Theory of the multiphoton cascade transitions with two photon links: comparison of quantum electrodynamical and quantum mechanical approaches

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Abstract
The theory of multiphoton cascade transitions with two-photon links is considered within two different approaches: quantum electrodynamical (QED) and phenomenological quantum mechanical (QM). A problem of regularization of the cascade contributions is investigated in detail. It is argued that the correct regularization should include both initial and intermediate level widths in the singular energy denominators. This result follows both from the QED and from the QM approach. Particular transitions $nl \rightarrow 1s + 2\gamma$ with $nl = 3s$, $4s$, $3d$, $4d$ and $nl \rightarrow 1s + 3\gamma$ with $nl = 3p$, $4p$ are considered as examples. The importance of the proper cascade regularization is also demonstrated.

Keywords: hydrogen, multiphoton processes, cascade emission, quantum electrodynamics, quantum mechanics

1. Introduction
The theory of multiphoton transitions in atoms on the basis of quantum mechanics (QM) started with the work by Göppert-Mayer [1]. The first evaluation of the two-photon decay rate $2s \rightarrow 1s + 2\gamma(E1)$ in hydrogen was performed by Breit and Teller [2] (see correction to this work in [3]). Accurate nonrelativistic evaluation of the $2s \rightarrow 1s + 2\gamma(E1)$ transition rate in hydrogen was performed in [4]. The first fully relativistic calculation of this transition applicable also to the highly charged H-like ions (H-like HCI) was given in [5] and later in [6–8]. Quantum electrodynamical (QED) corrections to the two-photon decay of H-like ions were studied in [9, 10]. The calculations of different two-photon transition rates in a hydrogen atom and H-like ions were given in [11–13] in the nonrelativistic approximation and in a fully relativistic approach in [14–18].

Accurate evaluations of the two-photon transition rates in hydrogen are important for several reasons. First, during the last decades the two-photon $2s \rightarrow 1s$ transition frequency measurements reached an exclusively high level of accuracy [19–21] and the accurate measurements of the two-photon $3s \rightarrow 1s$, $3d \rightarrow 1s$ frequency transitions were also reported [22, 23]. Theoretical studies of the accuracy limits for these measurements require the knowledge of the corresponding transition rates [24]. Second, the two-photon transition rates in hydrogen are important for astrophysics, since these processes were involved in the formation of the cosmic microwave background (CMB) during the epoch of the cosmological hydrogen recombination. Recent accurate measurements of the properties of CMB also require the accurate knowledge of the two-photon $2s \rightarrow 1s$, $3s \rightarrow 1s$, $3d \rightarrow 1s$ etc transition rates in hydrogen. For this purpose the two-photon transition rates in hydrogen were evaluated in [25–28].

There is an essential difference in the description of the two-photon $2s \rightarrow 1s$ and the two-photon transitions to the ground state from the higher levels. This difference is due to the presence of the cascade contributions which are absent...
only in the \(2s \rightarrow 1s\) transition. The cascade problem in the two-photon transitions in two-electron HCI was first discussed in [29], The same problem was considered later in [30]. In [31] (see also [32]) a general QED approach was developed which allows for the description of cascade transitions. In the theory of cascade transition rates, an important question is the regularization of the singular cascade contributions to the total two-photon transition rate. This regularization is performed by the introduction of the level widths in the energy denominators which otherwise turn to zero for the photon frequencies, corresponding to the cascade resonances. In the QM phenomenological approach only the width of the intermediate state is usually employed for regularization [12, 13, 27]. We call this approach phenomenological since the intermediate state is usually employed for regularization of the energy denominators which otherwise turn to zero for the frequency distributions for the two-photon decays are converted with some other functions. This also violates the equivalence of the total transition probability is not the equivalence of the total cascade contribution from the 'pure two-photon' contributions. Second, in the different applications of the interference terms between the cascade and 'pure two-photon' contributions. Hence, this is not the case for two reasons. First, this ambiguity of this separation was demonstrated in [34]. The separation of the cascade contribution from the 'pure two-photon' contribution in equation (2) was discussed in several papers: in [31] for the two-electron HCI and in [34, 37–39] for the two-photon transitions in hydrogen. An ambiguity of this separation was demonstrated in [34]. The cascade contribution can be presented as the sum of the contributions of two cascade links (resonances):

\[
dW_{2s-2p-1s}^{2y}(\text{cascade}) = dW_{2s-2p-1s}^{2y}(\text{resonance 1}) + dW_{2s-2p-1s}^{2y}(\text{resonance 2}),
\]

where two resonant frequencies are: \(\omega_{\text{res1}} = E_{3s} - E_{2p}\) and \(\omega_{\text{res2}} = E_{2p} - E_{1s}\). The corresponding resonance contributions were presented in [34] on the basis of the QED approach

\[
dW_{2s-2p-1s}^{2y}(\text{resonance 1}) = \frac{\Gamma_{3s} + \Gamma_{2p}}{\Gamma_{2p}} \times W_{3s-2p}^{2y}(\omega_{\text{res1}}) W_{2p-1s}^{2y}(\omega_{\text{res2}}) \, d\omega,
\]

\[
dW_{2s-2p-1s}^{2y}(\text{resonance 2}) = \frac{\Gamma_{3s} + \Gamma_{2p}}{\Gamma_{2p}} \times W_{3s-2p}^{2y}(\omega_{\text{res2}}) W_{2p-1s}^{2y}(\omega_{\text{res2}}) \, d\omega.
\]

Note that the factor \(\frac{\Gamma_{3s} + \Gamma_{2p}}{\Gamma_{2p}}\) in equation (4) was lost in [34] though this led to a relatively small numerical error since \(\Gamma_{3s} \approx 0.01 \Gamma_{2p}\).

Here \(\Gamma_{3s}, \Gamma_{2p}\) are the total widths of the levels 3s, 2p and \(W_{3s-2p}, W_{2p-1s}\) are the one-photon transition rates. In the nonrelativistic limit (i.e. retaining only \(E1\) transitions) \(\Gamma_{3s} = W_{3s-2p}, \Gamma_{2p} = W_{2p-1s}\). Then, integrating equations (4), (5) over \(\omega\) and taking into account equation (1) we find

\[
\frac{1}{2} \int_{0}^{\infty} dW_{3s-2p-1s}^{2y}(\text{resonance 1}) = \frac{1}{2} W_{3s-2p}^{1y} = \frac{1}{2} \Gamma_{3s},
\]

\[
\frac{1}{2} \int_{0}^{\infty} dW_{3s-2p-1s}^{2y}(\text{resonance 2}) = \frac{1}{2} W_{3s-2p}^{1y} = \frac{1}{2} \Gamma_{3s}.
\]

Hence,

\[
W_{3s-1s}^{2y}(\text{cascade}) = \Gamma_{3s},
\]

and

\[
W_{3s-1s}^{2y} = \Gamma_{3s} + \frac{1}{2} \int_{0}^{\infty} \left[ dW_{3s-1s}^{2y}(\text{pure}) + dW_{3s-1s}^{2y}(\text{interference}) \right].
\]

Within the QM approach the regularization was usually performed by introducing the widths for the intermediate \(np\) states, i.e. replacing the energy \(E_{np}\) by \(E_{np} - \frac{i}{2}\Gamma_{np}\). In the case of \(3s - 2p - 1s\) cascade \(\Gamma_{np} = \Gamma_{2p}\). Therefore, the contribution of resonance 1 instead of equation (4) looked like

\[
dW_{3s-2p-1s}^{2y}(\text{resonance 1}) = \frac{W_{3s-2p}^{1y}(\omega_{\text{res1}}) W_{2p-1s}^{2y}(\omega_{\text{res2}})}{\Gamma_{2p}} \, d\omega,
\]

while the contribution of \(dW_{3s-2p-1s}^{2y}(\text{resonance 2})\) remained the same as in equation (5).

We have to stress that the insertion of equation (10) into the integral, equation (1), gives exactly the same result, equation (6), as the insertion of equation (4). One could think that both methods of regularizations are equivalent. However, this is not the case for two reasons. First, this equivalence for the cascade contributions to the integral value of the transition probability is not the equivalence of the total frequency distributions to the same integral due to the existence of the interference terms between the cascade and 'pure two-photon' contributions. Second, in the different applications the frequency distributions for the two-photon decays are converted with some other functions. This also violates the equivalence mentioned above (see one example in section 4).
In the second part of the paper we address the same problem of the cascade regularization within the QM approach following mainly [40]. In [40] the derivation is given for one-photon transitions between two excited atomic states, which corresponds to one ‘link’ of the two-photon cascade transition. We extend this derivation to the full two-photon transition rate, including cascade, ‘pure’ two-photon and interference contributions. We demonstrate that with the proper treatment of the QM approach we arrive again at the expression, equation (4), and not equation (10)) for the cascade contribution in case of the $3s \rightarrow 1s + 2\nu$ transition. The QM approach is then generalized also to the three-photon transitions. In all cases, the QM approach gives exactly the same results as the QED theory.

The paper is organized as follows. Section 2 is devoted to the application of the QED approach to the cascade regularization problem. In section 2.1 we start with the QED derivation of the Lorentz profile for the one-photon transition from the excited state to the ground state. This derivation repeats shortly the derivations in [31, 32] but is necessary to introduce the basic formulas and notations. As an example the Lyman-alpha transition $2p - 1s$ is considered. In section 2.2 the two-photon transition rate to the ground state from the $ns$-state in the presence of cascades is described in general. In section 2.3 the regularization of the two-photon cascade transition $3s - 1s$ is analysed. In section 2.4 the same derivations are made for the two-photon $4s - 1s$ transition: there is an important difference between $4s - 1s$ and $3s - 1s$ two-photon transitions due to the existence of several cascade channels in the case of $4s - 1s$. In section 2.5 the three-photon transitions are analysed in the ‘two-photon approximation’ with three-photon decay $3p - 1s$ as an example. In section 3 the QM approach is applied to the two- and three-photon transitions. In section 4 the importance of the cascade contributions to the multiphoton transition rates.

2. QED approach for the two- and three-photon decays

The full QED description of any process in an atom should start with the ground state and end up with the ground state too, i.e. the excitation of the decaying state should always be included. For the resonant processes, e.g. for the resonant photon scattering, the absorption part of the process can be well separated from the emission part, so that the description of the decay process independent of the excitation becomes possible. In this way the theory of multiphoton processes in atoms was developed in [31, 32].

In the present paper our aim is to highlight the origin of the arrival of the initial state width in the regularization scheme for the cascade contributions to the multiphoton transition rates.

2.1. One-photon transition to the ground state

Having in mind the recombination processes in the hydrogen atom, we consider first the resonance photon scattering on the ground $1s$ state with resonances corresponding to the $np$ states. In our derivations we will fully neglect the photons other than $E1$ which is reasonable for the neutral hydrogen. It is important to stress that we consider the free atoms which are excited by the photons released by the source whose line width is comparable to (or larger than) the natural line width of the resonance atomic state. Thus we exclude the special cases of the excitation by the laser with the narrow bandwidths or something equivalent. Our condition (broad source width) should correspond to the cosmological recombination situation when every atom is excited by the photons emitted by another atom. The Feynman graph corresponding to the resonant photon scattering is depicted in figure 1(a). A scattering amplitude presented by figure 1(a) is [40, 41]

$$S_{1s}^{(2)nc} = -2\pi \delta(\omega_f - \omega_i)U_{1s}^{(2)nc},$$

$$U_{1s}^{(2)nc} = e^2 \sum_n \left( \gamma_{\mu} A_{\mu}^{(k, \vec{e})} \right)_{1s} \left( \gamma_{\mu} A_{\mu}^{(k, \vec{e})} \right)_{np, 1s} \alpha_f + E_{np} - E_{1s} = E_n,$$

where $E_n$ are the Dirac energies, $\gamma_\mu$ are the Dirac matrices, $A_\mu^{(k, \vec{e})}$ is the electromagnetic field vector potential, $\vec{e}$ is the polarization vector, $\vec{k}$ is the wave vector, $\omega = |\vec{k}|$ is the photon frequency, indices i, f denote the initial (absorbed) and final (emitted) photons. Summation in equation (12) extends over the entire Dirac spectrum. The energy conservation law implies that $|\vec{k}_{ij}| = |\vec{k}_f|$. For the resonant scattering process the photon frequency $\omega_i = \omega_f$ is close to the energy difference between two atomic levels. In the case of $np$ resonance $\omega_i \simeq E_{np} - E_{1s}$. Accordingly we have to retain only one term in the sum over $n$ in equation (12)

$$U_{1s}^{(2)nc} = e^2 \left( \gamma_{\mu} A_{\mu}^{(k, \vec{e})} \right)_{1s} \left( \gamma_{\mu} A_{\mu}^{(k, \vec{e})} \right)_{np, 1s} \alpha_f + E_{1s} - E_{np}.$$

Equation (13) reveals that in the resonance approximation the scattering amplitude is factorized into emission and
absorption parts. The energy denominator should be attached to the emission or absorption part depending on what we want to describe: the emission or absorption process. In particular, the first-order emission amplitude can be expressed as

$$U_{\text{em}}^{n\rightarrow 1s} = e^{-i\int_{E_{\text{em}}}^{E_{\text{em}}+E_{1s}} \frac{d\omega}{\omega_f + E_{1s} - E_{\text{em}}} \Gamma_{\text{em}}},$$ \hspace{1cm} (14)$$

The nonresonant corrections to the resonance approximation, first introduced in \[33\] were recently investigated in \[42-44\]. The role of these corrections appeared to be negligible in most cases. These corrections arise when one takes into account the terms other than the resonant ones in the sum over \(n\) in equation (12). Since in our present studies we are not aiming at the highly accurate determination of the resonant frequencies, these corrections can also be neglected.

The energy conservation law which follows from equation (11) reads \(\omega_n = \omega_f\). One can write the resonance condition in the form:

$$|\omega_n - E_{\text{em}} + E_{1s}| = |\omega_f - E_{\text{em}} + E_{1s}| \leq \Gamma_{\text{em}}.$$ \hspace{1cm} (15)$$

In the cases where we can neglect \(\Gamma_{\text{em}}\) in equation (15), this equation takes the form of the energy conservation law

$$\omega_f = E_{\text{em}} = E_{1s}.$$ \hspace{1cm} (16)$$

In particular we can use equation (16) in the numerator of equation (14) but not in its denominator.

To derive the Lorentz profile for the emission process we follow the low procedure \[35\], i.e. insert an infinite number of self-energy corrections in the resonance approximation into the electron propagator in figure 1(a). Repeating these insertions in the resonance approximation leads to a geometric progression. Summation of this progression leads to the arrival of the level width \(\Gamma_{\text{em}}\) in the energy denominator in equation (13) \[32\]

$$U_{\text{em}}^{n\rightarrow 1s} = e^{-i\int_{E_{\text{em}}}^{E_{\text{em}}+E_{1s}} \frac{d\omega}{\omega_f + E_{1s} - E_{\text{em}} + \frac{1}{3}\Gamma_{\text{em}}}}.$$ \hspace{1cm} (17)$$

As a next step one has to take the amplitude, equation (17), by square modulus, then integrate over the photon emission directions \(\vec{v}_f\) and sum over the photon polarizations. Then we obtain the Lorentz profile for the absolute probability of the photon emission with the frequency in an interval between \(\omega_f\) and \(\omega_f + d\omega_f\)

$$d\omega_{n\rightarrow 1s}(\omega_f) = \frac{1}{2\pi} \left(\frac{W_{1s}}{\omega_f + E_{1s} - E_{\text{em}} - \frac{L_{\text{em}}^2}{\Gamma_{\text{em}}^2}}\right)^2,$$ \hspace{1cm} (18)$$

where \(W_{1s}\) is the one-photon transition rate for the transition \(n\rightarrow 1s\). The Lorentz profile is normalized to the branching ratio for the transition \(n\rightarrow 1s\):

$$\int_{-\infty}^{\infty} d\omega_{n\rightarrow 1s} = \frac{W_{1s}}{\Gamma_{1s}} = b_{1s},$$ \hspace{1cm} (19)$$

Figure 2. Feynman graph describing the two-photon resonant scattering on the ground state of the hydrogen atom, with the excitation of the \(n\rightarrow s\) state and the resonance condition \(\omega_1 + \omega_2 = E_s - E_{1s}\). In (a) the basic process of the resonant scattering with the excitation of the \(n\rightarrow s\) state is depicted. In (b) the electron self-energy insertion in the central electron propagator is made. The notations are the same as in figure 1.

2.2. Two-photon \(n\rightarrow 1s\) transition

In this section we describe the two-photon transition to the ground state using \(n\rightarrow 1s\) two-photon transitions as an example. According to our approach we have to consider the Feynman graph depicted in figure 2(a). The two-photon resonant excitation is the most natural and convenient way to describe the excitation process in this case. The resonance condition is

$$\omega_1 + \omega_2 = \omega_0 = E_s - E_{1s}.$$ \hspace{1cm} (20)$$

A scattering amplitude corresponding to figure 2(a) looks like

$$U_{1s}^{\text{sc}} = e^{i\int_{E_{1s}}^{E_{1s}+E_s} \frac{d\omega}{\omega_f + E_{1s} - E_n} (\omega_f + \omega_f + E_{1s} - E_n) \times (\omega_f + \omega_f + E_{1s} - E_n)} \Gamma_{1s}.$$ \hspace{1cm} (21)$$

The energy conservation law in this process is implemented by the condition \(\omega_f + \omega_f = \omega_1 + \omega_2\) and the resonance condition is given by equation (20). From equation (20) follows the approximate energy conservation law similar to equation (21)

$$|\omega_f + \omega_f - E_s + E_{1s}| \leq \Gamma_{1s},$$ \hspace{1cm} (22)$$

which can be replaced by an equation similar to equation (16)

$$\omega_f + \omega_f = E_s - E_{1s},$$ \hspace{1cm} (23)$$

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when \( \Gamma_m \) can be neglected. According to equation (20) and the conservation law, the last energy denominator in equation (21) can be replaced by
\[
\omega_{f_1} + \omega_{f_2} - \omega_{i} + E_{1s} - E_{ns} = \omega_i + E_{1s} - E_{ns},
\]
i.e. does not depend on the frequencies of the emitted photons.

In the resonance approximation we retain only one term \( n_2 = ns \) in the sum over \( n_2 \). For \( n > 2 \) there is always a leading one-photon contribution to the total width \( \Gamma_m \), for example the \( W_{1s-2p}^{ly} \) transition rate in the case \( n = 3 \). Assuming the existence of such a contribution, we will continue our studies by inserting the one-loop electron self-energy corrections to the central propagator in figure 1(a) (the low procedure).

After summation of the Low sequence, the emission amplitude for the two-photon decay process \( ns \to 1s + 2\gamma \) looks like
\[
U^\text{em}_{ns-1s} = e^2 \sum_{n_1} \frac{(\gamma_{\mu_1} A_{\nu_1}^{\ast}(\vec{k}_{f_1}, \vec{\epsilon}_{f_1}))(\gamma_{\mu_2} A_{\nu_2}^{\ast}(\vec{k}_{f_2}, \vec{\epsilon}_{f_2}))(\gamma_{\mu_3} A_{\nu_3}^{\ast}(\vec{k}_{i}, \vec{\epsilon}_{i}))(\gamma_{\mu_4} A_{\nu_4}(\vec{k}_{i}, \vec{\epsilon}_{i}))}{(\omega_{f_1} + E_{1s} - E_{ns})(\omega_{f_1} + \omega_{f_2} + E_{1s} - E_{ns} + \frac{i}{2}\Gamma_m)}.
\]
(25)
The value \( \Gamma_m \) is defined in a different way for the different \( ns \) states. For example, \( \Gamma_3s = W_{1s-2p}^{ly} \) since there are no other one-photon decay channels for the 3s level. The further investigations of the two-photon transition probabilities should be performed separately for different \( n \). In the next section, we will continue these investigations for the 3s \( \to 1s + 2\gamma \) transition.

### 2.3. Two-photon 3s \( \to 1s \) transition

For further studies of the 3s \( \to 1s \) transition, we can start with the expression for the emission amplitude, equation (25), written for the case \( ns = 3s \). The Feynman graphs for the resonance two-photon scattering with the excitation of the 3s level are depicted in figure 3. To the expression, equation (25), we have to add another term corresponding to the Feynman graph figure 3(a) with the interchanged positions of the \( \vec{k}_{f_1}, \vec{\epsilon}_{f_1} \) and \( \vec{k}_{f_2}, \vec{\epsilon}_{f_2} \) photons.

For the 3s \( \to 1s \) two-photon transition only one cascade is possible: 3s \( \to 2p \to 1s \). Accordingly, the two new resonance conditions arise (these resonances were defined in section 1):
\[
\omega^{\text{res.1}} = E_{3s} - E_{2p},
\]
\[
\omega^{\text{res.2}} = E_{2p} - E_{1s}.
\]
To consider the cascade contribution to 3s \( \to 1s \) we have to set \( n_1 = 2p \) in equation (25). Applying the low procedure to the upper electron propagator in figure 3(b) we find
\[
U^\text{em, cascade}_{3s-2p-1s} = e^2 \left\{ \frac{(\gamma_{\mu_1} A_{\nu_1}^{\ast}(\vec{k}_{f_1}, \vec{\epsilon}_{f_1}))_{12p}(\gamma_{\mu_2} A_{\nu_2}^{\ast}(\vec{k}_{f_2}, \vec{\epsilon}_{f_2}))_{2p3s}}{\omega_{f_1} + E_{1s} - E_{2p} + \frac{i}{2}\Gamma_{2p}} \right. + \left. \frac{(\gamma_{\mu_2} A_{\nu_2}^{\ast}(\vec{k}_{f_2}, \vec{\epsilon}_{f_2}))_{12p}(\gamma_{\mu_1} A_{\nu_1}^{\ast}(\vec{k}_{i}, \vec{\epsilon}_{i}))_{2p3s}}{\omega_{f_1} + E_{1s} - E_{2p} + \frac{i}{2}\Gamma_{2p}} \right\} \times \frac{1}{\omega_{f_2} + \omega_{f_1} + E_{1s} - E_{3s} + \frac{i}{2}\Gamma_{3s}}.
\]
(27)

The first term in the curly brackets describes the resonance \( \omega^{\text{res.1}} \), the second term describes the resonance \( \omega^{\text{res.2}} \).

Now we take \( U^\text{em, cascade}_{3s-2p-1s} \) by square modulus, integrate over the directions of the emitted photons and sum over the polarizations of both photons. Consider first the square modulus of the first term in the curly brackets and the factor outside the curly brackets in equation (27). This term is represented by figure 3(a) and corresponds to the contribution of resonance 1 in equation (26). In this case we are interested to derive the Lorentz line profile for the upper link of the cascade 3s \( \to 2p \to 1s \). Therefore we have to integrate first over the frequency of the second emitted photon, i.e. \( \omega_{f_2} \). In principle, the integration over both photon frequencies should be done with equation (22) taken into account, i.e.
\[
\int_0^{\omega_0} d\omega_{f_1} \int_0^{\omega_0} d\omega_{f_2} = \frac{1}{2} \int_0^{\omega_0} d\omega_{f_1} \int_0^{\omega_0} d\omega_{f_2},
\]
(28)
where \( \omega_0 = E_{3s} - E_{1s} \). Equation (28) holds due to the symmetry of equation (27) with respect to permutation \( \omega_{f_1} \leftrightarrow \omega_{f_2} \).

Here, the integration over the frequency \( \omega_{f_2} \) in equation (27) we perform in the complex plane. Since only the pole terms contribute we can extend the interval of integration to \((-\infty, +\infty)\) and not to refer to equation (22). Then using the Cauchy theorem after some algebraic transformation we obtain the cascade contribution (resonance 1) to the differential branching ratio
\[
\mathcal{B}_{2\gamma(\text{resonance 1})}(\omega) = \frac{1}{2\pi} \times \frac{\Gamma_3s + \Gamma_{2p}}{\Gamma_3s + \Gamma_{2p}} \frac{W_{1s-2p}^{ly}(\omega^{\text{res.1}})W_{2p-1s}^{ly}(\omega^{\text{res.2}}) d\omega}{(\omega - \omega^{\text{res.1}})^2 + \frac{1}{4}(\Gamma_3s + \Gamma_{2p})^2}.
\]
(29)
The differential branching ratio \( d^2\gamma \) is connected with the differential transition rate \( dW_{\gamma} \) via
\[
d^2\gamma = \frac{dW_{\gamma}}{\Gamma_{\gamma}}.
\]

This definition concerns not only the cascade contributions but all of the contributions in equation (2) for the two-photon decay. Combining now the formulas (29), (30) we arrive at the expression (4) presented in the introduction. The integration of equation (30) over the remaining frequency will give the total branching ratio
\[
\mathcal{K}_{\gamma} = \frac{W_{\gamma}}{\Gamma_{\gamma}}.
\]

Note that this last integration according to equation (28) should be done within the interval \((0, \omega_0)\) since now no pole approximation can be used. In the same way we can obtain the expression (5) presented in the introduction.

To consider the ‘pure two-photon’ contribution to the transition amplitude \(3s \rightarrow 1s + 2\gamma\) we have to return to equation (25) and consider this expression with the state \(2p\) excluded from the summation over \(n_1\). The words ‘pure two-photon’ should not be understood literally: we have to remember that the exact separation of this ‘pure two-photon’ contribution is not possible [34]. This exclusion corresponds to the ‘pole approximation’ employed in section 2.3 for the description of the cascade contribution: extension of the first frequency integration over the interval \((−\infty, \infty)\). In [34] the more general approach was developed, when the resonances were regularized only within the ‘windows’ of the different breadth. Then the \(2p\) state should be eliminated from the sum over \(n_1\) only within ‘windows’. The ‘pole approximation’ corresponds to the window breadth \([0] \approx \infty\).

Since the energy denominators now become nonsingular we can employ the energy conservation law to replace the frequency \(\omega_{i}\) in the second denominator by \(\omega_{ij} = \omega_0 - \omega_{i}\).

Then taking equation (25) with the excluded term \(n_i = 2p\) by square modulus, integrating over the directions of the emitted photons, summing over the polarizations and integrating over \(\omega_{i}\) we obtain the ‘pure two-photon’ contribution to the differential transition rate \(3s \rightarrow 1s + 2\gamma\).

In a similar way, the interference between the cascade and the ‘pure two-photon’ parts of the amplitude, equation (25), can be evaluated.

### 2.4. Two-photon \(4s - 1s\) transition

Repeating the derivations for the \(3s - 1s\) transition for the case of the \(4s - 1s\) transition, we present the contributions for the two cascades: \(4s - 2p - 1s\) and \(4s - 3p - 1s\): (1) contribution from the upper link \(4s - 2p\) of the cascade \(4s - 2p - 1s\), (2) contribution of the lower link \(2p - 1s\) of the cascade \(4s - 2p - 1s\), (3) contribution from the upper link \(4s - 3p - 1s\), (4) contribution of the lower link \(3p - 1s\) of the cascade \(4s - 3p - 1s\), where the resonance frequencies are \(\omega_{0(1)} = E_{4s} - E_{2p}\), \(\omega_{0(2)} = E_{3p} - E_{1s}\) for the cascade \(4s - 2p - 1s\) and \(\omega_{0(3)} = E_{4s} - E_{2p}\), \(\omega_{0(4)} = E_{3p} - E_{1s}\) for the cascade \(4s - 3p - 1s\). This results
\[
W_{2\gamma(\text{cascade})} = \frac{1}{2} \int_{0}^{\infty} \sum_{i=1}^{4} dW_{2\gamma}\text{ (resonance }i)\]

\[
= W_{4s-2p}^{1/2} + \frac{W_{4s-2p-1s}^{1/2}W_{4s-3p}^{1/2}}{\Gamma_{3p}} = W_{4s-2p}^{1/2} + b_{4s-2p-1s}^{1/2}W_{4s-3p}^{1/2},
\]

where \(\omega_0 = E_{4s} - E_{1s}\), \(b_{4s-2p-1s}^{1/2}\) is the branching ratio for the transition \(3p - 1s\). We took into account that \(\Gamma_{3p} = W_{4s-2p}^{1/2} + W_{4s-2p-1s}^{1/2}\). Hence \(W_{2\gamma(\text{cascade})} \neq \Gamma_{4s}\), where \(\Gamma_{4s} = W_{4s-2p}^{1/2} + W_{4s-3p}^{1/2}\), unlike equation (8) in the case of the \(3s - 1s\) transition.

### 2.5. Three-photon \(3p \rightarrow 1s + 3\gamma\) transition

In this section we consider the three-photon transitions, taking as an example the \(3p \rightarrow 1s + 3\gamma\) transition. The main decay channels of the \(3p\) level are \(3p \rightarrow 1s + \gamma\) and \(3p \rightarrow 2s + \gamma\). Therefore as a resonance scattering process in this case we can choose the one-photon absorption and three-photon emission process depicted in figure 4. For the transition \(3p \rightarrow 1s + 3\gamma\) there are two cascades, containing two-photon links: \(3p \rightarrow 2p + 2\gamma \rightarrow 1s + 3\gamma\) and \(3p \rightarrow 2s + \gamma \rightarrow 1s + 3\gamma\). In the case of the three-photon transition we will take into account only the cascade contribution. This cascade contribution contains necessarily one ‘pure two-photon’ link.
(3p → 2p or 2s → 1s) and is, therefore, of the same order of magnitude as the ‘pure two-photon’ contribution to the 3s → 1s transition. The ‘pure three-photon’ transitions and the corresponding interference terms are essentially smaller than the cascade contributions and can be neglected in the ‘two-photon’ approximation [34]. In this sense the situation differs from the situation in 3s → 1s two-photon decay when we were interested in the ‘pure two-photon’ and interference terms.

The derivation similar to the two-photon case gives the following expression for the three-photon emission amplitude 3p → 1s in the resonance approximation:

\[
U_{3p \rightarrow 1s}^{\text{em.,3y}} = e^3 \sum_{n1n2} \frac{1}{(E_{1s} - E_{n1} + \omega f_1)(E_{1s} - E_{n2} + \omega f_2 + \omega f_3)} \times \left( \gamma^{\mu}_{\pi 1} A_{\pi 1}^{\mu \nu \rho} (\tilde{e}_{12}^{\nu \rho} \gamma^\mu)_{n1n2} (\gamma^{\mu}_{\pi 2} A_{\pi 2}^{\mu \nu \rho} (\tilde{e}_{12}^{\nu \rho} \gamma^\mu)_{n2} \right)_{3p}.
\]

(33)

In equation (33) we summed already all of the self-energy insertions in the lower electron propagator, as shown in figure 4. An exact energy conservation law in the case of the process, figure 4 is \( \omega = \omega f_1 + \omega f_2 + \omega f_3 \). The resonance condition and the approximate energy conservation law in the case of three-photon decay looks like

\[ |\omega_0 - (E_{3p} - E_{1s})| = |\omega f_1 + \omega f_2 + \omega f_3 - (E_{3p} - E_{1s})| \leq \Gamma_{3p}. \]

(34)

To fix the cascade 3p → 2s + γ → 1s + 3γ contribution we set \( n_2 = 2s \) in equation (33). This results

\[
U_{3p \rightarrow 1s}^{\text{em.,3y}} = e^3 \frac{1}{E_{1s} - E_{3p} + \omega f_1 + \omega f_2 + \omega f_3 + \frac{1}{2} \Gamma_{3p}} \times (\gamma^{\mu}_{\pi 1} A_{\pi 1}^{\mu \nu \rho} (\tilde{e}_{12}^{\nu \rho} \gamma^\mu)_{3p2s}) \times (E_{1s} - E_{2s} + \omega f_1 + \omega f_2 + \frac{1}{2} \Gamma_{2s}) \times \sum_{n1} \left( \gamma^{\mu}_{\pi 2} A_{\pi 2}^{\mu \nu \rho} (\tilde{e}_{12}^{\nu \rho} \gamma^\mu)_{n11s} \right) + \text{perm}. \]

(35)

In equation (35) we should include the contribution of the Feynman graphs with all the permutations of the photon lines (perm).

Now we have to take the right-hand side of equation (35) by square modulus, to integrate over the emitted photon directions and to sum over the photon polarizations. Then we have to integrate over the photon frequencies \( \omega f_1, \omega f_2, \omega f_3 \) taking into account the condition, equation (34). However when we integrate the contribution of the cascade 3p → 2s → 1s we have to take into account that the frequency \( \omega f_3 \) is fixed by the resonance condition

\[ |\omega f_3 - (E_{3p} - E_{2s})| \leq \Gamma_{3p} + \Gamma_{2s}. \]

(36)

Inserting equation (36) into the approximate conservation law (34) we obtain the equality

\[ \omega f_3 + \omega f_3 = E_{2s} - E_{1s}. \]

(37)

An integration over \( \omega f_3, \omega f_3 \) should be performed in the following way:

\[
\int_0^{E_{2s} - E_{1s}} d\omega f_3 \int_0^{E_{2s} - E_{1s}} d\omega f_3 = \frac{1}{2} \int_0^{E_{2s} - E_{1s}} d\omega f_3 \int_0^{E_{2s} - E_{1s}} d\omega f_3. \]

(38)

Equation (38) holds after symmetrization of equation (36) via the permutation of the photons with frequencies \( \omega f_3, \omega f_3 \). The integration over two frequencies (e.g. over \( \omega f_3 \) and \( \omega f_3 \) in equation (35)) can be extended to the interval \(-\infty, +\infty\) since we use the pole approximation. The third integration over \( \omega f_3 \) in equation (35) according to equation (38) should be performed over the finite interval \([0, E_{2s} - E_{1s}]\). The integration yields

\[ b_{3p \rightarrow 1s}^{3\gamma}(3p \rightarrow 2s - 1s) = \frac{W_{3p \rightarrow 2s}^{1\gamma} W_{2s \rightarrow 1s}^{2\gamma}}{\Gamma_3 \Gamma_{2s}}. \]

(39)

where \( \Gamma_0 = E_{2s} - E_{1s} \).

The physical sense of the dimensionless quantity \( b_{3p \rightarrow 1s}^{3\gamma}(3p \rightarrow 2s - 1s) \) should be discussed specially. This quantity should define the 3γ transition rate 3p → 1s via the channel 3p → 2s + γ → 1s + 3γ. This transition rate is very small compared to the main decay channel for the 3p state, i.e. \( W_{3p \rightarrow 1s}^{1\gamma} \). We assume that the quantity \( b_{3p \rightarrow 1s}^{3\gamma}(3p \rightarrow 2s - 1s) \) is the branching ratio for the three-photon transition rate \( W_{3p \rightarrow 1s}^{3\gamma}(3p \rightarrow 2s - 1s) \) to the direct two-photon transition rate \( W_{2s \rightarrow 1s}^{2\gamma} \). Then from equation (39) it follows

\[ W_{3p \rightarrow 1s}^{3\gamma}(3p \rightarrow 2s - 1s) = \frac{W_{3p \rightarrow 2s}^{1\gamma} W_{2s \rightarrow 1s}^{2\gamma}}{\Gamma_3 \Gamma_{2s}}. \]

(40)

In the same way the contribution of the three-photon cascade 3p → 2p + γ → 1s + 3γ can be analysed. The final result looks like

\[ b_{3p \rightarrow 1s}^{3\gamma}(3p \rightarrow 2p - 1s) = \frac{W_{3p \rightarrow 1s}^{1\gamma} W_{2p \rightarrow 2s}^{2\gamma}}{\Gamma_3 \Gamma_{2p}}. \]

(41)

Unlike \( b_{3p \rightarrow 1s}^{3\gamma}(3p \rightarrow 2s - 1s) \) the quantity, equation (41), should be considered as the branching ratio of the transition rate via channel 3p → 2p → 1s to the total width of the 3p level, i.e. \( \Gamma_{3p} \). Then

\[ W_{3p \rightarrow 1s}^{3\gamma}(3p \rightarrow 2p - 1s) = \frac{W_{3p \rightarrow 1s}^{1\gamma} W_{2p \rightarrow 2s}^{2\gamma}}{\Gamma_2}. \]

(42)

In the equation in [35], corresponding to equation (42), the factor \( W_{3p \rightarrow 1s}^{1\gamma}/\Gamma_{2p} = 1 \) was omitted. Here we keep it to demonstrate that the transition channel 3p → 2p → 1s is a three-photon channel. The total expression for the transition rate 3p → 1s (one-photon and three-photon) is

\[ W_{3p \rightarrow 1s}^{1\gamma} + W_{3p \rightarrow 1s}^{3\gamma} = W_{3p \rightarrow 1s}^{1\gamma} + \frac{W_{3p \rightarrow 2s}^{1\gamma} W_{2s \rightarrow 1s}^{2\gamma}}{\Gamma_3 \Gamma_{2s}}. \]

(43)

This expression coincides with one derived in [35] up to the coefficients before the second and third terms in the right-hand side of equation (43). In [35] these coefficients were evaluated incorrectly and were equal 3/4.
3. QM approach for the two- and three-photon decays

In this section we will follow along the lines of the derivation for the one-photon transition given in [40]. The QM approach is based on the time dependent Schrödinger equation

$$i \frac{\partial \psi(t)}{\partial t} = (\hat{H}_0 + \hat{V}(t)) \psi(t), \quad (44)$$

where \( \hat{H}_0 \) is a time-independent zero-order atomic Hamiltonian and \( \hat{V}(t) \) is the perturbation describing the interaction of an atom with the photon field. According to the Dirac perturbation theory we search for the solution of equation (44) in the form of the expansion

$$\psi(t) = \sum_\nu a_\nu(t) e^{-i\epsilon_\nu t} \psi_\nu^{(0)}, \quad (45)$$

where \( \psi_\nu^{(0)} \) are the eigenfunctions of \( \hat{H}_0 \):

$$\hat{H}_0 \psi_\nu^{(0)} = E_\nu \psi_\nu^{(0)} \quad (46)$$

and \( \epsilon_\nu \) are the energies of the system atom+photon field. These energies can be presented as

$$\epsilon_\nu = E_\nu - \frac{i}{2} \Gamma_\nu + \sum \omega, \quad (47)$$

where \( E_\nu \) are the eigenstates of the Hamiltonian \( \hat{H}_0 \), \( \Gamma_\nu \) are the corresponding level widths and \( \sum \omega \) is the sum of the photons which arrives in the field, and when an atom arrives in the state \( \nu \).

In equation (45) we also need a second subscript, indicating to which stationary state \( \psi_\nu^{(0)} \) we apply the perturbation theory:

$$\psi_\mu(t) = \sum_\nu a_{\mu\nu}(t) e^{-i\epsilon_\nu t} \psi_\nu^{(0)}. \quad (48)$$

For the coefficients \( a_{\mu\nu}(t) \) from equation (44) follows the system of equations

$$i \frac{\partial a_{\mu\nu}(t)}{\partial t} = \sum_{\nu'} \hat{V}_{\nu'\nu} a_{\mu\nu}(t) e^{i(\epsilon_{\nu'} - \epsilon_\nu t)}, \quad (49)$$

where \( \hat{V}_{\nu'\nu} \) are the matrix elements of the operator \( \hat{V} \). In these matrix elements we understand this operator as time-independent, the time dependence of it being included in the exponent \( e^{-i\epsilon_\nu t} \). We are interested in the evaluation of quantities

$$\lim_{t \to \infty} |a_{\mu\nu}(t)|^2 = W_{\mu\nu}, \quad (50)$$

which can be interpreted as the transition probabilities from the state \( \mu \) to the state \( \nu \) of the atom+field system. To derive the two-photon transition probability we need to employ the second-order perturbation theory. In the zero-order approximation for the initial state \( \mu = i \) we have \( a_0^{(0)} = \delta_{i\nu} \), where \( \delta_{i\nu} \) is the Kronecker symbol. This zero-order value should be inserted then into the right-hand side of equation (49). Then the equation for the first-order correction \( a_{i\nu}^{(1)} \) results

$$i \frac{\partial a_{i\nu}^{(1)}(t)}{\partial t} = \hat{V}_{i\nu} e^{i(\epsilon_\nu - \epsilon_i) t}. \quad (51)$$

Integrating equation (51) we find

$$a_{i\nu}^{(1)} = \frac{\hat{V}_{i\nu} (1 - e^{i(\epsilon_\nu - \epsilon_i) t})}{\epsilon_\nu - \epsilon_i}. \quad (52)$$

Take into account that

$$\epsilon_i = E_i - \frac{i}{2} \Gamma_i, \quad \epsilon_\nu = E_\nu - \frac{i}{2} \Gamma_\nu + \omega, \quad (53)$$

where \( \nu \) is the intermediate state in the two-photon transition and \( \omega \) is the frequency of the photon emitted in the transition \( i \to \nu \). As the next step we have to insert equation (52) into the right-hand side of equation (49) and to set \( \nu = f \) (final state). This yields

$$i \frac{\partial a_{if}^{(2)}(t)}{\partial t} = \sum_\nu \hat{V}_{if} \hat{V}_{\nu f} \left[ 1 - e^{i(\epsilon_\nu - \epsilon_f) t} \right] e^{i(\epsilon_f - \epsilon_i) t}. \quad (54)$$

Here \( \epsilon_f = E_f - \frac{i}{2} \Gamma_f + \omega' \) with the condition \( \omega_1 + \omega_2 = E_i - E_f \); if \( f \) is the ground state \( \Gamma_f = 0 \).

Integrating equation (54) over time we find the second-order correction

$$a_{if}^{(2)} = \sum_\nu \hat{V}_{if} \hat{V}_{\nu f} \left( \frac{1 - e^{i(\epsilon_\nu - \epsilon_f) t}}{\epsilon_f - \epsilon_\nu} - \frac{1 - e^{i(\epsilon_f - \epsilon_i) t}}{\epsilon_f - \epsilon_i} \right). \quad (55)$$

In equation (55) we should set \( t \to \infty \) according to definition (50). Taking into account permutation symmetry we find finally equation for transition probability

$$dW_{if}^{(2)} = \lim_{t \to \infty} |U_{if}^{(2)}|^2 d\omega d\omega', \quad (56)$$

where \( U_{if}^{(2)} = a_{if}^{(2)} + a_{fi}^{(2)} (\omega \leftrightarrow \omega') \). A simple algebraic transformation yields

$$U_{if}^{(2)} = \left\{ \sum_\nu \left( \frac{\hat{V}_{if} \hat{V}_{\nu f}}{E_f - E_\nu + \omega + \frac{i}{2} (\Gamma_\nu - \Gamma_f)} \right) \right\} + \left\{ \sum_\nu \left( \frac{\hat{V}_{ifi} \hat{V}_{\nu fi}}{E_f - E_\nu + \omega' + \frac{i}{2} (\Gamma_\nu - \Gamma_f)} \right) \right\} \times \frac{1}{E_f - E_i + \omega + \omega' + \frac{i}{2} (\Gamma_i - \Gamma_f)}. \quad (57)$$

We see that equation (57) coincides with QED equation (27) in the case of \( i = 3s, f = 1s, \nu = 2p, \omega = \omega_{1f} \), and \( \omega' = \omega_{2f} \). The further derivations are exactly the same as in sections 2.3 and the final results are also exactly the same.

Similar considerations can be used in the case of three-photon transitions. For the derivation of the three-photon transition probability we have to go to the third-order of the perturbation theory. Substituting equation (55) into the right-hand side of equation (49) for the third-order correction \( a_{i\nu}^{(3)} \), we arrive at

$$i \frac{\partial a_{i\nu}^{(3)}(t)}{\partial t} = \sum_{\nu'} \hat{V}_{i\nu'} \hat{V}_{\nu'\nu} \hat{V}_{\nu f} \left[ 1 - e^{i(\epsilon_\nu - \epsilon_i) t} \right] \left( \frac{1 - e^{i(\epsilon_{\nu'} - \epsilon_f) t}}{\epsilon_{\nu'} - \epsilon_f} - \frac{1 - e^{i(\epsilon_f - \epsilon_i) t}}{\epsilon_f - \epsilon_i} \right) e^{i(\epsilon_f - \epsilon_\nu) t}. \quad (58)$$
Here
\[
\epsilon_n = E_n - \frac{i}{2} \Gamma_n + \omega + \omega', \\
\epsilon_f = E_f - \frac{i}{2} \Gamma_f + \omega' + \omega''.
\] (59)

Taking the limit \( t \to \infty \) we can find from equation (58)
\[
a_{ij}^{(3)} = \sum_{n\nu} \frac{V_{fn} V_{\nu i}}{(\epsilon_f - \epsilon_n)(\epsilon_f - \epsilon_f - \epsilon_n)}.
\] (60)

Substituting equations (53) and (59) into equation (60) yields
\[
a_{ij}^{(3)} = \frac{V_{fn} V_{\nu i}}{(\epsilon_f - \epsilon_n + \omega + \omega' + \omega'' + \frac{i}{2} (\Gamma_n - \Gamma_f))} \times \frac{1}{(\epsilon_f - E_{\nu} + \omega + \omega' + \omega'' + \frac{i}{2} (\Gamma_n - \Gamma_f))}.
\] (61)

Equation (61) coincides with QED equation (33) in the case of \( i = 3p, f = 1s \). The further derivations will apparently give the same result as obtained with the QED approach in section 2.5.

4. Comparison of the different regularizations of the cascade contributions

To present an illustration of the proper choice of the cascade regularization we will evaluate an absorbability of the two-photon emission presented by the frequency distribution for the two- or three-photon transitions. A similar property was first introduced in [33] and employed in [45, 46] to characterize the probability of absorption of the radiation emitted by one atom and absorbed by another atom. In our case this means the probability for the two-photon radiation emitted by one atom to be absorbed in the one-photon absorption lines of another atom.

It is convenient to begin with the absorbability \( X_{2s-1s}^{2\gamma} \) of two-photon emission, to compare it with the absorbability of the other two-photon transitions. We define \( X_{2s-1s}^{2\gamma} \) as [46]
\[
X_{2s-1s}^{2\gamma} = \frac{1}{2} \int_0^\infty L_{2p-1s}^{1\gamma} (\omega) \, dW_{2s-1s}^{2\gamma} (\omega),
\] (62)

where \( dW_{2s-1s}^{2\gamma} (\omega) \) is the frequency distribution (differential transition rate) for the \( 2s \to 1s \) two-photon transition and \( L_{2p-1s}^{1\gamma} \) is the Lorentz line profile for the one-photon \( 1s \to 2p \) absorption line, which is the same as the Lorentz profile for the \( 2p \to 1s \) emission line. The Lorentz profile is given by equation (18).

A dimensionless frequency distribution \( \frac{dW_{2s-1s}^{2\gamma}}{d\omega} \) is normalized to the two-photon transition probability
\[
\frac{1}{2} \int_0^{\omega_{2\gamma}} dW_{2s-1s}^{2\gamma} (\omega) = W_{2s-1s}^{2\gamma} = 8.229 \, s^{-1}.
\] (63)

In the integral equation (63) outside the interval \([0, \omega_0]\) where \( \omega_0 = E_{2s} - E_{1s} \), the function \( dW_{2s-1s}^{2\gamma} (\omega) \) should be set to zero. A dimensionless quantity \( X_{2s-1s}^{2\gamma} \) can be considered as an absorbability, i.e. an absolute probability of the absorption of the \( 2s \to 1s \) two-photon emission by the Lyman-alpha one-photon absorption line. A numerical evaluation gives the result
\[
X_{2s-1s}^{2\gamma} = 6.50 \times 10^{-22},
\] (64)

which shows that the two-photon radiation \( 2s \to 1s \) emitted by one hydrogen atom cannot be absorbed via Lyman-alpha transition by another hydrogen atom in the ground state. Note that the transition \( 2s \to 1s \) does not contain cascades, i.e. the frequency distribution \( dW_{2s-1s}^{2\gamma} \) has no singularities which have to be regularized. The quantity
\[
X_{2s-1s}^{2\gamma} = 1 - X_{2s-1s}^{2\gamma}
\] (65)

can be called ‘nonabsorbability’ and for \( 2s \to 1s \) two-photon transition
\[
Y_{2s-1s}^{2\gamma} = 1.
\] (66)

In table 1 we present the results of calculations for the absorbabilities for the two-photon \( 3s \to 1s, 3d \to 1s, 4d \to 1s \) and for the three-photon \( 3p \to 1s, 4p \to 1s \) and \( 3p \to 1s, 4p \to 1s, 5p \to 1s \) transitions. All these transitions require the regularization of cascade contributions. The emission of the two- and three-photon transitions can be absorbed by several one-photon absorption \( 1s \to 2p, 1s \to 3p, 2p \to 3s, 2p \to 3d \). As explained in the introduction, we consider only \( E1 \) transitions in the hydrogen atom. Then the generalization of equation (62) to the \( 3s(3d) \to 1s \) two-photon transition should be
\[
X_{3s(3d)-1s}^{2\gamma} = \frac{1}{2} \int_0^\infty \left[ L_{2p-1s}^{1\gamma} (\omega) + L_{2p-1s}^{1\gamma} (\omega) \right. \\
+ L_{3p-2s}^{1\gamma} (\omega) + L_{3d-2s}^{1\gamma} (\omega) \left. \right] dW_{3s(3d)-1s}^{2\gamma} (\omega).
\] (67)

Note, that the Lorentz line profile for the transition between two excited states \( nl \) and \( nl' \) equation (18) is given by an expression [40]
\[
L_{nl \to nl'}^{1\gamma} (\omega) = \frac{1}{2\pi} \frac{W_{nl \to nl'}^{1\gamma}}{(\omega + E_{nl'} - E_{nl})^2 + \frac{1}{4} (\Gamma_{nl} + \Gamma_{nl'})^2},
\] (68)

which in the denominator contains the sum of two widths \( \Gamma_{nl} \) and \( \Gamma_{nl'} \). In a similar way, the absorbabilities for two-photon transitions \( 4d \to 1s \) and for the three-photon transition \( 3p \to 1s \) were evaluated. The Lamb shifts of the levels \( nl \), \( nl' \) are neglected.

In table 1 the two values for \( X_{nl-1s}^{2\gamma} (m = 2, 3) \) are given: first, the value \( X_{nl-1s}^{2\gamma} \) evaluated with the frequency distribution \( dW_{nl-1s}^{2\gamma} (\omega) \) where the regularization is performed with two widths (e.g. equation (4) for the \( 3s \to 1s \) transition) and the regularization with only one width (i.e. equation (10) for the \( 3s \to 1s \) transition). In table 1 the values for \( W_{nl-1s}^{2\gamma}, W_{2p-1s}^{2\gamma} \) are also indicated to stress the difference between the transition rate for the pure two- and three-photon transitions \( 2s \to 1s + 2\gamma, 3p \to 1s + 3\gamma \) and the transition rates for the two- and three-photon transitions with cascades \( 3s \to 1s + 2\gamma, nd \to 1s + 2\gamma \) \( (n = 3, 4), 4p \to 1s + 3\gamma \). This difference correlates with large difference in the values of the
absorptivities for the two types of transitions and highlights that the presence of cascades fully changes the characteristics of the transitions.

A difference between \( X_{nl}^{my(1)} \) and \( X_{nl}^{my(2)} \) demonstrates the importance of employing the proper regularization scheme. In the case of the \( 3d-1s \) transition, the deviation becomes more than 10\% of the total \( X^{2\gamma} \) value, for the \( 4p \rightarrow 1s + 3\gamma \) transition this percentage reaches 9.5\%. The latter result shows that the proper regularization of the cascade contributions is important for the description of not only the two-photon but also for the three-photon processes.

### 5. Conclusions

In this paper we analysed the problem of the multiphoton transitions with cascades in hydrogen taking as examples the two-photon \( ns \rightarrow 1s + 2\gamma \), \( nd \rightarrow 1s + 2\gamma \) transitions and the three-photon \( np \rightarrow 1s + 3\gamma \) transitions \((n = 3, 4)\). We proved that the regularization of the singularities in the expressions for the cascade contributions to the transition rates should include the widths of both initial and intermediate states. In the existing literature on the subject, the regularization with only intermediate state width is also used. We demonstrated that both methods of regularization (with the sum of the intermediate and initial level widths and with intermediate level width only) give the same result for the total transition rate if only the cascade contribution is considered. However the results begin to differ if we consider the differential (with respect to the frequency) transition rates, taking into account the ‘pure two-photon’ (noncascade) contributions as well as the interference terms between cascade and noncascade terms. To demonstrate that this difference may become important we evaluated the ‘absorbativity’ of the two-photon emission by one-photon absorption lines. The absorptivity characterizes the effectiveness of the absorption of the two-photon radiation emitted by one atom, by another atom of the same kind. This may be important for the laboratory experiments where the multiphoton processes are investigated and also in astrophysics for the description of the cosmological recombination.

The analysis of the problem was performed within two different approaches. First, we applied the QED approach which allows for the \textit{ab initio} description of any multiphoton processes with any number of cascades. Within this approach it is necessary to fix the method of preparation (excitation) of the initial excited state. We choose the most natural for our purposes, the excitation by the two-photon or one-photon radiation emitted by another hydrogen atom. Actually the results for the decay of the excited state do not depend on the particular method of excitation provided that the linewidth of the exciting source is as broad or broader than the level width of the excited state under consideration. With this condition fulfilled, the cascade regularization in the decay of the excited state will always contain the sum of the initial and intermediate level widths, i.e. the width of the excited state will be present in the regularization. Second, we applied a standard QM phenomenological approach which however was never applied directly to the cascade regularization. This approach is based on the assumption that the initial excited state possesses its natural level width which is considered as a phenomenological parameter. Then we are automatically led to the same conclusion as within the QED approach.

Our description (both QED and QM) does not refer to the special cases when an atom is excited by a laser with the narrow bandwidth or something similar. In these cases the level width of the excited state will be diminished or disappear. Then, however the line profile of the excitation source will enter the description and the whole problem will require a special investigation.

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### Table 1. Absorptibility for the two- and three-photon emission \( nl \rightarrow 1s + k\gamma \) \((k = 2, 3)\) with different cascade regularizations.

| \( nl \) | \( X_{nl-1s}^{my(1)} \times 10^{-22} \) | \( X_{nl-1s}^{my(2)} \times 10^{-22} \) | \( Y_{nl-1s}^{my(1)} \times 10^{-22} \) | \( Y_{nl-1s}^{my(2)} \times 10^{-22} \) | \( W_{nl-1s} \gamma, s^{-1} \)
|---|---|---|---|---|---|
| 2s | 6.393 \times 10^{-22} | 6.393 \times 10^{-22} | 1.0000 | 1.0000 | 8.229 35 |
| 3s | 0.00497 | 0.00502 | 0.9954 | 0.9948 | 0.06317 \times 10^{6} |
| 3d | 0.04652 | 0.05217 | 0.97349 | 0.94783 | 0.64686 \times 10^{6} |
| 4d | 0.02118 | 0.02385 | 0.97882 | 0.97615 | 0.26013 \times 10^{6} |
| 3p | 2.547 \times 10^{-22} | 2.547 \times 10^{-22} | 1.0000 | 1.0000 | 1.01909 |
| 4p | 0.00221 | 0.00253 | 0.99747 | 0.99770 | 0.003929 \times 10^{6} |
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