1)(2k+3)}\left[C^{(k-1)}\times\alpha^{(1)}\right]^{(k)}_{-q}\right]. \quad (15) $$

Thus, we have two different (equivalent) forms for the Ek-transition probabilities (Eqs. (9) and (14)) together with an arbitrary choice for the gauge constant K at our disposal. Analogous expressions for the emission probability of photons, characterized by energy and polarization, are provided in [35].

The aim of the present investigation concerns the derivation of the nonrelativistic limit of the E1M1-, E1E1- and E1E2-transition probabilities in different gauges and forms. Deriving the nonrelativistic limit of equations (9) and (14), implies two distinct nonrelativistic forms for the one-photon transition probability with arbitrary gauge constant K (see [36]):

$$W^{\mathrm{E}k}_{A\to A'} = \frac{2(k+1)(2k+1)\omega^{2k-1}}{k[(2k+1)!!]^2 c^{2k+1}} \times \sum_{q=-k}^{k} \left| (A'| \left(Q'^{(k)}_{-q} + K\sqrt{\frac{k}{k+1}} \left[Q'^{(k)}_{-q} - \omega Q^{(k)}_{-q} \right] \right) |A) \right|^2, \quad (16)$$

and

$$W^{\mathrm{E}k}_{A\to A'} = \frac{2(k+1)(2k+1)}{k[(2k+1)!!]^2} \sum_{q=-k}^{k} \left(\frac{\omega}{c}\right)^{2k+1} \times \left| (A'| \left(Q^{(k)}_{-q} + K\sqrt{\frac{k}{k+1}} \left[\frac{1}{\omega} Q'^{(k)}_{-q} - Q^{(k)}_{-q} \right] \right) |A) \right|^2. \quad (17)$$

Here $|A)$ and $(A'|$ are now nonrelativistic Schrödinger states (wave functions) together with operators

$$Q^{(k)}_{-q} = -r^k C^{(k)}_{-q}, \quad (18)$$

$$Q'^{(k)}_{-q} = -r^{-k-1} \left(k C^{(k)}_{-q} \frac{\partial}{\partial r} + \frac{i}{r}\sqrt{k(k+1)} \left[C^{(k)} \times L^{(1)} \right]^{(k)}_{-q} \right), \quad (19)$$

where $L^{(1)}$ is the orbital angular momentum of the atomic electron. Choosing $K = 0$, we find that the operator in equation (16) corresponds to the nonrelativistic transition operator in the "velocity" form, while for $K = -\sqrt{(k+1)/k}$ it is related to the transition operator in the "length" form. However, the correspondence of a certain gauge choice to a particular type of nonrelativistic transition operators is not unique. In view of equation (17) we can conclude, that within the nonrelativistic limit the expression (14) with $K = 0$ converts the transition operator into the "length" form and with $K = -\sqrt{(k+1)/k}$ into the "velocity" form, respectively.

3 Application of the Coulomb Green function

In order to calculate the transition probabilities for the processes $2p \to 2\gamma + 1s$ and $2s \to 2\gamma + 1s$ in the hydrogen atom we employed the nonrelativistic Coulomb Green function. The summations over the entire spectrum of the Schrödinger equation arise usually when perturbation theory is applied. The Green function approach allows us to express intermediate summations in a closed analytic form. This is very useful for the analysis and for tests of numerical evaluations.

The Green function of the Schrödinger equation is represented by the solution of the equation

$$\left(\hat{H} - E\right) G(E;\vec{r},\vec{r}\,') = \delta(\vec{r} - \vec{r}\,') \quad (20)$$

and can be always represented as a spectral decomposition

$$G(E;\vec{r},\vec{r}\,') = \sum_N \frac{\varphi^*_N(\vec{r})\varphi_N(\vec{r}\,')}{E_N - E}, \quad (21)$$

where the sum runs over the entire spectrum of the Schrödinger Hamiltonian (bound and continuous spectrum). The set of quantum numbers N may be specified as usual by the principal quantum number n, orbital-angular momentum number l and projections m. In view of the spherical symmetry it is sufficient to derive a closed expression for radial part $g_l(E;r,r')$ of the Green function, defined by the partial wave decomposition

$$G(E;\vec{r},\vec{r}\,') = \sum_{lm} \frac{1}{rr'} g_l(E;r;r')\, Y^*_{lm}(\Omega) Y_{lm}(\Omega'). \quad (22)$$

In the particular case of a Coulomb potential the Green function of equation (20) is called Coulomb Green Function (CGF). With the use of the expansion (22) the radial integrals occurring in equations (16) and (17) for the transition probabilities can be calculated explicitly (see Ref. [37] for details).

For the radial part of the Coulomb Green function it is convenient to employ the Sturmian expansion [38], which is defined in the entire complex energy plane via

$$\frac{1}{rr'} g_l(E;r,r') = \sum_{n_r=0}^{\infty} \frac{\Phi_{n_r l}(r)\Phi_{n_r l}(r')}{E_{n_r l} - E}, \quad (23)$$

where $\Phi_{n_r l}(r)$ denote the Sturmian functions. The Sturmian expansion of the CGF can be represented in an alternative form by introducing radial functions

$$R_{n_r l}\left(\frac{2r}{\nu}\right) = \frac{1}{r}\sqrt{\frac{Z}{\nu n_r}} \frac{1}{(2l+1)!} \times \sqrt{\frac{\Gamma(n_r+l+1)}{\Gamma(n_r-l)}} M_{n_r, l+\frac{1}{2}}\left(\frac{2r}{\nu}\right), \quad (24)$$

which are related to Whittaker functions $M_{n_r,l+\frac{1}{2}}(2r/\nu)$, where $\nu = Z/\sqrt{-2E}$. For integer values $\nu = n$ these functions coincide with the normalized, radial hydrogenic wave functions

$$\Phi_{n_r l}(r) = \sqrt{\frac{\nu n}{Z}} R_{nl}\left(\frac{2r}{\nu}\right). \quad (25)$$

Substitution of equation (25) into (23) yields

$$\frac{1}{rr'} g_l(\nu;r,r') = \frac{\nu^2}{Z^2} \sum_{n=l+1}^{\infty} \frac{n}{n-\nu} R_{nl}\left(\frac{2r}{\nu}\right) R_{nl}\left(\frac{2r'}{\nu}\right). \quad (26)$$

Within this paper we apply the Green function method for the evaluation of the two-photon decay probability in the hydrogen atom.

In [33] the two-photon transition process $2s \to \gamma(\mathrm{E1}) + \gamma(\mathrm{E1}) + 1s$ has been considered. The probability for a two-photon decay $A \to \gamma(\mathrm{E1}) + \gamma(\mathrm{E1}) + A'$ with photon frequencies ω_1 and ω_2 within the nonrelativistic limit and dipole approximation yields

$$dW^{\mathrm{E1E1}}_{A\to A'}(\omega_2) = \frac{8}{9\pi}\left(\frac{4\pi}{3}\right)^3 \times \sum_{M_1M_2}\left|(A'|rY_{1M_2}\left(\frac{\vec{r}}{r}\right)G(E_A-\omega_1;\vec{r},\vec{r}\,')r'Y^*_{1M_1}\left(\frac{\vec{r}\,'}{r'}\right)|A) + (A'|rY_{1M_1}\left(\frac{\vec{r}}{r}\right)G(E_A-\omega_2;\vec{r},\vec{r}\,')r'Y^*_{1M_2}\left(\frac{\vec{r}\,'}{r'}\right)|A)\right|^2 \times (\omega_1\omega_2)^2\,d\omega_2. \quad (27)$$

The energy conservation law implies $\omega_1 = E_A - E_{A'} - \omega_2$. After the evaluation of angular matrix elements in equation (27) the remaining radial integrals have the form

$$\int_0^\infty\int_0^\infty\int_0^\infty dr'drdx\,(r')^{s'+\frac{7}{2}}(r)^{s+\frac{7}{2}} \times \exp\left(-\frac{1}{\nu}(\beta'r'+\beta r)+(r+r')\cosh(x)\right) \times \left(\coth\left(\frac{x}{2}\right)\right)^{2\nu} I_{2l+1}\left(\frac{2\sqrt{rr'}}{\nu}\sinh(x)\right), \quad (28)$$

where n and n' are the principal quantum numbers of the initial and final states, respectively together with the parameters $\beta = \nu/n$, $\beta' = \nu/n'$ and $\nu = \sqrt{-2\,(E_{nl}-\omega)}$. These integrals can be evaluated analytically when inserting the series expansion for the modified Bessel functions I_{2l+1}. The integration over x should be evaluated in the end. Assuming emission of two identical E1 photons, one can write the total probability for such a two-photon decay as

$$W^{\mathrm{2E1}}_{A\to A'} = \frac{1}{2}\int_0^{\omega_0} dW^{\mathrm{2E1}}_{A\to A'}(\omega) \quad (29)$$

with $\omega_0 = E_A - E_{A'}$. For the process $2s \to 2\gamma(\mathrm{E1}) + 1s$ the result of the evaluations in [39] was reported as

$$W^{\mathrm{2E1}}_{2s\to 1s} = 8.226(\alpha Z)^6\,\mathrm{s}^{-1} \quad (30)$$

with an accuracy of about 1%.

In our calculation of the E1E1-transition probability we shall employ an alternative expression for the radial part of the CGF based on the Sturmian expansion (26).

4 E1E1 two-photon decay

In order to provide confidence in applications of the CGF expansions we consider at first the two-photon decay process $2s \to \gamma(E1) + \gamma(E1) + 1s$ in the hydrogen atom. As a test of the method the gauge constant $K = -\sqrt{(k+1)/k}$ is chosen in the expression equation (16) for the transition probability, which corresponds to the nonrelativistic "length" form as mentioned above. (This would be equivalent to the choice $K = 0$ together with the form Eq. (17).) Inspection of equations (16) and (18) reveals that the emission of electric photons (Ek) is described by the potentials

$$V^{\mathrm{E}k} = \frac{2(k+1)\omega^{k+\frac{1}{2}}}{k(2k+1)!!}r^kY^{(k)}_{-q}. \quad (31)$$

Accordingly, the two electric photon decay rate of the atomic state A can be written as

$$dW^{EkEk'}_{A\to A'} = \sum_{qq'm_Am_{A'}}\left|\sum_N\frac{(A'|V^{\mathrm{E}k}|N)(N|V^{\mathrm{E}k'}|A)}{E_N-E_A+\omega} + \sum_N\frac{(A'|V^{\mathrm{E}k'}|N)(N|V^{\mathrm{E}k}|A)}{E_N-E_A+\omega}\right|^2 \times \delta\left(\omega+\omega'-E_A+E_{A'}\right)d\omega d\omega' \quad (32)$$

Here the labels A, A' and N abbreviate the set of nonrelativistic quantum numbers (principal quantum number n, orbital momentum l and projection m_l) for characterizing the atomic electron in the initial (A), intermediate (N) and final (A') states. The photons will be characterized by the angular momentum and projections (kq) as well as by the type of the photon (electric or magnetic). Equation (32) also implies the summation over degenerate substates of the final atomic state A' and the average over the degenerate substates of the initial atomic state A as well as summations over the angular momentum projections of both emitted photons. The frequencies of the two photons ω and ω' are related by the energy conservation law $\omega' = \omega_0 - \omega$, where $\omega_0 = E_A - E_{A'}$.

Employing the eigenmode decomposition of the Coulomb Green function (Eqs. (22–26)) the probability of the two-photon decay process takes the form

$$dW^{\mathrm{E1E1}}_{A\to A'} = \frac{2\pi}{2l_A+1}\sum_{qq'm_Am_{A'}}\left|\sum_{lm_l}\iint d^3r_1d^3r_2\,R_{n_{A'}l_{A'}}(r_1)Y^*_{l_{A'}m_{A'}}\left(\frac{\vec{r_1}}{r_1}\right)V^{\mathrm{E1}}(\vec{r_1})g_l(\nu;r_1,r_2) \times Y_{lm_l}\left(\frac{\vec{r_1}}{r_1}\right)Y^*_{lm_l}\left(\frac{\vec{r_2}}{r_2}\right)V^{\mathrm{E1}}(\vec{r_2})R_{n_Al_A}(r_2)Y_{l_Am_A}\left(\frac{\vec{r_2}}{r_2}\right) + \sum_{lm_l}\iint d^3r_1d^3r_2\,R_{n_{A'}l_{A'}}(r_1)Y^*_{l_{A'}m_{A'}}\left(\frac{\vec{r_1}}{r_1}\right)V^{\mathrm{E1}}(\vec{r_1})g_l(\nu';r_1,r_2)Y_{lm_l}\left(\frac{\vec{r_1}}{r_1}\right)Y^*_{lm_l}\left(\frac{\vec{r_2}}{r_2}\right)V^{\mathrm{E1}}(\vec{r_2})R_{n_Al_A}(r_2)Y_{l_Am_A}\left(\frac{\vec{r_2}}{r_2}\right)\right|^2 d\omega, \quad (33)$$

where $V^{\mathrm{E}k}(\vec{r})$ is the potential (31) specified within the gauge $K = -\sqrt{(k+1)/k}$ and compatible with the form (16), together with parameters $\nu = Z/\sqrt{-2(E_A - \omega)}$, $\nu' = Z/\sqrt{-2(E_A - \omega')}$ and frequencies $\omega' = E_A - E_{A'} - \omega$. For the case of hydrogen one sets $Z = 1$.

Inserting the expression (31) for the potentials $V^{\mathrm{E}k}$ and evaluating at first the angular matrix elements in equation (33) (integrating over angles and summing over all projections) yields for quantum numbers $A = 2s$, $A' = 1s$, $k = k' = 1$ (the values for k and k' follow via selection rules for given initial and final atomic states)

$$dW^{2\mathrm{E}1}_{2s\to 1s}(\omega) = \left(\frac{2}{3}\right)^3 \frac{\omega^3\omega'^3}{\pi}\left[I_1(\nu) + I_1(\nu')\right]^2 \alpha^6 d\omega \quad (34)$$

together with a radial integrals I_1 of the type

$$I_1(\nu) = \frac{1}{\sqrt{2}}\int_0^\infty\int_0^\infty dr_1 dr_2 e^{-r_1-\frac{r_2}{2}} r_1^3 r_2^3 (2-r_2) g_1(\nu; r_1, r_2), \quad (35)$$

and

$$I_1(\nu') = \frac{1}{\sqrt{2}}\int_0^\infty\int_0^\infty dr_1 dr_2 e^{-r_1-\frac{r_2}{2}} r_1^3 r_2^3 (2-r_2) g_1(\nu'; r_1, r_2), \quad (36)$$

respectively. The further calculations utilize the representation of the radial Coulomb Green function in terms an expansion over Laguerre polynomials [32] (employed also in Ref. [34])

$$g_l(\nu; r, r') = \frac{4Z}{\nu}\left(\frac{4}{\nu^2} r r'\right)^l \exp\left(-\frac{r+r'}{\nu}\right) \times \sum_{n=0}^{\infty} \frac{n! L_n^{2l+1}\left(\frac{2r}{\nu}\right) L_n^{2l+1}\left(\frac{2r'}{\nu}\right)}{(2l+1+n)!(n+l+1-\nu)}. \quad (37)$$

The series (37) converges absolutely as $n^{-3/2}$ for arguments $r, r' > 0$ and $\Im(\nu) = 0$ [32]. The angular momentum quantum number $l = 1$ for intermediate states is determined by the angular integration. Inserting the expansion (37) for $l = 1$ into equation (35) (similarly for Eq. (36)) yields

$$I_1(\nu) = \frac{16\sqrt{2}}{\nu^3}\left(\frac{\nu}{2}\right)^{10} \times \sum_{m=0}^{\infty} \frac{m!}{(m+3)!(m+2-\nu)} \int_0^\infty d\xi \xi^4 e^{-\xi\left(\frac{\nu+1}{2}\right)} L_m^3(\xi) \times \int_0^\infty dt t^4 e^{-t\left(\frac{\nu+2}{4}\right)}\left(1 - \frac{\nu}{4}t\right) L_m^3(t). \quad (38)$$

These integrals can be analytically evaluated

$$I_1(\nu) = \frac{\nu^7 2^{13}\sqrt{2}}{(\nu+1)^5(\nu+2)^5} \times \sum_{m=0}^{\infty} \frac{(m+3)!}{m!(m+2-\nu)}\, {}_2F_1\left(-m, 5; 4; \frac{2}{\nu+1}\right) \times \left[{}_2F_1\left(-m, 5; 4; \frac{4}{4+\nu}\right) - \frac{5\nu}{\nu+2}\, {}_2F_1\left(-m, 6; 4; \frac{4}{2+\nu}\right)\right], \quad (39)$$

where ${}_2F_1(a, b; c; z)$ is a hypergeometric function. Inserting this result into equation (34) the integration over ω has to be performed in order to obtain the total transition probability for the E1E1 decay of the $2s$-state in the hydrogen atom. This is numerically achieved with the aid of the computer-algebra code MAPLE. The final result is

$$W^{2\mathrm{E}1}_{2s\to 1s} = \frac{1}{2}\int_0^{\omega_0} dW^{2\mathrm{E}1}_{2s\to 1s}(\omega) = 0.0013187\,(\alpha Z)^6\,\mathrm{a.u.} = 8.234\,\mathrm{s}^{-1}\,(Z=1) \quad (40)$$

with $\omega_0 = E_{2s} - E_{1s}$. In equation (40) we indicated the Z-dependence of the W^{E1E1}_{2s-1s} transition probability. The numerical value $8.234\,\mathrm{s}^{-1}$ coincides with earlier nonrelativistic results. The relative deviation from the result $8.228\,\mathrm{s}^{-1}$ reported in [3] is about 0.07%.

5 E1E2 transition probability for the 2p state

In this section we consider the E1E2 decay of the $2p$-state in hydrogen the atom. We employ again the set of quantum numbers nlm as far as the total angular momentum j is not important in this calculation performed within the nonrelativistic approach. Nevertheless, in order to compare our results with those obtained from the relativistic evaluation (see Refs. [14,15]), we shall perform the calculation within two different gauges according to equations (16–19). This will also elucidate possible influence of relativistic effects associated with the contribution of the negative-energy Dirac spectrum.

Specifying again equation (33) for the transition between levels $A = 2p$, $A' = 1s$ and taking into account that in this case the angular momenta of the photon can take values $k = 1, 2$, we receive four different terms contributing in equation (33). After angular integration and summation over projections we derive

$$dW^{\mathrm{E1E2}}_{2p\to 1s}(\omega) = \frac{2\omega^3\omega'^3}{3^2 5^2 \pi} \times \left|\omega' I_1(\nu) + \omega I_2(\nu') + \omega I_1(\nu') + \omega' I_2(\nu)\right|^2 d\omega, \quad (41)$$

where

$$I_1(\nu) = \frac{1}{\sqrt{6}}\int_0^\infty\int_0^\infty dr_1 dr_2\, r_1^3 r_2^5\, e^{-r_1-\frac{r_2}{2}}\, g_1(\nu; r_1, r_2) \quad (42)$$

and

$$I_2(\nu) = \frac{1}{\sqrt{6}} \int_0^\infty \int_0^\infty dr_1 dr_2\, r_1^4 r_2^4\, e^{-r_1 - \frac{r_2}{2}}\, g_2(\nu; r_1, r_2), \quad (43)$$

respectively. Inserting again the representation (37) for the CGF with $l = 1$ leads to radial integrals that can be analytically evaluated

$$I_1(\nu) = \frac{5\sqrt{6}\nu^9}{(\nu+1)^5(\nu+2)^7} \sum_{m=0}^{\infty} \frac{(m+3)!}{m!(m+2-\nu)} \times {}_2F_1\left(-m, 5; 4; \frac{2}{\nu+1}\right) {}_2F_1\left(-m, 7; 4; \frac{4}{\nu+2}\right) \quad (44)$$

and

$$I_2(\nu) = \frac{6^{3/2} 2^{12} \nu^9}{(\nu+1)^7(\nu+2)^7} \sum_{s=0}^{\infty} \frac{(s+5)!}{s!(s+3-\nu)} \times {}_2F_1\left(-s, 7; 6; \frac{2}{\nu+1}\right) {}_2F_1\left(-s, 7; 6; \frac{4}{\nu+2}\right). \quad (45)$$

Substituting the integrals (44) and (45) (with corresponding arguments ν and ν') into (41) and integrating over frequencies ω yields

$$W^{\rm E1E2}_{2p\to 1s} = \frac{1}{2} \int_0^{\omega_0} dW^{\rm E1E2}_{2p\to 1s} = 2.0075 \times 10^{-5}\, (\alpha Z)^8 \text{ a.u.} = 6.673 \times 10^{-6}\, \text{s}^{-1}\, (Z = 1) \quad (46)$$

with $\omega_0 = E_{2p} - E_{1s}$. In equation (46) we indicated the Z-dependence of the W^{E1E1}_{2p-1s} transition probability. Compared with the relativistic result in the "length" gauge (see [10]) the relative discrepancy is about 1%.

The calculation of the E1E2 two-photon decay with the nonrelativistic "velocity" form is more involved. Now the gauge constant should be chosen either $K = -\sqrt{(k+1)/k}$ for the form equation (17) or $K = 0$ for the form equation (16).

We choose $K = 0$ together with the form equation (16). The potential reads in this case

$$V^{\text{E}k}(\vec{r}\,) = \frac{4\omega^{k-\frac{1}{2}}}{(2k+1)!!} \sqrt{\frac{k+1}{k(2k+1)}}\, r^{k-1} \times \left[k Y^{(k)}_{-q}(\Omega) \frac{\partial}{\partial r} + \frac{i}{r} \sqrt{k(k+1)} \left[Y^{(k)} \times L^{(1)} \right]^k_{-q} \right]. \quad (47)$$

The formula for $dW^{\rm E1E2}_{2p\to 1s}$ follows again from equation (33). Performing angular integrations and summations over projections as discussed in previous cases now yields

$$dW^{\rm E1E2}_{2p\to 1s}(\omega) = \frac{2^4}{3^3 5^2 \pi} \omega' \omega \times \left[\omega'^2 \left| I_1(\nu) + I_2(\nu') \right|^2 + \omega^2 \left| I_1(\nu') + I_2(\nu) \right|^2 \right] d\omega \quad (48)$$

with radial integrals of the type

$$I_1(\nu) = \frac{1}{\sqrt{6}} \int_0^\infty \int_0^\infty dr_1 dr_2\, r_1^2 r_2^3\, e^{-r_1 - \frac{r_2}{2}} \times \left[1 - \frac{9i}{2} - \frac{r_2}{2} \right] \left[\frac{\partial}{\partial r_1} - \frac{2i}{r_1} \right] g_1(\nu; r_1, r_2), \quad (49)$$

and

$$I_2(\nu) = \frac{1}{\sqrt{6}} \int_0^\infty \int_0^\infty dr_1 dr_2\, r_1^3 r_2^2\, e^{-r_1 - \frac{r_2}{2}} \times \left[1 - 5i - \frac{r_2}{2} \right] \left[\frac{\partial}{\partial r_1} - \frac{3i}{r_1} \right] g_2(\nu; r_1, r_2) \quad (50)$$

together with parameters $\nu = Z/\sqrt{-2(E_{2p} - \omega)}$ and $\nu' = Z/\sqrt{-2(E_{2p} - \omega')}$, respectively. The integrations over r_1 and r_2 lead to a rather lengthy analytical formula containing various combinations of expressions similar to those in equations (44) and (45). The numerical evaluation yields finally

$$W^{\rm E1E2}_{2p\to 1s} = \frac{1}{2} \int_0^{\omega_0} dW^{\rm E1E2}_{2p\to 1s}(\omega) = 3.6896 \times 10^{-6}\, (\alpha Z)^8 \text{a.u.} \simeq 1.227 \times 10^{-6}\, \text{s}^{-1}\, (Z = 1). \quad (51)$$

where $\omega_0 = 3/8$ in a.u. This result differs from that obtained from relativistic calculations [10,11] by about 0.5%. Note, that unlike as in the case of the "length" form, the negative-energy contribution is no longer negligible when the "velocity" form is employed. Therefore, the result (Eq. (51)) does not coincide with equation (46) and represents only the positive-energy contribution to W^{E1E2}_{2p-1s} in the "velocity" form. Correspondingly, we compare this result to the positive-energy contribution calculated in [14,15]. The negative-energy contribution to W^{E1E2}_{2p-1s} in the "velocity" form for low Z values was evaluated analytically in [14].

6 E1M1 two-photon decay

For the mixed E1M1 two-photon transition probability the expression (32) should be replaced by

$$dW^{\rm E1M1}_{A\to A'} = \sum_{M_e M_m M_A m_{A'}} \left| \sum_N \frac{(A'|V^{\rm E1}(\omega)|N)(N|V^{\rm M1}(\omega')|A)}{E_N - E_A + \omega} + \frac{(A'|V^{\rm M1}(\omega')|N)(N|V^{\rm E1}(\omega)|A)}{E_N - E_A + \omega'} + \frac{(A'|V^{\rm E1}(\omega')|N)(N|V^{\rm M1}(\omega)|A)}{E_N - E_A + \omega'} + \frac{(A'|V^{\rm M1}(\omega)|N)(N|V^{\rm E1}(\omega')|A)}{E_N - E_A + \omega} \right|^2 d\omega. \quad (52)$$

Here $V^{\mathrm{E1}}(\omega) = (4/3)\omega^{3/2} r Y^{(1)}_{M_\mathrm{e}}$, $V^{\mathrm{M1}}(\omega) = \sqrt{4/3}\mu_0\omega^{3/2}(\hat{j}_{1M_\mathrm{m}} + \hat{s}_{1M_\mathrm{m}})$, $\mu_0 = \alpha/2$ is Bohr's magneton, $\hat{j}_{1M_m}$ and $\hat{s}_{1M_m}$ are the spherical components of the total angular-momentum and the spin operator (spherical tensors of rank 1) of the electron. This choice corresponds to the nonrelativistic "length" form for describing the emission electric photons. Since the potential for the magnetic photon includes total angular momentum and spin operator, coupled wave functions with the set of quantum numbers $N = \{nlsjm\}$ should be used, i.e.,

$$\phi_{nlsjm} = \sum_{m_l m_s} C^{jm}_{lm_l \ \ sm_s} R_{nl}(r) Y^{(l)}_{m_l}(\Omega)\eta_{sm_s}, \qquad (53)$$

where $C^{jm}_{lm_l \ \ sm_s}$ is a Clebsch-Gordan coefficient, $R_{nl}(r)$ is the solution of the radial Schrödinger equation and η_{sm_s} is the spin function. The magnetic potentials occurring in equation (54) do not depend on radial variables. Thus, only the intermediate state with $nl = n_A l_A$ will contribute in equation (52). After performing angular integrations and summations over all projections one arrives at the expressions

$$dW^{\mathrm{E1M1}}_{2p\to 1s} = \frac{2^8\mu_0^2}{\pi}\left(\frac{2}{3}\right)^{12}\omega\omega'^3 d\omega \qquad (54)$$

and

$$W^{\mathrm{E1M1}}_{2p\to 1s} = \frac{1}{2}\int_0^{3/8} dW^{\mathrm{E1M1}}_{2p\to 1s} = \frac{2^5}{\pi}\left(\frac{2}{3}\right)^{12}\alpha^8\int_0^{3/8}\omega\left(\frac{3}{8}-\omega\right)^3 d\omega. \qquad (55)$$

As the final result we obtain

$$W^{\mathrm{E1M1}}_{2p\to 1s} = \frac{2^5}{\pi}\left(\frac{2}{3}\right)^{12}\frac{243}{655360}(\alpha Z)^8 \text{ a.u.} = 9.6769\times 10^{-6}\ \mathrm{s}^{-1} \quad (Z=1). \qquad (56)$$

Here we again indicated the Z-dependence of the W^{E1M1}_{2p-1s} transition probability. Comparison with the result of a fully relativistic calculation reveals a discrepancy of about 0.1%.

7 Conclusions

The results of the analytic calculations presented in this paper confirm the results of the purely numerical calculations of $E1E2$, $E1M1$ two-photon transitions in H atom reported in references [14,15]. This support is especially important since no other numerical calculations for these transitions are available up to now. The evaluations are performed by means of the CGF method. All angular and radial integrations are analytically evaluated and only the final integration over the photon frequency has to be performed numerically. Looking at the pure dependence on parameters the results reveal that in the nonrelativistic limit the $E1E2$ and $E1M1$ transitions are by a factor $(\alpha Z)^2$ smaller, than the one for $E1E1$ transition. Still the additional numerical smallness provides two more orders of magnitude.

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