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A. Surzhykov, J.P. Santos, P. Amaro, Paul Indelicato. Negative continuum effects on the two-photon decay rates of hydrogen-like ions. 2009. hal-00410134

HAL Id: hal-00410134
https://hal.science/hal-00410134

Preprint submitted on 17 Aug 2009

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

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(Dated: August 17, 2009)

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 $2s_{1/2} \to 1s_{1/2}$ and $2p_{1/2} \to 1s_{1/2}$ transitions in neutral hydrogen H as well as for hydrogen–like xenon Xe^{53+} and uranium 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.

PACS numbers: 31.30.J-,32.80.Wr

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 $2p_{1/2} \to 1s_{1/2}$ total decay probabilities. The relativistic calculations have indicated the importance of negative–energy contributions not only for high–Z but also for low–Z hydrogen–like ions.

In this work, we apply the second–order perturbation theory based on relativistic Dirac's equation in order to re–analyze atomic two–photon decay. We pay special attention to the influence of negative continuum solutions on the evaluation of the transition amplitudes and, hence, on the total and energy–differential decay rates. For the sake of clarity, we restrict our analysis to the decay of hydrogen–like ions for which both the positive– and negative–energy parts of Dirac's spectrum can be still studied in a systematic way by making use of a finite basis set method [19]. Implementation of this method for computing relativistic second–order transition amplitudes is briefly discussed in Sections II A and II B. Later, in Section II C, we consider an alternative, semi–classical, approach which allows *analytical* evaluation of the negative–energy contributions to the two–photon matrix elements and transition rates. These two—semi–classical and fully relativistic—approaches are used in Section III to calculate the energy–differential and total decay rates for several multipole terms in the $2s_{1/2} \to 1s_{1/2}$ and $2p_{1/2} \to 1s_{1/2}$ two–photon decay of neutral hydrogen as well as hydrogen–like xenon Xe^{53+} and uranium U^{91+} ions. Based on the results of our calculations, we argue that both the total transition probabilities and the photon energy distributions can be strongly affected by the negative–state contributions; this effect is most clearly observed for the non–dipole transitions not only in high–Z but also in (non–relativistic) low–Z domain. Brief summary of these findings and outlooks are given finally in Section IV.

II. THEORY

A. Differential and total decay rates

Not much has to be said about the basic formalism for studying the two–photon transitions in hydrogen–like ions. In the past, this formalism has been widely applied in order to investigate not only the total decay probabilities [17, 18, 19, 34] but also the energy as well as angular distributions [23] and even the correlation in the polarization state of the photons [15, 24]. Below, therefore, we restrict ourselves to a rather brief account of the basic expressions, just enough for discussing the role of negative–energy solutions of Dirac's equation in computing of the two–photon (total and differential) rates.

The properties of the two–photon atomic transitions are evaluated, usually, within the framework of the second–order perturbation theory. When based on Dirac's equation, this theory gives the following expression for the differential in energy decay rate:

$$\frac{\mathrm{d}w}{\mathrm{d}\omega_1} = \frac{\omega_1\omega_2}{(2\pi)^3 c^3} \left| \sum_\nu \left(\frac{\langle f\,|\boldsymbol{A}_2^*|\,\nu\rangle\,\langle \nu\,|\boldsymbol{A}_1^*|\,i\rangle}{E_\nu - E_i + \omega_1} + \frac{\langle f\,|\boldsymbol{A}_1^*|\,\nu\rangle\,\langle \nu\,|\boldsymbol{A}_2^*|\,i\rangle}{E_\nu - E_i + \omega_2} \right) \right|^2 \mathrm{d}\Omega_1 \mathrm{d}\Omega_2\,, \qquad (1)$$

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

$$\boldsymbol{A}_j^* = \boldsymbol{\alpha} \cdot (\hat{\boldsymbol{e}}_j + G\boldsymbol{k}_j)\, \mathrm{e}^{-i\boldsymbol{k}_j \boldsymbol{r}} - G\mathrm{e}^{-i\boldsymbol{k}_j \boldsymbol{r}}\,, \qquad (2)$$

where $\boldsymbol{\alpha}$ 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), $|i\rangle \equiv |n_i\kappa_i\mu_i\rangle$ and $|f\rangle \equiv |n_f\kappa_f\mu_f\rangle$ denote solutions of the Dirac's equation for the initial and final ionic states while $E_i \equiv E_{n_i\kappa_i}$ and $E_f \equiv E_{n_f\kappa_f}$ are the corresponding one–particle energies. Because of energy conservation, E_i and E_f are related to the energies $\omega_{1,2} = ck_{1,2}$ of the emitted photons by:

$$E_i - E_f = \omega_1 + \omega_2\,. \qquad (3)$$

¿From this relation, it is convenient to define the so-called energy sharing parameter $y = \omega_1/(\omega_1 + \omega_2)$, 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 $\boldsymbol{A}_j^*$ (see Refs. [18, 19, 39] for further details). By using such an expansion, we are able to re–write Eq. (1) as a sum of partial multipole rates

$$\frac{\mathrm{d}w}{\mathrm{d}\omega_1} = \sum_{\Theta_1 L_1 \Theta_2 L_2} \frac{\mathrm{d}W_{\Theta_1 L_1 \Theta_2 L_2}}{\mathrm{d}\omega_1}\,, \qquad (4)$$

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

$$\frac{\mathrm{d}W_{\Theta_1 L_1 \Theta_2 L_2}}{\mathrm{d}\omega_1} = \frac{\omega_1\omega_2}{(2\pi)^3 c^3} \sum_{\lambda_{\Theta_1}\lambda_{\Theta_2}} \sum_{\kappa_\nu} \left[\left| S^{j_\nu}(1,2) \right|^2 + \left| S^{j_\nu}(2,1) \right|^2 + 2 \sum_{\kappa'_\nu} d(j_\nu, j'_\nu)\, S^{j_\nu}(2,1) S^{j'_\nu}(1,2) \right], \quad (5)$$

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

$$d(j_\nu, j'_\nu) = \sqrt{(2j_\nu+1)(2j'_\nu+1)} \times (-1)^{2j_\nu + L_1 + L + 2} \left\{ \begin{array}{ccc} j_f & j'_\nu & L_1 \\ j_i & j_\nu & L_2 \end{array} \right\}, \quad (6)$$

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

$$S^{j_\nu}(1,2) = \sum_{n_\nu} \frac{\left\langle n_f\kappa_f \left|\left| \hat{a}^{\lambda_{\Theta_1}*}_{L_1} \right|\right| n_\nu\kappa_\nu \right\rangle \left\langle n_\nu\kappa_\nu \left|\left| \hat{a}^{\lambda_{\Theta_2}*}_{L_2} \right|\right| n_i\kappa_i \right\rangle}{E_\nu - E_i + \omega_2} . \quad (7)$$

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

Until now, we have discussed the general expressions for the two–photon transition rates which are differential in energy ω_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:

$$w_{\mathrm{tot}} = \sum_{\Theta_1 L_1 \Theta_2 L_2} W_{\Theta_1 L_1 \Theta_2 L_2} \equiv \sum_{\Theta_1 L_1 \Theta_2 L_2} \int_0^{\omega_t} \frac{\mathrm{d}W_{\Theta_1 L_1 \Theta_2 L_2}}{\mathrm{d}\omega_1} \mathrm{d}\omega_1 , \quad (8)$$

where $\omega_t = E_i - E_f$ 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, 35]. Instead, in the next section we will focus on the summation over the intermediate states $|n_\nu\kappa_\nu\rangle$ which appears in the second–order transition amplitudes (see Eq. (7)).

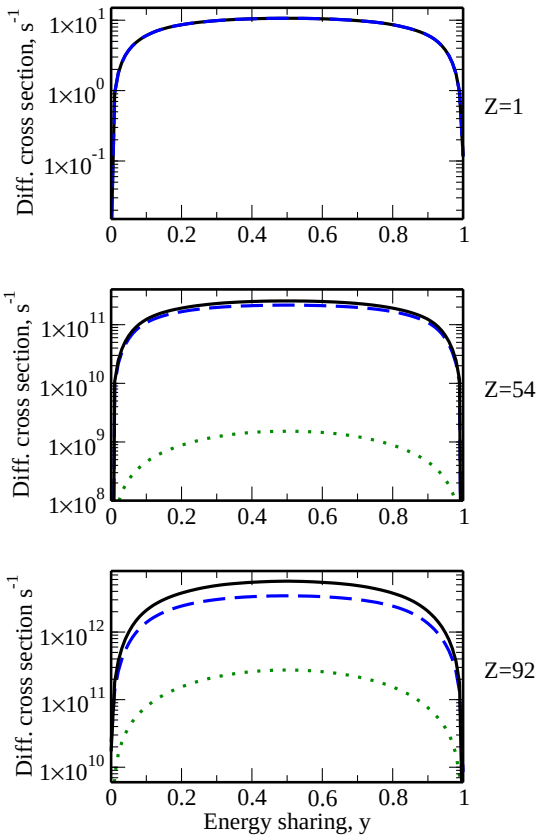


FIG. 1: (Color online) Energy–differential transition rates for the 2E1 $2s_{1/2} \to 1s_{1/2}$ two–photon decay of hydrogen and hydrogen–like ions. Relativistic calculations have been carried out by performing intermediate–state summation over complete Dirac's spectrum (solid line) as well as by restricting this summation to the positive– (dashed line) and negative–energy (dotted line) states only.

B. Summation over the intermediate states

The summation over the intermediate states in Eq. (7) runs over the complete one–particle spectrum $|n_\nu\kappa_\nu\rangle$, including a summation over the discrete part of the spectrum as well as an integration over the positive and negative–energy continuum. In practice, of course, performing such an infinite–state summation is a rather demanding task. A number of methods have been developed over the last decades in order to evaluate the second–order transition amplitudes consistently. Apart from the Green's function approach [23, 36] which — in case of a purely Coulomb potential— allows for the analytical computation of Eq. (7), the *discrete–basis–set* summation is widely used nowadays in two–photon studies [19]. A great advantage of the latter method is that it allows to separate the contributions from the positive– and negative–energy solutions in the intermediate–state summation. Since the effects that arise from the negative–energy spectrum are in the focus of the present study, we apply for the calculations below

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 $i = 1, .., 2N$ which, in turn, are found as solutions of the Dirac–Fock equation,

$$\begin{bmatrix} \frac{V(r)}{c} & \frac{\mathrm{d}}{\mathrm{d}r} - \frac{\kappa_\nu}{R} \\ -\left(\frac{\mathrm{d}}{\mathrm{d}r} + \frac{\kappa_\nu}{R}\right) & -2c + \frac{V(r)}{c} \end{bmatrix} \phi^i_\nu(r) = \frac{\epsilon^i_\nu}{c} \phi^i_\nu(r)\,, \qquad (9)$$

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 $i = 1, .., N$ describe the negative continuum with $\epsilon^i_\nu < -2mc^2$ while solutions labeled by $i = N+1, .., 2N$ 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 $1 \le Z \le 92$. 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. (1) in which we retain the sum only over the negative–energy continuum states. Since the total energy of these states is $E_\nu = -(T_\nu + mc^2)$, 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:

$$\frac{\mathrm{d}w^{(-)}}{\mathrm{d}\omega_1} = \frac{\omega_1 \omega_2}{(2\pi)^3 c^3} \frac{1}{4(mc^2)^2} \left| \sum_{\nu \in (-)} \Big(\langle f\,|\boldsymbol{A}^*_2|\,\nu\rangle \langle \nu\,|\boldsymbol{A}^*_1|\,i\rangle \right.$$

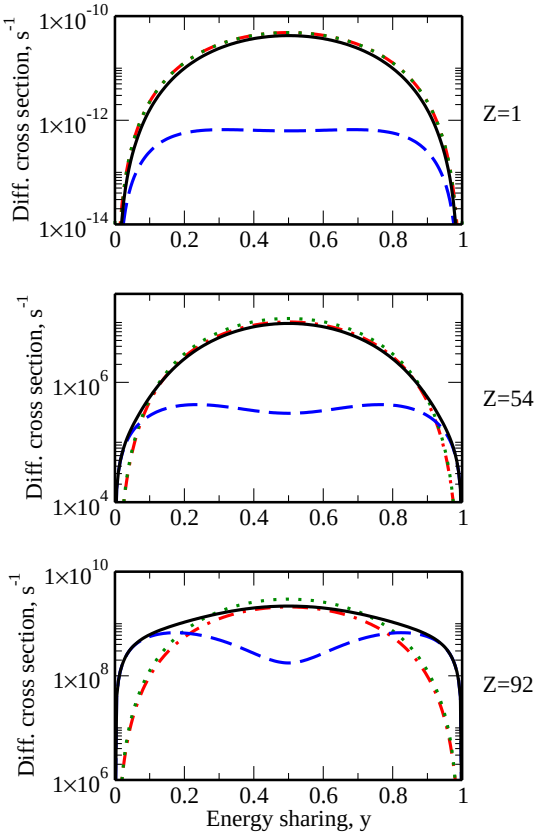


FIG. 2: (Color online) Energy–differential decay rates for the (sum of the) 2M1, 2E2 and E2M1 $2s_{1/2} \to 1s_{1/2}$ multipole two–photon transitions in hydrogen and hydrogen–like ions. Relativistic calculations have been carried out by performing intermediate–state summation over complete Dirac's spectrum (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. (14).

$$\left. + \langle f\,|\boldsymbol{A}^*_1|\,\nu\rangle \langle \nu\,|\boldsymbol{A}^*_2|\,i\rangle \Big) \right|^2 \mathrm{d}\Omega_1 \mathrm{d}\Omega_2\,. \qquad (10)$$

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 $\boldsymbol{A}^*_j$ can be written as:

$$\boldsymbol{A}^*_j = \boldsymbol{\alpha} \cdot \hat{\boldsymbol{e}}_j \left(1 - i\boldsymbol{k}\cdot\boldsymbol{r} + 1/2\,(-i\boldsymbol{k}\cdot\boldsymbol{r})^2 + ...\right), \qquad (11)$$

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

In contrast to the "standard" spherical tensor expansion [18, 39], the series (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. (11) describes—within

the non–relativistic limit—electric dipole (E1) transition, the term $(-i\boldsymbol{k}\cdot\boldsymbol{r})$ 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 $\boldsymbol{A}^*_j = -\boldsymbol{\alpha}\cdot\hat{\boldsymbol{e}}_j\,(i\boldsymbol{k}\cdot\boldsymbol{r})$ for both operators in Eq. (10) we may find the contribution from the negative spectrum to the 2M1, 2E2 and E2M1 $2s_{1/2} \to 1s_{1/2}$ transition probabilities:

$$\frac{\mathrm{d}w^{(-)}_{\mathrm{M1,E2}}}{\mathrm{d}\omega_1} = \frac{\omega_1\omega_2}{(2\pi)^3c^3}\,\frac{1}{4(mc^2)^2}$$
$$\times\left|\sum_{\nu\in(-)}\Big(\langle f\,|\boldsymbol{\alpha}\cdot\hat{\boldsymbol{e}}_2\,(\boldsymbol{k}_2\cdot\boldsymbol{r})|\,\nu\rangle\,\langle\nu\,|\boldsymbol{\alpha}\cdot\hat{\boldsymbol{e}}_1\,(\boldsymbol{k}_1\cdot\boldsymbol{r})|\,i\rangle\right.$$
$$\left.+\,\langle f\,|\boldsymbol{\alpha}\cdot\hat{\boldsymbol{e}}_1\,(\boldsymbol{k}_1\cdot\boldsymbol{r})|\,\nu\rangle\,\langle\nu\,|\boldsymbol{\alpha}\cdot\hat{\boldsymbol{e}}_2\,(\boldsymbol{k}_2\cdot\boldsymbol{r})|\,i\rangle\Big)\right|^2$$
$$\times\mathrm{d}\Omega_1\mathrm{d}\Omega_2\,. \qquad (12)$$

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 [40]. By employing a closure relation for such a set we re–write Eq. (12) in the form:

$$\frac{\mathrm{d}w^{(-)}_{\mathrm{M1,E2}}}{\mathrm{d}\omega_1} = \frac{\omega_1\omega_2}{(2\pi)^3c^3}\,\frac{1}{(mc^2)^2}$$
$$\times\,|(\hat{\boldsymbol{e}}_1\hat{\boldsymbol{e}}_2)\,\langle f\,|(\boldsymbol{k}_1\cdot\boldsymbol{r})(\boldsymbol{k}_2\cdot\boldsymbol{r})|\,i\rangle|^2\;\mathrm{d}\Omega_1\mathrm{d}\Omega_2\,, \qquad (13)$$

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 $2s_{1/2} \to 1s_{1/2}$ two–photon transition, i.e., when $|i\rangle = |2s\rangle$ and $|f\rangle = |1s\rangle$, this expression finally reads:

$$\frac{\mathrm{d}w^{(-)}_{\mathrm{M1,E2}}}{\mathrm{d}\omega_1} = \frac{2^{22}}{3^{13}}\,\frac{\alpha^{10}}{5\pi Z^4}\,\omega_1^3\omega_2^3\,, \qquad (14)$$

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,

$$w^{(-)}_{\mathrm{M1,E2}} = \int\frac{\mathrm{d}w^{(-)}_{M1,E2}}{\mathrm{d}\omega_1}\,\mathrm{d}\omega_1 = (\alpha Z)^{10}\,\frac{1}{14\,\pi\,5^2\,3^6}$$
$$= 1.247\,10^{-6}\,(\alpha Z)^{10}\,, \qquad (15)$$

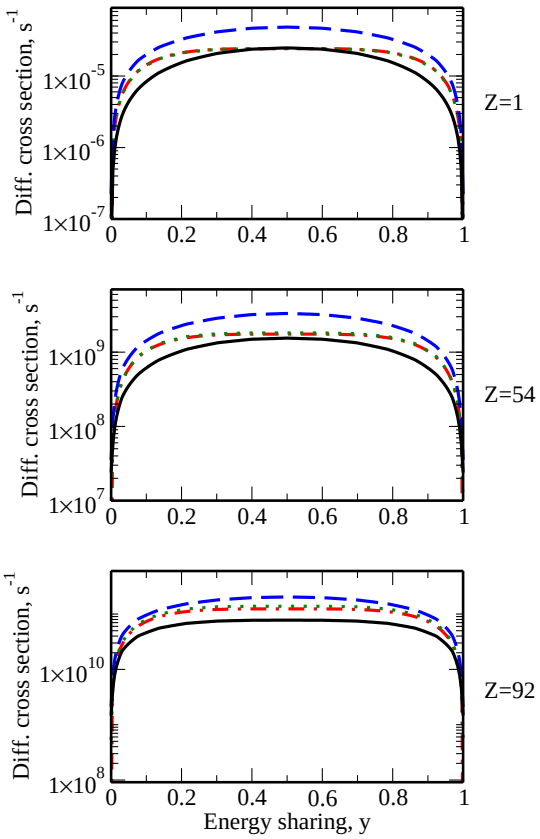


FIG. 3: (Color online) Energy–differential decay rates for the (sum of the) E1M1 and E1E2 $2p_{1/2} \to 1s_{1/2}$ multipole two–photon transitions in hydrogen and hydrogen–like ions. Relativistic calculations have been carried out by performing intermediate–state summation over complete Dirac's spectrum (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. (16).

where the integration over the photon energy ω_1 is performed.

Apart from the 2M1, 2E2 and E2M1 $2s_{1/2} \to 1s_{1/2}$ 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 $2p_{1/2} \to 1s_{1/2}$ decay read as:

$$\frac{\mathrm{d}w^{(-)}_{\mathrm{E1,M1,E2}}}{\mathrm{d}\omega_1} = \frac{2^{17}}{3^{12}}\,\frac{\alpha^8}{\pi Z^2}\,\omega_1\omega_2(\omega_1^2+\omega_2^2)\,, \qquad (16)$$

and

$$w^{(-)}_{\mathrm{E1,M1,E2}} = (\alpha Z)^8\,\frac{2}{5\pi\,3^7}$$
$$= 5.822\,10^{-5}\,(\alpha Z)^8\,, \qquad (17)$$

respectively [34]. 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 $2s_{1/2} \to 1s_{1/2}$ transition, which is well established both in theory [18, 19, 23] 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 $2s_{1/2} \to 1s_{1/2}$ 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 (5) 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 $2s_{1/2} \to 1s_{1/2}$ 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 $2s_{1/2} \to 1s_{1/2}$ 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 $2s_{1/2} \to 1s_{1/2}$ two–photon decay of the hydrogen–like ions. Apart from this —experimentally well studied—transition, recent theoretical interest has been focused also on the $2p_{1/2} \to 1s_{1/2}$ *two–photon* decay [34]. 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 [41]. A number of calculations [34, 42] 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 $2p_{1/2} \to 1s_{1/2}$ 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 (7) is of great importance for accurate evaluation of $2p_{1/2} \to 1s_{1/2}$ 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 $2s_{1/2} \to 1s_{1/2}$ 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 $2p_{1/2} \to 1s_{1/2}$ decay rates in low–Z domain. Again, while for neutral hydrogen both, semi–relativistic (16) 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 $2s_{1/2} \to 1s_{1/2}$ and $2p_{1/2} \to 1s_{1/2}$ 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 $2s_{1/2} \to 1s_{1/2}$ 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-

		Z=1		Z=54		Z=92	
		length	velocity	length	velocity	length	velocity
2E1	W_+	8.2291 (+00)	8.2291 (+00)	1.6311 (+11)	1.6023 (+11)	2.9041 (+12)	2.3939 (+12)
	W_-	2.4949 (-08)	6.2372 (-09)	3.8442 (+09)	9.6290 (+08)	6.8066 (+11)	1.7044 (+11)
	W_t	8.2291 (+00)	8.2291 (+00)	1.8592 (+11)	1.8592 (+11)	3.8256 (+12)	3.8256 (+12)
E1M2	W_+	2.5372 (-10)	2.5372 (-10)	4.7949 (+07)	4.7940 (+07)	8.2955 (+09)	8.2714 (+09)
	W_-	9.1743 (-21)	4.5871 (-21)	1.9521 (+04)	9.7905 (+03)	4.8084 (+07)	2.4070 (+07)
	W_t	2.5372 (-10)	2.5372 (-10)	4.9278 (+07)	4.9278 (+07)	9.1387 (+09)	9.1387 (+09)
2E2	W_+	3.7296 (-11)	4.8617 (-13)	9.1765 (+06)	1.9624 (+05)	2.4730 (+09)	9.7383 (+07)
	W_-	4.5092 (-11)	8.2822 (-12)	1.1000 (+07)	2.0202 (+06)	2.9087 (+09)	5.3305 (+08)
	W_t	4.9072 (-12)	4.9072 (-12)	9.8177 (+05)	9.8177 (+05)	1.7859 (+08)	1.7859 (+08)
2M1	W_+	5.9021 (-20)		1.2691 (+05)		3.3321 (+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 s^{-1}) for the several multipole combinations of $2s_{1/2} \to 1s_{1/2}$ 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.

creased for higher nuclear charges. For the hydrogen–like uranium, for example, the total 2E1 decay rate is enhanced from 2.9041×10^{12} s^{-1} in the velocity gauge and 2.3939×10^{12} s^{-1} in the length gauge to the—gauge independent—"exact" value of 3.8256×10^{12} 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 $2s_{1/2} \to 1s_{1/2}$ 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 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 $2s_{1/2} \to 1s_{1/2}$ 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 E1M2 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, $2s_{1/2} \to 1s_{1/2}$ 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:

$$w_{2M1} + w^{(-)}_{2E2} = 1.248\,10^{-6}\,(\alpha Z)^{10}\,, \qquad (18)$$

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 $2s_{1/2} \to 1s_{1/2}$ decay but also for the leading E1M1 and E1M2 (two–photon) channels of $2p_{1/2} \to 1s_{1/2}$ 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:

$$w^{(-)}_{E1M1} + w^{(-)}_{E1E2} = 5.822\,10^{-5}\,(\alpha Z)^{8}\,, \qquad (19)$$

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	
		length	velocity	length	velocity	length	velocity
E1M1	W_+	4.1934 (-05)	3.2256 (-05)	3.0381 (+09)	2.2422 (+09)	2.1280 (+11)	1.4126 (+11)
	W_-	1.9355 (-05)	9.6773 (-06)	1.5701 (+09)	7.7417 (+08)	1.3745 (+11)	6.5902 (+10)
	W_t	9.6767 (-06)	9.6767 (-06)	6.3731 (+08)	6.3731 (+08)	3.8633 (+10)	3.8633 (+10)
E1E2	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)	6.7698 (+08)	2.1653 (+11)	4.4598 (+10)
	W_t	6.6117 (-06)	6.6117 (-06)	4.2942 (+08)	4.2942 (+08)	2.3584 (+10)	2.3584 (+10)

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

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 positive 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, hence, 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 obtained, both the total decay probabilities and the energy distributions of the simultaneously emitted photons can be strongly affected by the negative–state summation not only for heavy ions but also for low–Z domain. We demonstrate, however, that the role of Dirac's negative continuum becomes most pronounced for the higher (non–dipole) terms in the expansion of the electron–photon interaction; similar effect has been recently reported for the theoretical description of hydrogen–like systems exposed to intense electromagnetic pulses [30].

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. Theoretical 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.

Acknowledgements

A.S. acknowledges support from the Helmholtz Gemeinschaft and GSI under the project VH–NG–421. This research was supported in part by FCT Project No. POCTI/0303/2003 (Portugal), financed by the European Community Fund FEDER, by the French-Portuguese collaboration (PESSOA Program, Contract No. 441.00), and by the Acções Integradas Luso-Francesas (Contract No. F-11/09) and Luso-Alemãs (Contract No. A-19/09). Laboratoire Kastler Brossel is " Unité Mixte de Recherche du CNRS, de l' ENS et de l' UPMC No. 8552." P.A. acknowledges the support of the FCT, under Contract No. SFRH/BD/37404/2007. This work was also partially supported by Helmholtz Alliance HA216/EMMI.

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