Relativistic evaluation of the two-photon decay of the metastable $1s^22s^2p^3P_0$ state in berylliumlike ions with an active-electron model

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The two-photon $1s^22s^2p^3P_0 \rightarrow 1s^22s^21S_0$ transition in berylliumlike ions is theoretically investigated within a full relativistic framework and a second-order perturbation theory. We focus our analysis on how electron correlation, as well as the negative-energy spectrum can affect the forbidden $E1M1$ decay rate. For this purpose we include the electronic correlation by an effective potential and within an active-electron model. Due to its experimental interest, evaluation of decay rates are performed for berylliumlike xenon and uranium. We find that the negative-energy contribution can be neglected in the present decay rate. On the other hand, if contributions of electronic correlation are not carefully taken into account, it may change the lifetime of the metastable state by 20%. By performing a full-relativistic $jj$-coupling calculation, we found discrepancies for the decay rate of an order of 2 compared to non-relativistic $LS$-coupling calculations, for the selected heavy ions.

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I. INTRODUCTION

Two-photon decay has been studied several times since it was originally discussed by Göppert-Mayer [1]. In low-$Z$ atomic systems, the $2s - 1s$ transitions in hydrogenlike and heliumlike ions occur primarily by two electric dipole photons ($E1E1$), and the respective decay rates provided by theory and experiment are in good agreement. These works focused not only on the total and energy differential decay rates [2–4], but also on the angular and polarization correlations of the emitted two photons [5–9]. Detailed analysis of these two-photon properties have been used to reveal unique information about electron densities in astrophysical plasmas and thermal x-ray sources, as well as highly precise values of physical constants [10]. The study of two-photon decay in high-$Z$ ions also provided a sensitive tool for exploring the relativistic and quantum electromagnetic (QED) effects that occur in the strong atomic fields of those systems. As in the case of low-$Z$ ions, predictions for two-photon decay rates are in good agreement with experimental data [11–14].

Scarcely investigations have been performed so far for other atomic systems with more than two electrons. In the case of lithiumlike ions, this lack of research might be attributed to almost all two-photon transitions being in direct competition with dominant allowed (single $E1$) transitions, thus reducing the importance of the former process in practical applications. However, this is not the case for berylliumlike ions with zero nuclear spin ($I = 0$). Owing to the $0 \rightarrow 0$ selection rule, the first excited state $1s^22s^2p^3P_0$ is metastable and its transition to the ground state $1s^22s^21S_0$ is strictly forbidden for all single-photon multipole modes. The most dominant decay process is a rare two-photon transition with a magnetic dipole mode ($E1M1$) that is very sensitive to relativistic and electronic correlation effects and can have lifetimes from few decades to few minutes, depending on the atomic electromagnetic field of the nucleus.

Knowledge of metastable decay rates are essential in collision-radiative modelling of astrophysical low-density plasmas that occurs in stellar coronae [15], thus many studies have been dedicated to the measurement and calculation of higher-order ($M1, E2$) and hyperfine-induced $E1$ transitions modes [16–18]. First measurements of the metastable hyperfine-induced decay rate in N$^{3+}$ was first performed at the Hubble Space Telescope with important implications to the isotopic abundance in an observed nebula [19].

Values of $E1M1$ decay rate in berylliumlike sulphur can also play an important role, specially because the majority of stable isotopes ($^{32}S$ and $^{34}S$) contains $I = 0$ and have observable quantities in the solar coronae [18, 20, 21].

Besides this astrophysical interest, there is also motivation for calculating the $E1M1$ two-photon decay mode coming from experiments aimed to test the standard model via the observation of parity nonconservation in
berylliumlike uranium [22] [23]. Moreover, some subtle X-ray lines coming from an electron cyclotron resonance (ECR) plasma might be attributed to charge state mechanisms involving the Be-like metastable state 1s22s22p 3P0 [24].

There are no experimental results for the E1M1 decay rates in zero-spin berylliumlike ions available. Only recently, several dielectronic recombination resonances were clearly identified as coming from a parent 1s22s22p 3P0 metastable state in xenon (136Xe+50 with I = 0), which will lead to a forthcoming measurement of the respective E1M1 decay rate [25] [26]. In these recently published works, the need of full relativistic calculations for this decay rate is emphasised.

In isotopes with non-zero I the importance of the E1M1 mode is reduced: the hyperfine mixing between the term 3P0 and the closely-lying above term 3P1 produces states with total angular momentum F ≠ 0, thus circumventing the 0 → 0 selection rule. This drastically reduces the lifetime of the metastable 1s22s22p 3P0 state since it opens an E1 single-photon channel. Decay rates for this hyperfine-induced E1 mode have been known theoretically for some years [18] [27] [29] and the first measurements have already been performed recently, both in laboratory [30] [31] and in galaxy nebula [19]. A review of this topic can be found in Ref. [32].

From the theoretical point of view, the calculation of this two-photon decay rate offers a challenge not only because berylliumlike ions have a compact electron structure, which makes electronic correlation of paramount importance, but also due to relativistic effects, such as the negative-energies. Previous studies about two-photon decay with a M1 component have shown that this negative-energy contribution is mandatory for both low-Z and high-Z ions [33] [34] and improves gauge invariance [35] [37]. Furthermore, similar investigations have concluded that the inclusion of negative-energy continuum gives better agreement with experimental data [38]. Both effects have to be efficiently incorporated in the second-order summation over the intermediate states that characterizes two-photon transitions.

Figure 1 illustrates the compact atomic structure in berylliumlike uranium, where the initial, intermediate and final states are plotted.

Up to now, only two estimations of the E1M1 decay rate for berylliumlike systems are available [39] [40], both assuming a non-relativistic approximation and using LS-coupling, which for high-Z ions may lead to significant deviations. Moreover, the summation over the intermediate states was only restricted to the first terms, 1s22s22p 3P1 and 1s22s22p 1P1.

In this work, we calculate the two-photon decay rate of the metastable 1s22s22p 3P0 state in berylliumlike ions considering a relativistic evaluation of the second-order summation in a jj-coupling active-electron. Negative energies are thus included and investigated. In order to take into account the electronic correlation, we perform the evaluation of the second-order summation via a finite-basis-set and an effective local potential, with a few key intermediate states calculated using the MultiConfiguration Dirac-Fock (MCDF) method. For these evaluations, we consider xenon and uranium, following the reasoning above. For elements below xenon, we notice that the strong electronic correlation prevents the present method of retrieving a reliable decay rate. A model beyond the active electron model is currently under investigation.

II. THEORY

The evaluation of two-photon related quantities have been discussed several times in the literature [11] [12] [41], we, therefore, present here only the final form suitable for further discussion of the influence of the relativistic and electronic correlation effects.

Two-photon processes are evaluated following a second-order perturbation theory, which overall contains a summation over the complete spectrum of a given Hamiltonian. Its elements are often referred as intermediate states. For the present case of the E1M1 two-photon decay between the states 1s22s22p 3P0 and 1s22s2 1S0 (terms are given for state identification), the differential decay rate is given by (atomic units),

\[
\frac{dW}{d\omega_1} = \frac{\omega_1 \omega_2}{(2\pi)^3 c^2} \left| \sum_{j_n=1/2}^{3/2} \left[ S_j^h (2,1) + S_j^h (1,2) \right] \right|^2 ,
\]

(1)

where \(\omega_1\) and \(\omega_2\) are the energies of the two emitted photons, c is the light speed and \(j_n\) is the total angular momentum of the active electron performing the transition. From now on, we write configurations without 1s² for shortness. The sum of both photon energies is
equal to transition energy due to energy conservation, $E_{2s^2 2p^3} - E_{2s^2 2p^4} = \omega_1 + \omega_2 = \omega_t$. The two-photon amplitudes $S^{3s}(2, 1)$ and $S^{3s}(1, 2)$ contain the summation over the reduced matrix elements of the $E1$ and $M1$ multipole components, which are given by,

$$S^{3s}(2, 1) = \sum_n \frac{\langle 2s | 3sP_0 \rangle | n_{vJ}\rangle \langle n_{vJ} | 2p^3P_0 \rangle}{E_n - E_{2s2p^3P_0} + \omega_1}. \quad (2)$$

with the multipole components, electric dipole and magnetic dipole being given by the relativistic radiative operators $R_1 = E_1$ and $R_2 = M_1$, respectively [11]. $S^{3s}(1, 2)$ is given by an equation similar to Eq. (2) by interchanging 1 with 2. In Fig. 1 are represented the first states of the summation for the four two-photon amplitudes allowed by selection rules, which are $2s3s_{1/2}^13s_0$, $2s3d_{3/2}^11D_1$, $2s2p_{3/2}^13P_1$ and $2s2p_{1/2}^11P_1$, for $S^{3s}(2, 1)$, $S^{3s}(2, 1)$, $S^{1s}(1, 2)$ and $S^{3s}(1, 2)$, respectively.

In this work, we consider the active electron model (AEM) [11], i.e., only intermediate states with variations of the active electron’s quantum numbers $n'l'_{j'}$ that participates in the transition $2p \rightarrow n'l'_{j'} \rightarrow 2s$ are taken into account in the summation over the intermediate states. Other intermediate states with excitation of the spectator electron, (like the 1s and 2s occupied orbitals) are thus not taken into account. Atomic states are usually given as a linear combination of configurations within a MCDF or configuration interaction (CI). We hereby define a state with major contribution of a configuration with a spectator-orbital excitation as $C^{\text{exc}}$. $C^{\text{non-exc}}$ are usual states within the AEM, where the major contribution addresses to non-excitation configurations of the spectator-orbital. In order to better justify the AEM, we give in Figure 2 a pictorial representation of one $C^{\text{exc}}$ and one $C^{\text{non-exc}}$. The path (a) corresponds to a state $C^{\text{non-exc}}$ in the AEM. This path is also represented in Fig. 1. Because the radiative operator is a one-body operator, $C^{\text{exc}}$ states give non-null matrix elements only by considering either of these two cases:

- **Path (b)** - Electron orbitals are almost orthogonal between all states, thus if the radiative operator connects the active orbitals, there is a small contribution of $C^{\text{exc}}$ due to $(2s_{\text{ini}}^1|3s_{\text{fin}}^1) \neq 0$ and $(3s_{\text{ini}}^1|2s_{\text{fin}}^1) \neq 0$, where $2s_{\text{ini}}^1$, $3s_{\text{fin}}^1$ and $2s_{\text{fin}}^1$ are spectator orbitals in the initial, $C^{\text{exc}}$ and final states.

- **Path (c)** - The final and initial states can have a reasonable contribution of a configuration with the same spectator orbital as $C^{\text{exc}}$ due to configuration mixing. The configuration coefficients can be obtained by MCDF or CI.

These two cases show how multiconfiguration and fully relaxed orbitals can play a role in two-photon processes by allowing $C^{\text{exc}}$ states beyond the AEM.

![Figure 2](image_url)
TABLE I. Optimal values of $k_1$ and energy differences (eV) obtained by the FBS method without $k_1$ optimization ($E_{\text{FBS}}$), as well as the respective ones obtained by the MCDF method ($E_{\text{MCDF}}$). Energy of $2s^2\ 1^3P_0$ is relative to final state $2s^2\ 1^3S_0$ ($\omega_1$), while the rest are relative to the initial state, $E_n - E_{2s2p_{1/2}}\ 3\nu_0$. Values provided by Ref. [46] are also listed.

| Species | $k_1$ | $E_{\text{FBS}}$ | $E_{\text{MCDF}}$ | Ref. [46] |
|---------|-------|------------------|------------------|-----------|
| Xe$^{50+}$ | $3^3P_0 - 1^3S_0$ | 0.79 | 118.5 | 104.1 | 104.5 |
| | $3^3P_1 - 3^3P_0$ | 0.64 | 23.9 | 22.8 | |
| | $1^3P_1 - 3^3P_0$ | 0.62 | 400.8 | 430.1 | 428.3 | |
| U$^{88+}$ | $3^3P_0 - 1^3S_0$ | 0.69 | 252.7 | 258.1 | 258.3 |
| | $3^3P_1 - 3^3P_0$ | 0.58 | 0.0 | 41.6 | 39.9 | |
| | $1^3P_1 - 3^3P_0$ | 0.72 | 4259.3 | 4245.3 | 4243.3 | |

The energy differences $E_n - E_{2s2p_{1/2}}\ 3\nu_0$ (denominators of Eq. (2)), obtained by the FBS method and with the respective ones of the MCDF method. Table I lists the optimal values of $k_1$ that minimizes the differences between the FBS and MCDF of the mentioned energy differences. The MCDF calculations were performed using the general relativistic MCDF code (MDFGME) [47].

Calculations of the decay rate were performed in both length and velocity gauges. The quality of the evaluation of the two-photon amplitudes, if the potential remains local in all states, is directly connected to the gauge invariance [11] [43]. Although we introduced different local-exchange potentials in the states and MCDF energies, we notice that the gauge invariance is still at a level of few percent.

With the application of the present formalism to the decay of $1s2p\ 3^3P_0$ to the ground state in heliumlike ions, and with an effective potential of $v_0(1s, r)$, we reproduce the results of Ref. [45] within the respective accuracy.

### III. RESULTS AND DISCUSSION

The results of our calculations for the $2s2p\ 3^3P_0 \rightarrow 2s^2\ 1^3S_0\ E1M1$ decay rate $W$ are presented in Table II. The obtained lifetimes corresponds to $\sim 3$ min and 12 s for Xe$^{50+}$ and U$^{88+}$ ions, respectively. Other allowed higher-order multipole contributions to this transition, like the E2M2 or E3M3, are severely reduced. The obtained value for the E2M2 decay rate in berylliumlike uranium of $8.2 \times 10^{-18}$ s$^{-1}$ shows the minimal impact to the total decay rate.

Calculations were performed in both velocity and length gauges, showing differences of up to 4% due to the

TABLE II. Decay rate (s$^{-1}$) for $2s2p\ 3^3P_0 \rightarrow 2s^2\ 1^3S_0\ E1M1$ transition in xenon and uranium. Relativistic calculations have been performed in velocity (V) and length (L) gauges for several cases: with ($W_{\text{opt}}$) and without $k_1$-optimization ($W_{\text{non-opt}}$); with the summation carried without negative energies ($W^+$); having the energies provided by Ref. [46] ($W^*$); without the effective exchange potential ($W_{\text{no-exc}}$). Values of Refs [40] and [23] are listed.

| Species | $W_{\text{opt}}$ | $W_{\text{opt}}$ | $W_{\text{non-opt}}$ | $W_{\text{non-opt}}$ |
|---------|------------------|------------------|---------------------|---------------------|
| Xe$^{50+}$ | $4.78 \times 10^{-3}$ | $4.97 \times 10^{-3}$ | $4.05 \times 10^{-3}$ | $4.05 \times 10^{-3}$ |
| U$^{88+}$ | $8.04 \times 10^{-2}$ | $8.06 \times 10^{-2}$ | $9.35 \times 10^{-2}$ | $9.35 \times 10^{-2}$ |

$W^+$ | $W^*$ | $W^*$ |
|-------|-------|-------|
| Xe$^{50+}$ | $4.78 \times 10^{-3}$ | $4.98 \times 10^{-3}$ | $5.20 \times 10^{-3}$ | $5.40 \times 10^{-3}$ |
| U$^{88+}$ | $8.08 \times 10^{-2}$ | $8.11 \times 10^{-2}$ | $8.18 \times 10^{-2}$ | $8.20 \times 10^{-2}$ |

$W_{\text{no-exc}}$ | Ref. [40] | Ref. [23] |
|------------------|----------|----------|
| Xe$^{50+}$ | $5.30 \times 10^{-3}$ | $3.4 \times 10^{-2}$ | $5.2 \times 10^{-2}$ |
| U$^{88+}$ | $8.31 \times 10^{-2}$ | $2.6 \times 10^{-1}$ | $4.9 \times 10^{-1}$ |

$a$ Extension of Ref. [40] having the energy splitting $3^3P_0 - 3^3P_1$ into account.
different local-exchange potentials in the states. The case without these effective exchange potentials ($W_{\text{no-exc}}$) results in a gauge invariance of $10^{-10}$%.

Differences between the values of the decay rate with and without $k_1$-optimization in Table 1 are mostly due to the respective transition energies, for which the decay rate depends quadratically, as well on the different $^3P_1$ and $^1P_1$ energies. These values can also be compared with the case of not considering the effective exchange potential of Eq. 1. Differences of up to 20% and 16% in xenon and uranium, respectively, shows how sensitive the decay of this transition is to the electronic correlation, in particular to the non-local part of the electron-electron interaction.

Residual differences of $0.5 - 2$ eV between MCDF energy values and those of Ref. 40 results in relative differences of 8% in the decay rate. Most of the experimental observations 26, 39, 50 and theoretical calculations 40, 51 - 54 of these energies are included in a energy range of 2 eV, resulting in differences up to 10%.

To be conservative, we consider the uncertainty in the decay rate as the combined uncertainty of the previous effects. The final result of the E1M1 decay rate is thus equal to $(5 \pm 1) \times 10^{-3}$ s$^{-1}$ and $(8 \pm 1) \times 10^{-2}$ s$^{-1}$ for xenon and uranium, respectively.

In contrast to previous studies of the negative continuum, where it shows that its contribution has to be included in relativistic calculations of two-photon decay rates 43, 44, the present case is of order of few percent, even for berylliumlike uranium ions. Following the semirelativistic approach of Ref. 48, the estimation of the negative-continuum contribution to the decay is proportional to $\omega_k^2/Z^2$. Previous studies deal with transitions between principal quantum numbers (e.g. 34), for which the transitions energies scales as $Z^4$. In the present case, the transition addresses the same quantum number and scales roughly as $Z$. This makes a smaller contribution of the negative energy continuum.

We notice evident differences relative to previous calculations by factors from 10 to 300. The differences can be attributed to our full relativistic approach in a $jj$-coupling scheme. This can be further investigated in the differential decay rate that is illustrated in Fig. 3, where it is shown normalized values (to the integral) of Eq. 1. Here, values for Xe$^{50+}$ and U$^{88+}$ ions obtained in this work and by Ref. 39 are displayed, which show evident differences in the differential decay rate.

The values of Refs. 39, 40 where obtained by considering only the $2s2p^1P_1$ and $2s2p^3P_1$ states in the intermediate-state summation and were calculated in a non-relativistic $LS$-coupling framework. Moreover, the non-relativistic form of the electric and magnetic dipole operators was also employed in Refs. 39, 40, which forbids intercombination transitions with a spin-flip of the total spin in a $LS$-coupling. Therefore, spin-orbit and spin-spin interactions were included in first approximation in order to mix the $^3P_1$ and $^1P_1$ terms. For highly charged ions, intercombination transitions are allowed in a $jj$-coupling scheme with relativistic wavefunctions, as the spin-orbit interaction is already included non-perturbately. Other investigations of the E1E1 have shown that relativistic effects increase the decay rate by 30% 12, 55 in heliumlike Xe. In the present case, the $M_1$ mode is even more sensitive to the $LS$-coupling scheme that is not appropriate for highly charged ions, where the strong spin-orbit interaction is included perturbately. A similar factor of 300 was already obtained in a relativistic calculation 56.

IV. CONCLUSION

We have presented the results of the two-photon forbidden $E1M1$ decay rate for two selected heavy elements obtained with an effective potential. The limitations of the active electron model for this particular decay is investigated and found that while this approach cannot be applied to low- and middle-Z ions, for berylliumlike Xe and heavier elements, each state is well described by a single configuration with orthogonal orbitals. Therefore, excitations of the spectator electron that forbids the use of this model can be neglected. We have found a negligible contribution of negative-energy states to this decay rate, which is in agreement with semirelativistic estimations. On the other hand, we observe significant relativistic effects relative to non-relativistic calculations performed for middle-Z ions, which can be attributed to the fact that the $LS$-coupling scheme is not appropriate for the evaluation of this decay rate in highly charged ions.

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