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Application of B-splines finite basis sets to relativistic two-photon decay rates of 2s level in hydrogenic ions

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Abstract. A theoretical study of the one- and two-photon spontaneous emission rates from the $2s_{1/2}$ state of one-electron ions is presented. High-precision values of the relativistic emission rates for ions with nuclear charge Z up to 100 are obtained through the use of finite basis sets for the Dirac equation constructed from B-splines. Furthermore, we analyze the influence of the inclusion of quantum electrodynamics corrections in the initial and final state energies.

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1 Introduction

One of the most widely studied atomic transition in which selection rules forbid the emission of one electric dipole photon is the radiative decay of the metastable $2s_{1/2}$ state to the $1s_{1/2}$ ground state of H-like systems. This metastable state can decay by either one of following two competing processes: the emission of a single photon, or the emission of two photons (which proceeds via intermediate sates). In the former case, the process

$$2s_{1/2} \to 1s_{1/2} + h\nu \tag{1.1}$$

which is highly inhibited by the angular momentum and parity selection rules, can only proceed trough relativistic corrections to the magnetic dipole ($M1$) matrix element [1–3] and is therefore slower than an ordinary allowed electric dipole ($E1$) decay rate by a factor of Z^6, where Z is the atomic number of the hydrogenic system.

In the two photons decay mode,

$$2s_{1/2} \to 1s_{1/2} + h\nu_1 + h\nu_2 \tag{1.2}$$

the two photons are emitted with a continuous distribution due to conservation of energy, such that

$$h\nu_1 + h\nu_2 = E_{2s_{1/2}} - E_{1s_{1/2}}\,. \tag{1.3}$$

There are an endless number of possibilities for this process, such as $2M1$, $2E2$, $2M2$, $2E3$, .., $E1M1$, $E2M1$, $E2M3$, ..., where $2On$ means a transition which involves two photons On, and $OnO'm$ means a transition that involves one photon On and one photon $O'm$. In this nomenclature E and M stands, respectively, for a electric type and magnetic type photon, and the adjacent numbers indicate the angular momentum quantum number, ℓ, of the photon. The $2E1$ transition is the most probable of all the combinations.

Early interest in the decay of metastable states of hydrogen and helium stemed from astrophysics. Under the low-density conditions that prevail in planetary nebulae, for example, the simultaneous emission of two photons by an H atom in the metastable $2s_{1/2}$ level is considered as a source of continuum [4]. About 32 per cent of electron captures lead directly to the $2s_{1/2}$ level and subsequently to two-photon emission, since collisional deexcitation proves unimportant. A similar mechanism is verified for the $1s2s\ ^1S_0$ state of helium, which depopulates primarily by two-photon emission, while the $1s2s\ ^3S_1$ state depopulates at approximately equal rates by collisions and by radiation [5]. Consequently, the ratio of line intensities in the helium triplet spectrum can be used as a density probe for the nebula [6,7]. The spectra of heliumlike ions in the solar corona and solar flare provide similar information [5] that can lead to the determination of electron densities in thermal cosmic ray x-ray sources [5,8].

More recently, attention has been payed to two-photon transitions in high-Z ions, *e.g.*, U^{90+}, in the context of the study of parity-violation effects in atoms [9].

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In many-electron systems two-photon transitions are normally overwhelmed by allowed electric dipole single photon emission and Auger transitions. However, some two-photon experiments have been performed in such systems [10–14].

This work is the first step in obtaining a general, many-electron code in the framework of the multiconfiguration Dirac-Fock method (MCDF) [15–19] where we will use B-splines basis sets to compute high-precision value of two-photon spontaneous emission rates for many-electron systems.

2 Historical overview

2.1 Theory

The earliest theoretical work on two-photon processes is due to Goppert-Mayer [20], who, after studying spontaneous and induced two-photon emission on the basis of Dirac's theory of dispersion, proposed that the simultaneous emission of two photons in hydrogen was the dominant decay mode for the $2s_{1/2}$ state.

This conclusion was confirmed by Breit and Teller [21]. They applied Goppert-Mayer's theory and estimated both $M1$ and $2E1$ $2s \to 1s$ transition rates in atomic hydrogen and found that two-photon emission is the most probable radiative decay mode, and is therefore the principal cause of the mechanism of interstellar $2s$ hydrogen atoms. They also found upper and lower limits for the decay rate, $1/\tau$, corresponding to this mode of decay: $6.5 < 1/\tau < 8.7$ s^{-1}.

Later, more detailed nonrelativistic calculations of the $2E1$ $2s \to 1s$ rate in hydrogen were carried out by Spitzer and Greenstein [4] and by Shapiro and Breit [5], which involved term-by-term numerical evaluation of the infinite summation over intermediate states in the second-order matrix elements responsible for the decay. The former found 8.227 s^{-1} for hydrogen and the latter found $(8.226 \pm 0.001)Z^6$ s^{-1} for the decay of a hydrogenic ion of nuclear charge Z.

Klarsfeld [22], obtained an analytic expression for the two-photon transition matrix element in terms of hypergeometric functions and performed a highly accurate nonrelativistic calculation for the $2E1$ decay rate for hydrogenic ions $2s \to 1s$ reaching the result $(8.2283 \pm 0.0001)Z^6$ s^{-1}. This result, when corrected for revised values of the fundamental constants, becomes 8.22938 Z^6 s^{-1}, according to Drake [23].

In the framework of nonrelativistic dipole approximation other calculations were performed, for instance, by Tung *et al.* [24] (who got the result 8.2284 s^{-1} for the hydrogenic $2s \to 1s$ transition) and by Costescu *et al.* [25]. They introduced improvements in the mathematical formalism by expressing the finite summations over intermediate states in terms of hypergeometric functions, but did not improve significantly upon Klarsfeld's result. Using the nonrelativistic approach as well, Bassani [26] and Quattropani [27] made calculations for the two-photon excitations from the ground state of atomic hydrogen to ns excited states.

More recently, Drake [23] used a finite basis-set method to calculate the nonrelativistic $2E1$ $2s \to 1s$ rate in hydrogen and obtained the high-precision value of 8.2293810 s^{-1}.

Florescu [28] developed a theory of two-photon transitions in hydrogenlike systems including the transitions from higher shells. According to her results, the $3d \to 1s$ double-photon transition should be 5 times stronger than $2s \to 1s$ double-photon transition.

The first relativistic calculations of the decay rates was done by Johnson [29]. By avoiding any expansion in (αZ) and replacing the usual summations over intermediate states by a numerical evaluation of the relativistic Green's function for the electron, Johnson performed a relativistic numerical calculation for the $2E1$ $2s \to 1s$ rate in the hydrogenic isoelectronic sequence and obtained results correct to lowest orders in α for a large number of $Z < 92$. He also calculated $M1$ decay rates in a closed analytical form, and the total decay rate for the $2s \to 1s$ transition in hydrogenic ions for the same set of Z values.

Goldman and Drake [30] carried out a calculation of the two-photon decay rate for $2s_{1/2}$ hydrogenic ions. They included relativistic effects and some combinations of photon multipoles that make significant contributions in the numerical evaluation of the results for a set of Z values. These authors summed over intermediate states by constructing a finite-basis-set representation of the Dirac Green's function. These calculations have yielded results substantially different from the previous direct numerical calculations of Johnson [29]. Goldman and Drake have pointed out that these differences arise because higher-order terms in the expansion of the photon multipole potential were omitted in Johnson's evaluation.

In a later work, Parpia and Johnson [31] used a direct numerical Green's function technique to treat the multipole potential and obtained results that agree closely with those of Goldman and Drake [30], confirming their hypothesis. After Parpia and Johnson, the small remaining discrepancies between their results and Goldman and Drake ones could be attributed to the difference in the methods used to treat the hydrogenic Green's function.

Goldman [32] introduced a variational method based on the orthogonality properties of the Laguerre polynomials, in which all the matrix elements of the Dirac Hamiltonian are calculated in closed form. This method was applied to a calculation of the two-photon rates for hydrogenic ions and resolved the discrepancy between the previous calculation of Parpia and Johnson and that by Goldman and Drake in favor of the latter.

Barut and Salamin [33], describing the level width as the imaginary part of the self-energy, calculated the relativistic rates of the $M1$ $2s \to 1s$ transition in the hydrogen isoelectronic sequence. These authors compared their results with those of Johnson [29] and of Parpia and Johnson [31] and found an almost complete agreement with the former ones.

Recently, using a representation of the Dirac Coulomb Green function in terms of an expansion over a Sturmian basis, Szymanowski [34] computed two-photon probability amplitudes for transitions in hydrogenic atoms from the ground state towards states in the L and M shells.

2.2 Experiments

Precision lifetime measurements can readily test the theory of atomic structure by providing experimental results that are sensitive to both the wave functions and energies of given configurations.

Although the basic theory of two-photon emission has been available since the beginning of the thirties with the work of Goppert-Mayer [20], laboratory experimentation were not performed before 1965. Lipeles, Novick and Tolk [35] by the application of atomic beam techniques pioneered the study of the $2s \to 1s$ transition in ionized helium. Their basic method was subsequently enhanced by Marrus and Schmieder [36] who studied a number of heavy hydrogenic and heliumlike ions, using the newly developed beam-foil technology.

Since that time, the increased availability of highly charged heavy ions from accelerator facilities around the world have enabled many experiments, among which the ones by Kocher, Clendenin and Novick [37] (H-like He), Marrus and Schmieder [36] (H-like and He-like Argon), Prior [38] (H-like He), Cocke *et al.* [39] (H-like F and O), Hinds, Clendenin and Novick [40] (H-like He), Gould and Marrus [41] (H-like Ar), Marrus *et al.* [42] (He-like Kr), Dunford *et al.* [43] (H-like and He-like Ni), Dunford *et al.* [44] (He-like Br) and Simionovici *et al.* [45] (He-like Nb).

All these experiments have been limited to the study of two-photon transitions from the metastable $2s$ level for which there is no competing strongly allowed one-photon transition. This is because two-photon emission is typically 4−8 orders of magnitude weaker than strongly allowed one-photon emission. Indeed, the importance of the atomic beam technique pioneered by Lipeles, Novick and Tolk [35] is that it yields a beam of nearly pure metastable atoms downstream of the region in which all strongly allowed one-photon transitions take place. Short-pulse excitation would, in some circumstances, offer a similar advantage.

Of course, two-photon transitions from nonmetastable states are also of intrinsic interest, as is the study of such transitions between the inner shells of many-electron atoms. For such systems beam techniques appear to offer no particular advantage since they cannot be used to eliminate strong one-photon emission. For these systems, only ingenious state-of-the-art experimental techniques have made it possible recently to detect two-photon transitions in many-electron systems, in which the process is overwhelmed by the allowed single-photon emission. Bannett and Freund [10,11], followed by Ilakovac *et al.* [12] and Mu and Crasemann [13,14], have been able to report measurements of two-photon x-ray emission from one-electron inner-shell transitions, *i.e.*, from a transition other than $2s \to 1s$.

Contrary to the large number of experiments in the study of two-photon $2s \to 1s$ transition, as far as our knowledge goes, few precise measurements of the single-photon $M1$ decay rates have yet been reported. Besides the reference to their effect, without measurement, made by Gould and Marrus [41], there are the measurements of the $M1$ $2s \to 1s$ decay amplitude in Ag^{46+} and in Kr^{35+}, made by Simionovici *et al.* [45] and by Cheng *et al.* [46], respectively.

3 Theory of relativistic radiative transitions

3.1 One-photon transitions

In the theory of quantum electrodynamics, the interaction potential between the electromagnetic field and the electro-positron field depends on the choice of gauge [47]. Therefore, the expression for the electric multipole transition matrix becomes gauge dependent. The spontaneous emission rate for a one-photon transition $i \to f$, is [48], in atomic units,

$$w_{i\to f} = 2\alpha\omega \frac{[j_f]}{[L]} \begin{pmatrix} j_i & L & j_f \\ 1/2 & 0 & -1/2 \end{pmatrix}^2 \left|\overline{M}_{fi}\right|^2, \tag{3.1}$$

where $\overline{M}_{fi}$ involves only radial integrals. The notation $[j,k,\ldots]$ means $(2j+1)(2k+1)\ldots$ For magnetic type multipoles

$$\overline{M}^{m}_{fi} = \overline{M}^{(0,L)}_{fi} = \frac{2L+1}{[L(L+1)]^{1/2}} \left(\kappa_f + \kappa_i\right) I^{+}_{L} \tag{3.2}$$

whereas for electric type multipoles, the value depends linearly on the gauge parameter

$$\overline{M}^{e}_{fi}(G) = \overline{M}^{(1,L)}_{fi} + G\overline{M}^{(-1,L)}_{fi}, \tag{3.3}$$

where

$$\begin{aligned} \overline{M}^{(1,L)}_{fi} &= \left(\frac{L}{L+1}\right)^{1/2} \left[(\kappa_f - \kappa_i) I^{+}_{L+1} + (L+1) I^{-}_{L+1} \right] \\ &\quad - \left(\frac{L+1}{L}\right)^{1/2} \left[(\kappa_f - \kappa_i) I^{+}_{L-1} - L I^{-}_{L-1} \right] \end{aligned} \tag{3.4}$$

$$\begin{aligned} \overline{M}^{(-1,L)}_{fi} &= (2L+1) J_L \\ &\quad + (\kappa_f - \kappa_i)\left(I^{+}_{L+1} + I^{+}_{L-1}\right) \\ &\quad - L I^{-}_{L-1} + (L+1) I^{-}_{L+1}. \end{aligned} \tag{3.5}$$

In the notation used by Rosner and Bhalla [49], the $I^{\pm}_{L}(\omega)$ and $J_L(\omega)$ integrals are defined as follows:

$$I^{\pm}_{L}(\omega) = \int_0^\infty \left(P_f Q_i \pm Q_f P_i\right) j_L\left(\frac{\omega r}{c}\right) dr, \tag{3.6}$$

and

$$J_L(\omega) = \int_0^\infty \left(P_f P_i + Q_f Q_i\right) j_L\left(\frac{\omega r}{c}\right) dr, \tag{3.7}$$

where P and Q are the large and small components of the radial Dirac wave function respectively. The photon frequency $(E_i - E_f)$ is denoted by ω; j_i, κ_i, and E_i are respectively, the total angular momentum, relativistic number, and energy of the initial state. The corresponding quantities for the final state are j_f, κ_f, and E_f.

Grant [48] discussed the implications of this dependence of the gauge parameter G on the transition rate of by considering the nonrelativistic limits for electric dipole ($E1$) transitions. Among the great variety of possible gauges, he showed that there are two values of G which are of particular utility because they lead to well known nonrelativistic operators. First, if $G = 0$, one has the so called Coulomb gauge, or velocity gauge, which leads to the dipole velocity form in the nonrelativistic limit. Second, if $G = [(L+1)/L]^{1/2}$, or more specifically $G = \sqrt{2}$ for $E1$ transitions ($L = 1$), one obtains nonrelativistically an expression which reduces to the dipole length form of the transition operator.

3.2 Two-photon transitions

The theory of two-photon transitions is given in detail in reference [30]. We repeat here only the relevant points. The basic expression for the differential emission rate is, in atomic units, [47]

$$\frac{dw}{d\omega_1} = \frac{\omega_1\omega_2}{(2\pi)^3c^2}\left|\sum_n \frac{\langle f|\,\widetilde{A}_2^*\,|n\rangle\,\langle n|\,\widetilde{A}_1^*\,|i\rangle}{E_n - E_i + \omega_1} + \frac{\langle f|\,\widetilde{A}_1^*\,|n\rangle\,\langle n|\,\widetilde{A}_2^*\,|i\rangle}{E_n - E_i + \omega_2}\right|^2 d\Omega_1 d\Omega_2 \tag{3.8}$$

where i and f denote the initial and final states, ω_j is the frequency, and $\mathrm{d}\Omega_j$ the element of solid angle for the jth photon. The summation over n includes integrations over the continua for both positive and negative energy solutions for the Dirac equation. Conservation of the energy requires

$$E_i - E_f = \omega_1 + \omega_2, \tag{3.9}$$

which permits only one of the two photon frequencies to be independent.

Integrating the differential emission rate expression (3.8) we get the transition probability per unit time for a transition in which is emitted one photon $\Theta_1 L_1$ and one photon $\Theta_2 L_2$ [50],

$$\overline{W}_{\Theta_1 L_1,\Theta_2 L_2} = \int_0^{\omega_{if}} \frac{d\overline{W}_{\Theta_1 L_1,\Theta_2 L_2}}{d\omega_1}\, d\omega_1\,, \tag{3.10}$$

where

$$\Theta = E, M \qquad\qquad L = 1, 2, \ldots \tag{3.11}$$

The averaged decay rate $d\overline{W}/d\omega_1$ is given by the expression

$$\begin{aligned}\frac{d\overline{W}}{d\omega_1} = {} & \frac{\omega_1\omega_2}{(2\pi)^3c^2\,(2j_i+1)} \\ & \times \sum_{L_1,\lambda_1,L_2,\lambda_2,j}\Bigg\{[S^j(2,1)]^2 + [S^j(1,2)]^2 \\ & + 2\sum_{j'}(-1)^{2j'+L_1+L_2}[j,j']^{1/2}\begin{pmatrix} j_f & j' & L_1 \\ j_i & j & L_2\end{pmatrix} \\ & \times S^j(2,1)S^{j'}(1,2)\Bigg\},\end{aligned} \tag{3.12}$$

where

$$S^j(2,1) = \sum_{n_j}\frac{\overline{M}_{f,n_j}^{(\lambda_2,L_2)}(\omega_2)\,\overline{M}_{i,n_i}^{(\lambda_1,L1)}(\omega_1)}{E_{n_j} - E_i + \omega_1}\Delta^j(2,1), \tag{3.13}$$

and

$$\begin{aligned}\Delta^j(2,1) = {} & \frac{4\pi\,[j_i,j,j_f]^{1/2}}{[L_1,L_2]^{1/2}} \\ & \times \begin{pmatrix} j_f & L_2 & j \\ 1/2 & 0 & -1/2\end{pmatrix}\begin{pmatrix} j & L_1 & j_i \\ 1/2 & 0 & -1/2\end{pmatrix}.\end{aligned} \tag{3.14}$$

The matrix elements $\overline{M}_{f,i}^{(\lambda,L)}$ were defined above by the expressions (3.2), (3.4) and (3.5).

Finally, the spontaneous emission transition probability per unit time for a two-photon transition is given by

$$w = \sum_{\text{all }\Theta_1,L_1,\Theta_2,L_2} d_{\Theta_1 L_1,\Theta_2 L_2}\overline{W}_{\Theta_1 L_1,\Theta_2 L_2} \tag{3.15}$$

where

$$d_{\Theta_1 L_1,\Theta_2 L_2} = \begin{cases} 1 & \text{if } \Theta_1 L_1 \neq \Theta_2 L_2 \\ 1/2 & \text{if } \Theta_1 L_1 = \Theta_2 L_2. \end{cases} \tag{3.16}$$

The factor (1/2) is included to avoid counting each pair twice when both photons have the same characteristics.

4 Solution of the Dirac-Fock equation on a B-splines basis set

We will suppose that the atom, or ion, is enclosed in a finite box with a radius large enough to get a good approximation of the wavefunctions, with some suitable set of boundary conditions. In that case, one obtains a finite set, with the continuum described by discrete functions.

Let us denote by $\{\phi_n^i(r),\ i = 1,\ldots,2N\}$ such a set of solutions where n is the level number and i the position of the solution in the set. For each n the set $\{\phi_n^i(r),\ i = 1,\ldots,2N\}$ is complete. Each $\phi_n^i(r)$ obeys [51]

$$\begin{bmatrix} V_{DF}(r) & -\dfrac{d}{dr} + \dfrac{\kappa}{r} \\ \dfrac{d}{dr} + \dfrac{\kappa}{r} & -2mc + V_{DF}(r)\end{bmatrix}\phi_n^i(r) = \alpha\epsilon_n^i\phi_n^i(r). \tag{4.1}$$

In the relativistic case, a complete set spans both positive and negative energy solutions. Solutions $i = 1, \ldots, N$ represent the continuum $\epsilon_n^i < -2mc^2$ and solutions $i = N+1, \ldots, 2N$ represent bound states (the few first ones) and the $\epsilon_n^i > 0$ continuum using the conventions of equation (4.1). For practical reasons, such as easy numerical implementation, this set of solutions is itself expressed as linear combination of another basis set. We have choosen the B-splines basis set, for which integrations of numerical functions are easy to handle. The method we used and the derivation of the solutions of equation (4.1) in terms of B-splines are exactly identical to those described in detail by Johnson, Blundell and Saperstein in reference [52], and will not be reproduced here.

5 Quantum electrodynamics corrections to the initial and final States

In order to test the influence of the quantum electrodynamics (QED) corrections on the $2s$–$1s$ transition, we have performed calculations of the one-photon $M1$ decay rates $w(M1)$ and of the total two-photon decay rates $w(2\gamma)$ adding radiative corrections to the initial and final state energies.

Since we are studying hydrogenlike ions, the only QED corrections we have considered are the one-electron radiative corrections. For that case one has to evaluate a perturbation expansion in powers of the fine structure constant $\alpha \approx 1/137$, each power of α corresponding to one exchanged photon. For high Z the self-energy must be calculated to all orders in $(Z\alpha)$ which represent the strength of the electron-nucleus interaction, which cannot be treated perturbatively at high-Z. The one-electron corrections are of order $\alpha(Z\alpha)^4mc^2$. The one-electron corrections of order $\alpha(Z\alpha)^4mc^2$ are the self-energy and vacuum polarization.

The self-energy part contributions and the correspondent finite nuclear size corrections included in this work are those calculated by Mohr and collaborators [53–56] for several (n, ℓ), and by Mohr and Soff [57], respectively. The vacuum polarization part can be evaluated using well known potentials [58,59] and can, in contrast to the self-energy, be approximated by a few terms in the expansion in $Z\alpha$. Here we include the first two contributions scaling as $\alpha(Z\alpha)^4\ mc^2$ (Uehling potential [58]) and as $\alpha(Z\alpha)^6$ mc^2. All-order calculations have been performed and they show that the convergence in $Z\alpha$ is fast [60–62]. These calculations were calculated with the MCDF code developed by J.P. Desclaux, P. Indelicato and collaborators [15–19].

The second order one-electron radiative corrections have not been fully calculated. Still missing (for high-Z) is the two-loop self-energy (except for a piece of an irreducible diagram [63]), which can only be calculated by extrapolating recent calculations for low-Z [64]. This kind of extrapolation has been shown to be unreliable for the one loop self-energy [53]. For uranium ($Z = 92$) all other pieces (mixed self-energy vacuum polarization diagrams [64,65]) have been calculated recently. The two-loop vacuum polarization can be easily calculated by combining the Källén and Sabry contribution [66], with the difference between self-consistent and perturbative calculations of the Uehling potential.

Finally, and to be complete, we include a nuclear polarization correction [67–69], which in this case is small compared to the experimental precision and second-order QED corrections.

Table 1. Different calculations of the $2E1$ decay rates for the $2s_{1/2}$ state in hydrogenic ions in s^{-1}, for selected values of the nuclear charge Z.

Z	$Z^{-6}w(2E1)$ Goldman and Drake[a]	Parpia and Johnson[b]	Goldman[c]	This calculation
1	8.2291	8.2291	8.2290626	8.229063
20	8.1181	8.1196	9.1174035	8.117111
40	7.8096	7.8116	7.8092612	7.809031
60	7.3446	7.3453	7.3446482	7.344138
80	6.7440	6.7426	6.7428876	6.741590
92	6.3097	6.3093	6.3096623	6.309096

[a] S.P. Goldman, G.W.F. Drake, reference [30]
[b] F.A. Parpia, W.R. Johnson, reference [31]
[c] S.P. Goldman, reference [32]

6 Results and discussion

6.1 2s–1s decay rates

We apply now the B-splines basis set to the relativistic calculation of the one- and two-photon decay rates of the $2s_{1/2}$ state in hydrogenic ions, where we used finite nuclear size assuming a uniform nuclear charge distribution. The stability and accuracy of the results have been verified with respect to variations of both the gauge invariance and the basis set parameters: the size of the basis set, *i.e.*, the number of B-splines (ns), the degree of the B-splines (k) and the radius of the cavity (R). The parameters used in the calculation of the present results are: $k = 9$, $ns = 59$ and $R = 40$ a.u.

The integration with respect to the photon frequency has been performed using a Gauss-Legendre integration with 15 points. We believe that the present calculations are accurate to all figures quoted, since the variation relative to the the Gauss-Legendre integration points, k, and ns, assures a convergence of the results to at least one figure beyond those shown in the tables, while the agreement between length- and velocity-gauge values continues to at least six figures beyond those shown in the tables. Enough multipoles have been included in the calculation of the total 2-photon decay rates to obtain an accuracy of six significant digits for values of the nuclear charge from $Z = 1$ to $Z = 100$.

The results obtained for the $2E1$ contributions for the total decay rate of H-like ions for a set of nuclear charge values Z is presented in Table 1,

where we compare with other calculations. We note that the difference between the results is larger than the last digit quoted. In the case of the $Z = 20$ Goldman's

Table 2. Multipole combinations included in the present calculation of the total two-photon decay rates, with the allowed intermediate states in the summation of equation (3.13).

Multipoles	Intermediate states	Contribution (s^{-1}) $Z = 1$	$Z = 54$	$Z = 92$
$2E1$	$p_{1/2}, p_{3/2}$	8.229063	1.859221×10^{11}	3.825552×10^{12}
$E1M2$	$p_{3/2}$	2.537183×10^{-10}	4.927784×10^{7}	9.138566×10^{9}
$2M1$	$s_{1/2}, d_{3/2}$	1.380359×10^{-11}	3.402664×10^{6}	1.109270×10^{9}
$2E2$	$d_{3/2}, d_{5/2}$	4.907232×10^{-12}	9.817756×10^{5}	1.785817×10^{8}
$2M2$	$p_{3/2}, f_{5/2}$	3.069354×10^{-22}	5.502043×10^{2}	9.906518×10^{5}
$E2M3$	$d_{5/2}$	1.422935×10^{-22}	2.637946×10^{2}	4.949021×10^{5}
$E2M1$	$d_{3/2}$	1.637802×10^{-23}	4.416201×10^{1}	1.766901×10^{5}
$2E3$	$f_{5/2}, f_{7/2}$	5.526719×10^{-24}	9.721688×10^{0}	1.614230×10^{4}
$E3M2$	$f_{5/2}$	3.193886×10^{-34}	6.236436×10^{-3}	1.586533×10^{2}

Table 3. Contributions from different combinations of multipoles to the integrated decay rate w (s^{-1}). The Goldman's results are from reference [32] and the Goldman and Drake's results are from reference [30].

	$2E1$ $Z^{-6}w$	$E1M2$ $Z^{-10}10^{10}w$	$2M1$ $Z^{-10}10^{11}w$	$2E2$ $Z^{-10}10^{12}w$	
This calculation	8.2290626	2.5371825	1.3803590	4.9072324	
Goldman	8.22906	2.53718	1.38036	4.90723	
Goldman and Drake	8.2291	2.5371	1.3804	4.9072	

	$2M2$ $Z^{-14}10^{22}w$	$E2M3$ $Z^{-14}10^{22}w$	$E2M1$ $Z^{-14}10^{23}w$	$2E3$ $Z^{-14}10^{24}w$	$E3M2$ $Z^{-18}10^{34}w$
This calculation	3.0693540	1.4229345	1.6384276	5.5267188	3.1936424
Goldman	3.06935	1.42293	1.63936	5.52671	3.19817
Goldman and Drake	4.0890		5.7520		

Table 4. Frequency distribution of the two-photon decay rate. The spectral function $\psi(y, Z)$ is defined by equation (6.1) in the text.

$y \backslash Z$	1	20	40	60	80	92	92 (incl. QED)
0.0625	2.031203	1.943383	1.711577	1.413992	1.121326	0.972174	0.965492
0.1250	3.155900	3.083222	2.873568	2.560789	2.183861	1.952175	1.941688
0.1875	3.841947	3.783957	3.609879	3.334509	2.971781	2.729653	2.716475
0.2500	4.281391	4.235743	4.094683	3.863922	3.542724	3.317943	3.302706
0.3125	4.566370	4.530352	4.416121	4.224868	3.947630	3.747587	3.730800
0.3750	4.745181	4.716023	4.621301	4.459965	4.218603	4.040831	4.022954
0.4375	4.843859	4.818805	4.735867	4.592954	4.374439	4.211431	4.192909
0.5000	4.875429	4.851738	4.772716	4.635913	4.424934	4.266742	4.248013

Table 5. Decay rates of the $2s_{1/2}$ state (s^{-1}). Z is the nuclear charge.

Z	$w(M1)$	$w(2\gamma)$	$w(\text{tot})$
1	$2.49591901 \times 10^{-6}$	8.229063	8.229065
2	$2.55626238 \times 10^{-3}$	5.266042×10^{2}	5.266068×10^{2}
3	$1.47448885 \times 10^{-1}$	5.997292×10^{3}	5.997440×10^{3}
4	2.61939935	3.368840×10^{4}	3.369102×10^{4}
5	2.44075507×10^{1}	1.284703×10^{5}	1.284948×10^{5}
6	1.51219742×10^{2}	3.834621×10^{5}	3.836133×10^{5}
7	7.06964753×10^{2}	9.665073×10^{5}	9.672143×10^{5}
8	2.68960303×10^{3}	2.152429×10^{6}	2.155119×10^{6}
9	8.74246719×10^{3}	4.361002×10^{6}	4.369745×10^{6}
10	2.51003240×10^{4}	8.200570×10^{6}	8.225670×10^{6}
11	6.51818482×10^{4}	1.451723×10^{7}	1.458241×10^{7}
12	1.55805459×10^{5}	2.444948×10^{7}	2.460528×10^{7}
13	3.47395680×10^{5}	3.948826×10^{7}	3.983565×10^{7}
14	7.30032314×10^{5}	6.154224×10^{7}	6.227228×10^{7}
15	1.45779185×10^{6}	9.300828×10^{7}	9.446608×10^{7}
16	2.78454759×10^{6}	1.368469×10^{8}	1.396314×10^{8}
17	5.11524642×10^{6}	1.966626×10^{8}	2.017779×10^{8}
18	9.07769992×10^{6}	2.767891×10^{8}	2.858668×10^{8}
19	1.56211038×10^{7}	3.823790×10^{8}	3.980001×10^{8}
20	2.61488033×10^{7}	5.194978×10^{8}	5.456466×10^{8}
21	4.26945535×10^{7}	6.952229×10^{8}	7.379175×10^{8}
22	6.81531341×10^{7}	9.177457×10^{8}	9.858988×10^{8}
23	1.06578441×10^{8}	1.196478×10^{9}	1.303057×10^{9}
24	1.63563871×10^{8}	1.542164×10^{9}	1.705728×10^{9}
25	2.46724142×10^{8}	1.966992×10^{9}	2.213716×10^{9}
26	3.66298161×10^{8}	2.484712×10^{9}	2.851010×10^{9}
27	5.35897415×10^{8}	3.110759×10^{9}	3.646656×10^{9}
28	7.73430228×10^{8}	3.862374×10^{9}	4.635804×10^{9}
29	1.10222453×10^{9}	4.758729×10^{9}	5.860953×10^{9}
30	1.55241116×10^{9}	5.821062×10^{9}	7.373473×10^{9}
32	2.98171369×10^{9}	8.539707×10^{9}	1.152142×10^{10}
34	5.50996703×10^{9}	1.223449×10^{10}	1.774446×10^{10}
36	9.84022291×10^{9}	1.716321×10^{10}	2.700343×10^{10}
38	$1.70476667 \times 10^{10}$	2.362974×10^{10}	4.067741×10^{10}
40	$2.87414359 \times 10^{10}$	3.198853×10^{10}	6.072997×10^{10}
42	$4.72840541 \times 10^{10}$	4.264918×10^{10}	8.993323×10^{10}
45	$9.57795011 \times 10^{10}$	6.400027×10^{10}	1.597798×10^{11}
46	$1.19994177 \times 10^{11}$	7.281768×10^{10}	1.928119×10^{11}
47	$1.49641928 \times 10^{11}$	8.261063×10^{10}	2.322526×10^{11}
50	$2.82903863 \times 10^{11}$	1.186870×10^{11}	4.015908×10^{11}
54	$6.27019120 \times 10^{11}$	1.859758×10^{11}	8.129949×10^{11}
55	$7.58563275 \times 10^{11}$	2.069368×10^{11}	9.655000×10^{11}
58	$1.31864299 \times 10^{12}$	2.816997×10^{11}	1.600343×10^{12}
60	$1.87950004 \times 10^{12}$	3.428003×10^{11}	2.222300×10^{12}
62	$2.65095005 \times 10^{12}$	4.142831×10^{11}	3.065233×10^{12}
65	$4.36179752 \times 10^{12}$	5.438258×10^{11}	4.905623×10^{12}
66	$5.12606662 \times 10^{12}$	5.936543×10^{11}	5.719721×10^{12}
70	$9.58256840 \times 10^{12}$	8.313011×10^{11}	1.041387×10^{13}
74	$1.73921410 \times 10^{13}$	1.140289×10^{12}	1.853243×10^{13}
75	$2.01035177 \times 10^{13}$	1.230364×10^{12}	2.133388×10^{13}
78	$3.07616136 \times 10^{13}$	1.535228×10^{12}	3.229684×10^{13}
80	$4.05520829 \times 10^{13}$	1.769879×10^{12}	4.232196×10^{13}
82	$5.31771608 \times 10^{13}$	2.032135×10^{12}	5.520930×10^{13}
85	$7.91168010 \times 10^{13}$	2.482088×10^{12}	8.159889×10^{13}
86	$9.01056465 \times 10^{13}$	2.648282×10^{12}	9.275393×10^{13}
90	$1.49999200 \times 10^{14}$	3.401841×10^{12}	1.534010×10^{14}
92	$1.92403666 \times 10^{14}$	3.835980×10^{12}	1.962396×10^{14}
100	$5.03848601 \times 10^{14}$	6.008640×10^{12}	5.098572×10^{14}

Table 6. Comparison between theoretical and experimental total lifetime values (s) of the $2s_{1/2}$ state.

Ion	Experiment	This calculation
He^{+}	$(2.04^{+0.81}_{-0.34}) \times 10^{-3}$ [a]	1.898950×10^{-3}
	$(1.922 \pm 0.082) \times 10^{-3}$ [b]	
	$(1.923 \pm 0.002) \times 10^{-3}$ [c]	
O^{7+}	$(4.53 \pm 0.43) \times 10^{-7}$ [d]	4.640116×10^{-7}
F^{8+}	$(2.37 \pm 0.17) \times 10^{-7}$ [d]	2.288463×10^{-7}
S^{15+}	$(7.3 \pm 0.7) \times 10^{-9}$ [e]	7.161711×10^{-9}
Ar^{17+}	$(3.54 \pm 0.25) \times 10^{-9}$ [e]	3.498133×10^{-9}
	$(3.487 \pm 0.036) \times 10^{-9}$ [f]	
Ni^{27+}	$(2.171 \pm 0.018) \times 10^{-10}$ [g]	2.157123×10^{-10}
Ag^{46+}	$(4.49 \pm 0.08) \times 10^{-12}$ [h]	4.305658×10^{-12}

[a] C. A. Kocher, J.E. Clendenin, and R. Novick [37]
[b] M.H. Prior [38]
[c] E.A. Hinds, J. E. Clendenin, and R. Novick [40]
[d] C.L. Cocke *et al.* [39]
[e] R. Marrus and R.W. Schmieder [36]
[f] H. Gould and R. Marrus [41]
[g] R.W. Dunford *et al.* [43]
[h] A. Simionovici *et al.* [45]

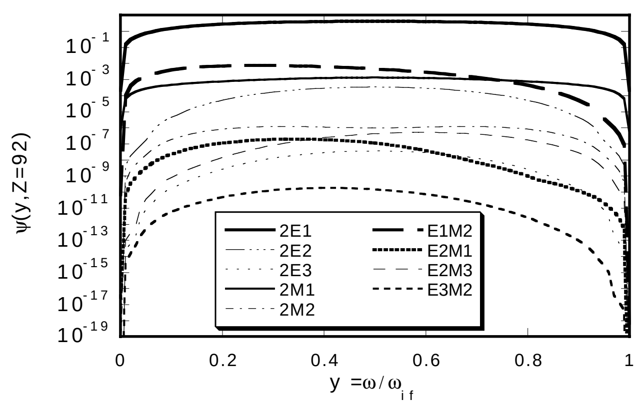

Fig. 1. Frequency distribution of each multipole contribution included in the calculation of the total two-photon decay rates for $Z = 92$.

value, which differs more than one unit, we believe that there was a misprint.

In Table 2 we list all the multipole combinations included in the calculation in order to reach the accuracy presented. We also show which intermediate states are to be included in each case in the summations in equation (4.1), as well as the values of each contribution for $Z = 1$, 54 and 92.

The breakdown of the integrated decay rate w into contributions from different combinations of multipoles is shown in Table 3, as well the contributions calculated by Goldman [32] and Goldman and Drake [30], where, except for the $2M2$ and $E2M3$ cases, we notice a fair agreement between the 3 sets of results. In the mentioned 2 cases Goldman and Drake's results differ more than one unit from the other 2 sets of results.

Table 7. The influence of the QED corrections on one-photon decay rates $w(M1)$, in s^{-1}, for the $2s_{1/2}$ state of H-like ions, for selected values ofthe nuclear charge Z.

Z	$w(M1)$	$w(M1)$ incl. QED corr.	Δ
1	$2.49591901 \times 10^{-6}$	$2.49455106 \times 10^{-6}$	0.05%
20	2.61488033×10^{7}	2.61394546×10^{7}	0.04%
40	$2.87414359 \times 10^{10}$	$2.87163893 \times 10^{10}$	0.09%
60	$1.87950004 \times 10^{12}$	$1.87675182 \times 10^{12}$	0.15%
80	$4.05520829 \times 10^{13}$	$4.04647736 \times 10^{13}$	0.22%
92	$1.92403666 \times 10^{14}$	$1.91896638 \times 10^{14}$	0.26%

Table 8. The influence of the QED corrections on two-photon decay rates $w(2\gamma)$, in s^{-1}, for the $2s_{1/2}$ state of H-like ions, for selected values of the nuclear charge Z.

	$w(2\gamma)$	$w(2\gamma)$ including QED corr.		
Z	Coulomb and length gauges	Coulomb gauge	length gauge	Δ
1	8.2290626143	8.2234343414	8.2234650306	0.07%
20	$5.1949783956 \times 10^{8}$	$5.1918037265 \times 10^{8}$	$5.1918056077 \times 10^{8}$	0.06%
40	$3.1988531806 \times 10^{10}$	$3.1940268092 \times 10^{10}$	$3.1940326673 \times 10^{10}$	0.15%
60	$3.4280025068 \times 10^{11}$	$3.4192756713 \times 10^{11}$	$3.4192930844 \times 10^{11}$	0.25%
80	$1.7698790233 \times 10^{12}$	$1.7631831699 \times 10^{12}$	$1.7632042151 \times 10^{12}$	0.38%
92	$3.8359800476 \times 10^{12}$	$3.8180133338 \times 10^{12}$	$3.8180892579 \times 10^{12}$	0.47%

To present the spectral distribution for a specific value of Z, it is convenient to express the results in the form suggested by Spitzer and Greenstein [4]

$$\frac{d\overline{W}}{dy} = Z^6 \left(\frac{9\alpha^6}{2^{10}}\right) \psi(y, Z) \quad Ry \tag{6.1}$$

where $y = \omega/\omega_{if}$ is the fraction of the photon energy carried by one of the two photons. Values of $\psi(y, Z)$ are given in Table 4 for a number of H-like ions. All the contributions listed in Table 2 are included in these results. In Figure 1 we show the spectral distribution of each contribution for $Z = 92$.

Finally, our final values for $2s$–$1s$ decay rates for a large selection of hydrogenic ions are presented in Table 5. In column 2 of this table we list the one-photon $M1$ decay rates $w(M1)$ and in the column 3 the total two-photon decay rates $w(2\gamma)$. In the final column we give the total decay rates for this transition $w(\text{tot})$, which is the sum of the $w(M1)$ contribution with the $w(2\gamma)$ contribution. All the values are in s^{-1}.

The total decay rates are compared with the available experimental data in Table 6. In all the cases, except in the result for He^{+} by Hinds, Clendenin and Novick, the theoretical value are within the experimental error bar. Moreover, all the measurements are in agreement with theory in such a way that the difference between the experimental and theoretical results are, for almost all cases, less than 2.5%. For Ar^{17+} and Ni^{27+}, the difference is less than 1%: 0.3% in the former and 0.6% in the later.

6.2 The influence of the QED corrections

In Table 7 we list the one-photon $M1$ decay rates for a set of Z values calculated with and without QED corrections. We observe that differences between the two results Δ go from 0.05% for $Z = 1$, to 0.26% for $Z = 92$, which confirms that the effects from QED corrections become more important for high values of Z.

The influence of QED corrections on two-photon decay rates is presented in Table 8. In this table we observe, besides the expected differences between the results with and without the mentioned corrections Δ, which go from 0.07% for $Z = 1$, to 0.47% for $Z = 92$, in the Coulomb gauge case, the breakdown of gauge invariance when we include radiative corrections. As it was stated above, the necessary and sufficient condition for the transition matrices for all multipoles to be gauge invariant is that the transition matrix of electric multipoles for longitudinal photons vanish identically. This condition is automatically satisfied for exact functions or for a single-particle model, but it may not necessary hold, for example, for a Dirac-Fock model due to the effect of the nonlocal potential, or for a model that, based in operators not included in the Hamiltonian, corrects the energy, which is the present case.

In the last column of Table 4 we also listed the frequency distribution of the two-photon decay rate for $Z = 92$ calculated with QED corrections.

7 Conclusions

In this article we have demonstrated, by the results of Section 6, that the finite basis sets for the Dirac equation constructed from B-splines is a very powerful tool for relativistic variational calculations, namely when applied to calculation of one- and two-photon decay rates. Besides its ability to avoid numerical problems, a great advantage of this method, over the commonly used Slater method, for instance, is that the Hamiltonian matrix is sparse (all the matrix elements between nonoverlapping splines is zero) and therefore "well behaved". Generation of a B-splines basis for many-electron systems is also well known and has been widely used in many-body perturbation theory. By applying this method to the $2s \to 1s$ radiative transition, we have been able to calculate the one-photon $M1$ decay rates $w(M1)$, the total two-photon decay rates $w(2\gamma)$ and the total decay rates $w(\mathrm{tot})$ for a set of hydrogenlike ions with nuclear charge from $Z = 1$ to $Z = 100$. In the two-photon decay rates $w(2\gamma)$ were included the nine most important multipoles contributions. We have shown also the breakdown of the gauge invariance when QED corrections are included in the energy of the initial and final states.

The calculations presented in this work have been done using the computer facilities at the Centro de Física Atómica da Universidade de Lisboa and at the Laboratoire Kastler-Brossel. This research was supported in part by JNICT (Portugal) under project Praxis/2/2.1/FIS/7223/94. J.P.S. acknowledges support from the Embassy of France in Portugal and JNICT for his stay in the Laboratoire Kastler-Brossel and P.I. for his stay in the Centro de Física Atómica da Universidade de Lisboa.

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