Negative continuum effects on the two–photon decay rates of hydrogen–like ions

Andrey Surzhykov1,2, José Paulo Santos3,4, Pedro Amaro1,3,4 and Paul Indelicato5
1Physikalisches Institut, Universität Heidelberg, Philosophenweg 12, D–69120 Heidelberg, Germany
2GSI Helmholtzzentrum für Schwerionenforschung, Planckstr. 1, D–64291 Darmstadt, Germany
3Departamento de Física, Faculdade de Ciências e Tecnologia, FCT, Universidade Nova de Lisboa, 2829-516 Caparica, Portugal
4Centro de Física Atómica da Universidade de Lisboa, Av. Prof. Gama Pinto 2, 1649-003 Lisboa, Portugal
5Laboratoire Kastler Brossel, École Normale Supérieure, CNRS, Université P. et M. Curie – Paris 6, Case 74; 4, place Jussieu, 75252 Paris CEDEX 05, France
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Two–photon decay of hydrogen–like ions is studied within the framework of second–order perturbation theory, based on relativistic Dirac’s equation. Special attention is paid to the effects arising from the summation over the negative–energy (intermediate virtual) states that occurs in such a framework. In order to investigate the role of these states, detailed calculations have been carried out for the \(2s_{1/2} \rightarrow 1s_{1/2}\) and \(2p_{1/2} \rightarrow 1s_{1/2}\) transitions in neutral hydrogen \(\text{H}\) as well as for hydrogen–like xenon \(\text{Xe}^{54+}\) and uranium \(\text{U}^{91+}\) ions. We found that for a correct evaluation of the total and energy–differential decay rates, summation over the negative–energy part of Dirac’s spectrum should be properly taken into account both for high–\(Z\) and low–\(Z\) atomic systems.

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I. INTRODUCTION

Experimental and theoretical studies on the two–photon transitions in atomic systems have a long tradition. Following seminal works by Göppert–Mayer 1 and by Breit and Teller 2 a large number of investigations have been performed in the past which focused on the decay of metastable states of light neutral atoms and low–\(Z\) ions. These investigations have dealt not only with the total and energy–differential decay rates 3,4,5 but also with the angular distributions 6,7,8,9 and even polarization correlations between the two emitted photons 10,11,12. Detailed analysis of these two–photon properties have revealed unique information about electron densities in astrophysical plasmas and thermal X–ray sources, highly precise values of physical constants 13, structural properties of few–electron systems including subtle quantum electrodynamical (QED) effects 14 as well as about the basic concepts of quantum physics such as, e.g., non–locality and non–separability 15.

Beside the decay of metastable states of low–\(Z\) systems, much of today’s interest is focused also on the two–photon transitions in high–\(Z\) ions and atoms which provide a sensitive tool for improving our understanding of the electron–photon interactions in the presence of extremely strong electromagnetic fields 16. In such strong fields produced by heavy nuclei, relativistic and retardation effects become of paramount importance and may strongly affect the properties of two–photon emission. To explore these effects, therefore, theoretical investigations based on Dirac’s equation have been carried for the total and energy–differential decay rates 17,18,19,20,21 as well as for the angular and polarization correlations 22,23,24. In general, relativistic predictions for the two–photon total and differential properties have been found in a good agreement with experimental data obtained for the decay of inner–shell vacancies of heavy neutral atoms 25,26 and excited states of high–\(Z\) few–electron ions 27.

Although intensive experimental and theoretical efforts have been undertaken recently to understand relativistic effects on the two–photon transitions in heavy ions and atoms, a number of questions still remain open. One of the questions, which currently attracts much of interest, concerns the role of negative energy solutions of Dirac’s equation in relativistic two–photon calculations. Usually, these calculations are performed within the framework of the second–order perturbation theory and, hence, require summation over the (virtual) intermediate ion states. Such a summation, running over the complete spectrum, should obviously include not only positive– (discrete and continuum) but also negative–eigenenergy Dirac’s states. One might expect, however, that since the energy release in two–photon bound–bound transitions is less than the energy required for the electron–positron pair production, the contribution from the negative part of Dirac’s spectrum should be negligible even for the decay of heaviest elements. From practical viewpoint, this assumption justifies the restriction of the intermediate–state summation to the positive–energy solutions only. Exclusion of the negative continuum would lead, in turn, to a significant simplification of the the second–order relativistic calculations especially for many–electron systems for which the problem of (many particle) negative continuum still remains unsolved.

Despite the (relatively) small energy of two–photon transitions, the influence of Dirac’s negative continuum in second–order calculations should be further questioned because of possibility for production and subsequent annihilation of the virtual anti–particles. It has been ar-
gued, for example, that transitions involving positron states have to be taken into account for the proper description of Thomson scattering \[28\], interaction of ions with intense electromagnetic pulses \[29, 30\] in the “undercritical” regime as well as magnetic transitions in two–electron ions \[31, 32, 33\]. Moreover, the first step towards the analysis of negative–energy contributions to the two–photon properties has been done by Labzowsky and co–workers \[34\] who focused on E1M1 and E1E2

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dependence of the energy which is carried away by the “first” photon.

\[
dw{dω_1} = \frac{dω_1}{(2\pi)^3} \left| \sum_ν \left( \frac{⟨f | A^*_j j ν | ν | A^*_i i⟩}{E_ν - E_i + ω_1} \right) \right|^2 dΩ_1 dΩ_2, \tag{1}
\]

where the transition operators \(A^*_j\) with \(j = 1, 2\) describe the (relativistic) electron–photon interaction. For the emission of photons with wave vectors \(k_j\) and polarization vectors \(e_j\) these operators read as:

\[
A^*_j = α \cdot (\hat{e}_j + G k_j) e^{-i k_j r} - Ge^{-i k_j r}, \tag{2}
\]

where \(α\) is a vector of Dirac matrices and \(G\) is an arbitrary gauge parameter. In the calculations below, following Grant \[35\], we employ two different gauges that are known to lead to well known non–relativistic operators. First, we use the so–called Coulomb gauge, when \(G = 0\), which corresponds to the velocity form of electron–photon interaction operator in the non–relativistic limit. As the second choice we adopt \(G = \sqrt{(L + 1)/L}\) in order to obtain Babushkin gauge which reduces, for the particular case of \(L=1\), to the dipole length form of the transition operator.

In Eq. (1), \(i\) \(\equiv |n_f κ_f μ_f⟩\) and \(f\) \(\equiv |n_f κ_f μ_f⟩\) denote solutions of the Dirac’s equation for the initial and final ionic states while \(E_i \equiv E_{n_i κ_i}\) and \(E_f \equiv E_{n_f κ_f}\) are the corresponding one–particle energies. Because of energy conservation, \(E_i\) and \(E_f\) are related to the energies \(ω_{1,2}\) of the emitted photons by:

\[
E_i - E_f = ω_1 + ω_2. \tag{3}
\]

From this relation, it is convenient to define the so–called energy sharing parameter \(y = ω_1/(ω_1 + ω_2)\), i.e., the fraction of the energy which is carried away by the “first” photon.

As usual in atomic physics, the second–order transition amplitudes in Eq. (1) and, hence, the two–photon transitions rates can be further simplified by applying the techniques of Racah’s algebra if all the operators are presented in terms of spherical tensors and if the (standard) radial–angular representation of Dirac’s wavefunctions are employed. For the interaction of electron with electromagnetic field, the spherical tensor components are obtained from the multipole expansion of the operator \(A^*_j\) (see Refs. \[38, 39\] for further details). By using such an expansion, we are able to re–write Eq. (1)

\[
\frac{dω}{dω_1} = \sum_{θ_1 L_1 θ_2 L_2} \frac{dW_{θ_1 L_1 θ_2 L_2}}{dω_1}, \tag{4}
\]

as a sum of partial multipole rates

\[
\frac{dω}{dω_1} = \sum_{θ_1 L_1 θ_2 L_2} \frac{dW_{θ_1 L_1 θ_2 L_2}}{dω_1}, \tag{4}
\]
which describe the emission of two photons of electric ($\Theta_j = E$) or magnetic ($\Theta_j = M$) type carrying away the angular momenta $L_1$ and $L_2$. For the decay of unpolarized ionic state $|n_j\kappa_i\rangle$, in which the emission angles as well as polarization of both photons remain unobserved, these partial multipole rates are given by [18]:

$$\frac{dW_{\Theta_1L_1\Theta_2L_2}}{d\omega_1} = \frac{\omega_1\omega_2}{(2\pi)^3} \sum_{\lambda_{\Theta_1}\lambda_{\Theta_2}} \sum_{\nu} \left| S^{j_1}(1, 2) \right|^2$$

$$+ \left| S^{j_{\nu}}(2, 1) \right|^2 + 2 \sum_{\kappa_{\nu}} d(j_{\nu}, j_{\nu}') S^{j_{\nu}}(2, 1) S^{j_{\nu}'}(1, 2),$$

where the angular coefficient $d(j_{\nu}, j_{\nu}')$ is defined by the phase factor and 6j Wigner symbol:

$$d(j_{\nu}, j_{\nu}') = \sqrt{(2j_{\nu} + 1)(2j_{\nu}' + 1)}$$

$$\times (-1)^{2j_{\nu} + L_1 + L_2} \left\{ \begin{array}{ccc} j_f & j_i & L_1 \\ j_{\nu} & j_{\nu}' & L_2 \end{array} \right\},$$

and the radial integral part is expressed in terms of the reduced matrix elements of the multipole (electric and magnetic) field operators:

$$S^{j_{\nu}}(1, 2)$$

$$= \sum_{\nu} \left\langle n_f\kappa_f|a_{\lambda_{\Theta_1}^*} n_j\kappa_i \right\rangle \left\langle n_j\kappa_i |a_{\lambda_{\Theta_2}^*} n_{\nu}\kappa_{\nu} \right\rangle \left| S^{j_{\nu}} \right|_{10}^{10} = \left| S^{j_{\nu}} \right|_{10}^{10}.$$ (7)

The summation over $\lambda_{\Theta_j}$ in Eq. (5) is restricted to $\lambda_{\Theta_j} = \pm 1$ for the electric ($\Theta_j = E$) and $\lambda_{\Theta_j} = 0$ for the magnetic ($\Theta_j = M$) photon transitions.

Until now, we have discussed the general expressions for the two–photon transition rates which are differential in energy $\omega_1$ of one of the photons. By performing an integration over this energy one may easily obtain the total rate that is directly related to the lifetime of a particular excited state against the two–photon decay. As it follows from Eq. (4), such a total rate can be represented as a sum of its multipole components:

$$w_{\text{tot}} = \sum_{\Theta_1L_1\Theta_2L_2} W_{\Theta_1L_1\Theta_2L_2}$$

$$= \sum_{\Theta_1L_1\Theta_2L_2} \omega_i \int_0^{\omega_i} \frac{dW_{\Theta_1L_1\Theta_2L_2}}{d\omega_1} d\omega_1,$$ (8)

where $\omega_i = E_i - E_f$ is the transition energy.

As seen from Eqs. (4)–(8), any analysis of the differential as well as total two–photon decay rates can be traced back to the (reduced) matrix elements that describe the interaction of an electron with the (multipole) radiation field. Since the relativistic form of these matrix elements is applied very frequently in studying the various atomic processes, we shall not discuss here their evaluation and just refer the reader for all details to references [18,19,32]. Instead, in the next section we will focus on the summation over the intermediate states $|n_j\kappa_{\nu}\rangle$ which appears in the second–order transition amplitudes (see Eq. (7)).
the finite (discrete) basis solutions constructed from the B–spline sets.

Although the B–spline basis set approach has been discussed in detail elsewhere [19, 37, 38], here we briefly recall its main features. In this way, we shall consider the ion (or atom) under consideration to be enclosed in a finite cavity with a radius \(R\) large enough to get a good approximation of the wavefunctions with some suitable set of boundary conditions, which allows for discretization of the continua. Wavefunctions that describe the quantum states \(|\nu\rangle \equiv |n_\nu\kappa_\nu\rangle\) of such a “particle in box” system can be expanded in terms of basis set functions \(\phi_i^\nu(r)\) with \(i = 1, \ldots, 2N\) which, in turn, are found as solutions of the Dirac–Fock equation,

\[
\begin{bmatrix}
\frac{V(r)}{c} - \left(\frac{\partial}{\partial r} + \frac{\kappa_\nu}{R}\right) - 2c + \frac{V(r)}{c} \\
- \left(\frac{\partial}{\partial r} + \frac{\kappa_\nu}{R}\right) - 2c + \frac{V(r)}{c}
\end{bmatrix}
\phi_i^\nu(r) = \frac{\epsilon_i^\nu}{c} \phi_i^\nu(r) ,
\]

where \(\epsilon_i^\nu = E_i^\nu - mc^2\) and \(V(r)\) is a Coulomb potential of a uniformly charged finite–size nucleus. Due to computational reasons, each of \(\phi_i^\nu(r)\) function is expressed as a linear combination of B–splines as it was originally proposed in Ref. 37 by Johnson and co–workers.

For each quantum state \(|\nu\rangle\) the set of basis functions \(\phi_i^\nu(r)\) spans both positive and negative energy solutions. Solutions labeled by \(i = 1, \ldots, N\) describe the negative continuum with \(\epsilon_i^\nu < -2mc^2\) while solutions labeled by \(i = N + 1, \ldots, 2N\) correspond to the first few states of the bound–state spectrum as well as to positive continuum with \(\epsilon_i^\nu > 0\). Thus, by selecting the proper sub–set of basis functions \(\phi_i^\nu(r)\) we may explore the role of negative continuum in computing of the properties of two–photon emission from hydrogen–like ions.

### C. Semi–relativistic approximation

Based on the relativistic theory, the expressions obtained in the previous section allow to study the influence of the Dirac’s negative continuum on the properties of two–photon emission from hydrogen–like ions with nuclear charge in the whole range \(1 \leq Z \leq 92\). For the low–Z ions, moreover, it is also useful to estimate the negative–energy contributions within the semi–relativistic approach as proposed in the work by Labzowsky and co–workers [34]. To perform such a semi–relativistic analysis let us start from Eq. \(\text{(11)}\) in which we retain the sum only over the negative–energy continuum states. Since the total energy of these states is \(E_\nu = -(E_i + \omega_j)\), the corresponding energy denominator of the second–order transition amplitude can be written as \(E_\nu - E_i + \omega_j \approx -2mc^2\) which leads to the following expression for the differential decay rate:

\[
\frac{d\omega^{(-)}}{d\omega_1} = \frac{\omega_1\omega_2}{(2\pi)^3c^3} \frac{1}{4(mc^2)^2} \sum_{\nu \in (-)} \left( f |A_1^\nu| \langle \nu | A_2^\nu | i \rangle \right)^2 d\Omega_1 d\Omega_2 .
\]

For the further simplification of this expression we shall make use of the multipole expansion of the electron–photon interaction operators [2]. For the sake of simplicity, we restrict this semi–relativistic analysis to the case of Coulomb gauge \((G = 0)\) in which operator \(A_j^*\) can be written as:

\[
A_j^* = \alpha \cdot \hat{e}_j \left(1 - i\mathbf{k} \cdot \mathbf{r} + 1/2(-i\mathbf{k} \cdot \mathbf{r})^2 + \ldots\right) ,
\]

if one expand the photon exponential \(\exp(i\mathbf{k} \cdot \mathbf{r})\) into the Taylor series.

In contrast to the “standard” spherical tensor expansion [18, 59], the series \(\text{(11)}\) usually does not allow one to make a clear distinction between the different multipole components of the electromagnetic field. For instance, while the first term in Eq. \(\text{(11)}\) describes—within
the non–relativistic limit—electric dipole (E1) transition, the term \((-\textbf{ik} \cdot \textbf{r})\) gives rise both to magnetic dipole (M1) and electric quadrupole (E2) channels. Such an approximation, however, is well justified for our (semi–relativistic) analysis which just aims to estimate the role of negative continuum states in the different (groups of) multipole two–photon transitions in light hydrogen–like ions. In particular, by adopting \(A^a_j = -\alpha \cdot \hat{e}_j (i\textbf{k} \cdot \textbf{r})\) for both operators in Eq. (10) we may find the contribution from the negative spectrum to the 2M1, 2E2 and E2M1 \(2s_{1/2} \rightarrow 1s_{1/2}\) transition probabilities:

\[
\frac{d\omega^{(-)}_{\text{M1,E2}}}{d\omega_1} = \frac{\omega_1 \omega_2}{(2\pi)^3e^3} \frac{1}{4(mc^2)^2} \times \sum_{\nu \in (-)} \left( \langle f | \alpha \cdot \hat{e}_2 (k_2 \cdot r) | \nu \rangle \langle \nu | \alpha \cdot \hat{e}_1 (k_1 \cdot r) | i \rangle \right)^2 \\
\times d\Omega_1 d\Omega_2.
\]  

(12)

Here, summation over the intermediate states \(|\nu\rangle\) is restricted by the negative–energy solutions of the Dirac equation for the electron in the field of nucleus. In the non–relativistic limits these states form a complete set of solutions of the Schrödinger equation for the particle in a repulsive Coulomb filed. By employing a closure relation for such a set we re–write Eq. (12) in the form:

\[
\frac{d\omega^{(-)}_{\text{M1,E2}}}{d\omega_1} = \frac{\omega_1 \omega_2}{(2\pi)^3e^3} \frac{1}{4(mc^2)^2} \times \langle [\hat{e}_1 \hat{e}_2] (f | k_1 \cdot r) (k_2 \cdot r) | i \rangle^2 d\Omega_1 d\Omega_2,
\]

(13)

where \(|i\rangle\) and \(|f\rangle\) denote now the solutions of the Schrödinger equation for the initial and final ionic states, respectively. For the particular case of \(2s_{1/2} \rightarrow 1s_{1/2}\) two–photon transition, i.e., when \(|i\rangle = |2s\rangle\) and \(|f\rangle = |1s\rangle\), this expression finally reads:

\[
\frac{d\omega^{(-)}_{\text{M1,E2}}}{d\omega_1} = \frac{22}{315} \frac{\alpha^{10}}{\pi^2 Z^3} \omega_1^3 \omega_2^3,
\]

(14)

if one performs an integration over the photon emission angles as well as a summation over the polarization states (see Ref. 34 for further details).

Eq. (14) provides the differential rate for the 2M1, 2E2 and E2M1 two–photon transitions as obtained within the non–relativistic framework and by restricting the summation over the intermediate spectrum \(|\nu\rangle\) to the negative energy states only. Being valid for low–Z ions, this expression may also help us to analyze the negative energy contribution to the total decay rate,

\[
\omega_{\text{M1,E2}}^{(-)} = \int \frac{d\omega^{(-)}_{\text{M1,E2}}}{d\omega_1} d\omega_1 = (\alpha Z)^{10} \frac{1}{14 \pi^5 3^6} = 1.247 \times 10^{-6} (\alpha Z)^{10},
\]

(15)

FIG. 3: (Color online) Energy–differential decay rates for the (sum of the) E1M1 and E1E2 \(2p_{1/2} \rightarrow 1s_{1/2}\) multipole two–photon transitions in hydrogen and hydrogen–like ions. Relativistic calculations have been carried out by performing intermediate–state summation over complete Dirac’s spectrum (solid line) as well as by restricting this summation to the positive– (dashed line) and negative–energy (dotted line) states only. Results of relativistic calculations are compared also with the semi–relativistic prediction (dot–dashed line) as given by Eq. (10).

where the integration over the photon energy \(\omega_1\) is performed.

Apart from the 2M1, 2E2 and E2M1 \(2s_{1/2} \rightarrow 1s_{1/2}\) two–photon transitions, Eqs. (10) and (11) may also be employed to study other decay channels. For example, the negative energy contributions to the differential as well as total rates for the E1M1 and E1E2 \(2p_{1/2} \rightarrow 1s_{1/2}\) decay read as:

\[
\frac{d\omega^{(-)}_{\text{E1,M1,E2}}}{d\omega_1} = \frac{217}{312} \frac{\alpha^8}{\pi Z^2} \omega_1 \omega_2 (\omega_1^2 + \omega_2^2),
\]

(16)

and

\[
\omega^{(-)}_{\text{E1,M1,E2}} = (\alpha Z)^8 \frac{2 \times 10^{-5}}{\sqrt{\pi Z^3}} (\alpha Z)^8,
\]

(17)

respectively. Together with Eqs. (14) and (15), we shall later use these non–relativistic predictions in order
to check the validity of our numerical calculations in low–Z domain.

III. RESULTS AND DISCUSSION

Having discussed the theoretical background for the two–photon studies, we are prepared now to analyze the influence of the Dirac’s negative continuum on the total as well as energy–differential decay rates. We shall start such an analysis from the $2s_{1/2} \rightarrow 1s_{1/2}$ transition, which is well established both in theory and in experiment. For all hydrogen–like ions this transition is dominated by the 2E1 decay channel while all the higher multipoles contribute by less than 0.5% to the decay probability. The energy–differential decay rate given by Eq. (5) for the emission of two electric dipole photons is displayed in Fig. 1 for the decay of neutral hydrogen (H) as well as hydrogen–like xenon $Xe^{53+}$ and uranium $U^{91+}$ ions. For these ions, relativistic second–order calculations have been done within the Coulomb gauge and by performing intermediate–state summation over the complete Dirac’s spectrum (solid line) as well as over the positive– (dashed line) and negative–energy (dotted line) solutions only. As seen from the figure, the negative–energy contribution to the energy–differential decay rate is negligible for low–Z ions but becomes rather pronounced as the nuclear charge $Z$ is increased. For the 2E1 decay of hydrogen–like uranium, for example, exclusion of the negative solutions from the intermediate–state summation in Eq. (7) leads to about 20% reduction of the decay rate when compared with the “exact” result.

While for the leading, 2E1 $2s_{1/2} \rightarrow 1s_{1/2}$ transition the negative continuum effects arise only for rather heavy ions, they might strongly affect properties of the higher multipole decay channels in low–Z domain. In Fig. 2, for example, we display the energy distributions of photons emitted in 2M1 and 2E2 transitions. As seen from the upper panel of the figure corresponding to the decay of neutral hydrogen, negative energy part of the Dirac’s spectrum gives the dominant contribution to the (sum of the) differential rates for these decay channels. With the increasing nuclear charge $Z$, the role of positive energy solutions also becomes more pronounced. However, these solutions allow one to describe reasonably well the differential rates only if one of the photons is much more energetic than the second one, i.e., when either $y < 0.1$ or $y > 0.9$. For a nearly equal energy sharing ($y \approx 0.5$), in contrast, accurate relativistic calculations of the 2M1 and 2E2 rates obviously require summation over both, the negative and the positive energy states.

Apart from the results of relativistic calculations, we also display in Fig. 2 the (sum of the) negative–energy contributions to the 2M1, 2E2, M1E2 and E2M1 $2s_{1/2} \rightarrow 1s_{1/2}$ transition probabilities as obtained within the semi–relativistic approach discussed in Section II C. As expected, for low–Z ions both the relativistic (dotted line) and semi–relativistic (dot–dashed line) results basically coincide and are well described by Eq. (14). As the nuclear charge $Z$ is increased, however, semi–relativistic treatment leads to a slight underestimation of the negative–energy contribution to the two–photon (differential) transition probabilities. For the $2s_{1/2} \rightarrow 1s_{1/2}$ decay of hydrogen–like uranium ion, for example, results obtained from Eq. (14) is about 30% smaller than the corresponding relativistic predictions.

Up to now, we have been considering the $2s_{1/2} \rightarrow 1s_{1/2}$ two–photon decay of the hydrogen–like ions. Apart from this —experimentally well studied—transition, recent theoretical interest has been focused also on the $2p_{1/2} \rightarrow 1s_{1/2}$ two–photon decay. Although such a channel is rather weak comparing to the leading one–photon E1 transition, its detailed investigation is highly required for future experiments on the parity violation in simple atomic systems. A number of calculations have been performed, therefore, for the transition probabilities of the dominant E1M1 and E1E2 multipole components. In order to discuss the role of Dirac’s negative continuum in these calculations, we display in Fig. 3 the energy–differential rate for the sum of the E1M1 and E1E2 $2p_{1/2} \rightarrow 1s_{1/2}$ two–photon transitions. Again, the calculations have been carried out within the Coulomb gauge for the electron–photon coupling and for three nuclear charges $Z = 1, 54$ and $92$. As seen from the figure, negative–energy summation in the second–order transition amplitude is of great importance for accurate evaluation of $2p_{1/2} \rightarrow 1s_{1/2}$ transition probabilities both for low–Z and high–Z ions. That is, restriction of the intermediate–state summation to positive part of Dirac’s spectrum results in an overestimation of the E1M1 and E1E2 differential in energy decay rates by factors of about 2 and 2.5 for the neutral hydrogen and hydrogen–like uranium, respectively.

Similarly to the $2s_{1/2} \rightarrow 1s_{1/2}$ multipole transitions, we make use of semi–relativistic formulae from Section II C to cross–check our relativistic computations for the negative–energy contribution to the E1M1 and E1E2 $2p_{1/2} \rightarrow 1s_{1/2}$ decay rates in low–Z domain. Again, while for neutral hydrogen both, semi–relativistic and relativistic approximations produce virtually identical results, they start to differ as the nuclear charge $Z$ is increased.

So far we have discussed the energy–differential decay rates both for $2s_{1/2} \rightarrow 1s_{1/2}$ and $2p_{1/2} \rightarrow 1s_{1/2}$ two–photon transitions. Integration of these rates over the energy of one of the photons (see Eq. (8)) will yield the total decay rates. In Table I we display the total decay rates for the various multipole channels of $2s_{1/2} \rightarrow 1s_{1/2}$ two–photon decay. In contrast to the photon energy distributions from above, here relativistic calculations have been performed in Coulomb (velocity) as well as Babushkin (length) gauges. In both gauges, negative–energy contribution to the (total) probability of the leading 2E1 transition is about eight orders of magnitude smaller than positive–energy term if decay of low–Z ions is considered but is significantly in-
creased for higher nuclear charges. For the hydrogen–like uranium, for example, the total E21 decay rate is enhanced from \(2.9041 \times 10^{12} \text{ s}^{-1}\) in the velocity gauge and \(2.3939 \times 10^{12} \text{ s}^{-1}\) in the length gauge to the—gauge independent—“exact” value of \(3.8256 \times 10^{12} \text{ s}^{-1}\) if, apart from the positive–energy states, the Dirac’s states with negative energy are taken into account in transition amplitude Eq. (7). These results clearly indicate the importance of the negative–state summation for the accurate evaluation of 2E1 \(2s_{1/2} \rightarrow 1s_{1/2}\) total rates in both, velocity and length gauges. It worth mentioning, however, that while for velocity gauge our findings are in perfect agreement with results reported in Ref. [34] some discrepancy was found for calculations performed in length gauge for which Labzowsky and co–workers [34] some discrepancy was found for calculations performed in length gauge for which Labzowsky and co–workers have argued that the contribution from the Dirac’s negative continuum is negligible even for heaviest ions. The reason for this discrepancy is not apparent for the moment and, hence, further investigations are highly required.

In Table I, besides the leading 2E1 decay channel, we present the results of relativistic calculations for the higher multipole contributions to the \(2s_{1/2} \rightarrow 1s_{1/2}\) two–photon transition. The influence of Dirac’s negative continuum is obviously different for various multipole combinations. While, for example, the negative–energy contribution to the intermediate–state summation in low–\(Z\) domain is negligible for the 2E1 decay it becomes of paramount importance for the 2E2 and 2M1 decay channels; an effect that has been already discussed for the case of the energy–differential decay rates (see upper panel of Fig. 2). Moreover, \(2s_{1/2} \rightarrow 1s_{1/2}\) transition with emission of two magnetic dipole (2M1) photons in light ions seems to happen almost exclusively via the negative energy (virtual) intermediate states. The total decay rate for this transition together with the negative–energy contribution to the probability of the 2E2 channel (evaluated in Coulomb gauge) gives in atomic units:

\[
w_{2M1} + w^{(-)}_{2E2} = 1.248 \times 10^{-6} (\alpha Z)^{10},
\]

which is in perfect agreement with the semi–relativistic formula [15].

As mentioned above for the computation of the photon energy distributions in low–\(Z\) domain, negative–energy contribution to the intermediate–state summation is rather pronounced not only for the higher multipole terms of \(2s_{1/2} \rightarrow 1s_{1/2}\) decay but also for the leading E1M1 and E1M2 (two–photon) channels of \(2p_{1/2} \rightarrow 1s_{1/2}\) transition. Our relativistic calculations displayed in Table II indicate that one should account for negative–continuum summation also for an accurate evaluation of the total decay rates for these two decay channels. For the decay of light elements, sizable contribution from the negative–continuum intermediate states arises both in length and velocity gauges. Again, these results partially question the predictions by Labzowsky and co–workers [34] who claimed a minor role of negative energy terms for E1M1 and E1M2 calculations in length gauge. For the velocity gauge, in contrast, our relativistic calculations:

\[
w^{(-)}_{E1M1} + w^{(-)}_{E1E2} = 5.822 \times 10^{-5} (\alpha Z)^{8},
\]

are in a good agreement both, with the semi–relativistic prediction [17] and data presented in Ref. [34].

**IV. SUMMARY AND OUTLOOK**

In conclusion, the two–photon decay of hydrogen–like ions has been re–investigated within the framework of second–order perturbation theory, based on Dirac’s relativistic equation. Special attention has been paid to the summation over the intermediate ionic states which

| \(Z=1\) | \(Z=54\) | \(Z=92\) |
|---|---|---|
| \(\text{length}\) | \(\text{velocity}\) | \(\text{length}\) | \(\text{velocity}\) | \(\text{length}\) | \(\text{velocity}\) |
| 2E1 | 8.2291 (+00) | 1.6311 (+11) | 1.6023 (+11) | 2.9041 (+12) | 2.3939 (+12) |
| \(W_+\) | 2.5372 (-10) | 1.9521 (+04) | 2.9087 (+09) | 3.3321 (+08) | 2.4730 (+09) |
| \(W_{-}\) | 3.7296 (-11) | 9.1765 (+06) | 9.1743 (-21) | 2.5372 (-10) | 2.5372 (-10) |
| \(W_t\) | 4.5092 (-11) | 4.9278 (+07) | 4.2790 (+03) | 1.3804 (-11) | 1.3804 (-11) |
| \(W_{\overline{t}}\) | 4.9072 (-12) | 9.8177 (+05) | 9.8177 (+05) | 1.3804 (-11) | 1.3804 (-11) |

| \(2M1\) | \(W_+\) | 5.9021 (-20) | 1.2691 (+05) | 3.3231 (+08) |
| \(W_{-}\) | 1.3804 (-11) | 3.2695 (+06) | 7.9720 (+08) |
| \(W_t\) | 1.3804 (-11) | 3.4027 (+06) | 1.1093 (+09) |

**TABLE I:** Total rates (in \(\text{s}^{-1}\)) for the several multipole combinations of \(2s_{1/2} \rightarrow 1s_{1/2}\) two–photon decay. Relativistic calculations have been performed within the velocity and length gauges and by carrying out intermediate–state summation over the complete Dirac’s spectrum \((W_t)\) as well as over the positive– (\(W_+\)) and negative–energy (\(W_{-}\)) solutions only.
occurs in such a framework and runs over complete one–
particle spectrum, including a summation over discrete
(bound) states as well as the integration over the posi-
tive and negative continua. In particular, we discussed
the role of the negative energy continuum in an accurate
evaluation of the second–order transition amplitudes and,
herefore, the energy–differential as well as total decay rates.
Detailed calculations of these rates have been presented
for the $2s_{1/2} \rightarrow 1s_{1/2}$ and $2p_{1/2} \rightarrow 1s_{1/2}$ two–photon
transitions in neutral hydrogen as well as hydrogen–like
xenon and uranium ions. As seen from the results ob-
tained, both the total decay probabilities and the en-
ergy distributions of the simultaneously emitted photons

In the present work, we have restricted our discussion
of the negative energy contribution to the second–order
calculations of the total and energy–differential decay rates.
Even stronger effects due to the Dirac’s negative
continuum can be expected, however, for the angular and
polarization correlations between emitted photons. Theo-
retical investigation of these correlations which requires
also detailed analysis of interference terms between the
various (two–photon) multipole combinations is currently
underway and will be published soon.

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**Table II:** Total rates (in s$^{-1}$) for the several multipole combinations of $2p_{1/2} \rightarrow 1s_{1/2}$ two–photon decay.

|       | $Z=1$ | $Z=54$ | $Z=92$ |
|-------|-------|--------|--------|
|       | length | velocity | length | velocity | length | velocity |
| $W_+$ | 4.1934 (-05) | 3.2256 (-05) | 3.0381 (+09) | 2.2422 (+09) | 12.180 (+11) | 1.4126 (+11) |
| $W_-$ | 1.9355 (-05) | 9.6773 (-06) | 1.5701 (+09) | 7.7417 (+08) | 1.3745 (+11) | 6.5902 (+10) |
| $W_1$ | 9.6767 (-06) | 9.6767 (-06) | 6.3731 (+08) | 6.3731 (+08) | 3.8633 (+10) | 3.8633 (+10) |
| $W_+$ | 3.6716 (-05) | 1.2339 (-06) | 2.6077 (+09) | 8.6699 (+07) | 1.7827 (+11) | 6.3367 (+09) |
| $W_-$ | 4.5159 (-05) | 9.6769 (-06) | 3.1980 (+09) | 8.6699 (+07) | 2.1653 (+11) | 4.598 (+10) |
| $W_1$ | 6.6117 (-06) | 6.6117 (-06) | 4.2942 (+08) | 4.2942 (+08) | 2.3584 (+10) | 2.3584 (+10) |

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