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Spontaneous decay of an unstable atomic state in non-relativistic QED: a complete treatment including gauge invariance, renormalization and non-Markovian time behaviour

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Abstract. It is shown that the application of a self-consistent projection-operator method, developed recently by the author, makes possible a complete and gauge-invariant treatment of spontaneously decaying atomic state. All the shortcomings of the Weisskopf–Wigner gauge-dependent method (Markov approximation and the neglect of virtual transition, retardation and electron-spin effects) are removed. After deriving a gauge-invariant equation of motion for the decaying state, a new physical interaction picture (in which the unobservable interaction of the free electron with the vacuum radiation field is eliminated) is introduced. In this new interaction picture, apart from a natural solution of the renormalization problem, explicit finite analytic results for the whole non-Markovian Lyman-α spontaneous decay are derived. Furthermore, a quasi-Lorentzian expression for the radiative line shape containing the correct (complete) finite non-relativistic frequency shift, stemming from the Lamb shifts of both levels 1S and 2P, is calculated.

1. Introduction

The sixty-year-old problem of spontaneously decaying atomic states has been treated by many authors in the past. (Because of the very large number of papers dealing with the subject, here we quote only those papers which are relevant to the problem treated in the present paper. Further references can be found in the quoted papers and books.) The shortcomings contained in the Weisskopf–Wigner method (gauge dependence, two-level-atom, rotating-wave, dipole and Markov approximations, and the neglect of electron-spin effects) [1, 2] were removed in various degrees. Thus, for example within the two-level-atom approximation, it has been pointed out that the dipole approximation (neglect of retardation effects) requires the introduction of an unnatural cutoff frequency and leads to incorrect asymptotic results [3]. By removing the dipole and Markov approximation some authors calculated asymptotic deviations from exponential decay for a two-level atom (see e.g., [3–5], and further references quoted in these works). In the case of Lyman-α spontaneous emission, a complete non-Markovian treatment including deviations from exponential decay for finite times within the scope of the two-level atom has been presented in our previous paper [6]. Without investigating the time development of a decaying state, the line shape has been studied by Arnous and Heitler [7] (who applied a very cumbersome formalism) and by Low [8] who used a covariant S-matrix method. However, the non-Markovian

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effects, which, as will be shown in this paper, change the Lorentzian frequency spectrum to a quasi-Lorentzian one, were not included in their calculations [7, 8].

The objective of this paper is to present a complete (gauge-invariant) treatment of the spontaneously decaying state. This treatment, unlike all previous treatments in the literature, should yield complete results for the non-Markovian time behaviour and radiative line shape including virtual transition effects. In order to achieve this, the two-level approximation (appearing in [1–6]), which leads to an incomplete frequency (Lamb) shift (stemming only from the real transition to the lower-lying level), should be avoided in our treatment. In other words, the aim of the present paper is to include the whole discrete and continuous atomic spectrum in our calculations. However, since the inclusion of virtual ('energy-non-conserving') transitions automatically leads to divergent results, the renormalization problem has to be solved first, by using a consistent method. This has been done in our previous paper [9] by applying a *self-consistent projection-operator method* (SCPOM) developed recently [10, 11].

The application of the SCPOM makes it possible to introduce a new *physical real-transition interaction picture*, in which the dynamics caused by virtual transitions and the unobservable interaction of the free electron with the vacuum radiation field is eliminated. The latter, as has been shown in [9], leads to a natural and *unambiguous solution* of the longstanding *renormalization problem* in non-relativistic quantum electrodynamics without using the conventional concept of mass renormalization, which, as pointed out by Au and Feinberg [12], is not unambiguous. The main differences between the mass renormalization method and our method can be summarized as follows.

(i) In our Hamiltonian, from the very beginning, only the *experimentally observable (physical) mass* appears (we do not differentiate between observable, bare and electromagnetic mass).

(ii) No unnatural *mass-renormalization counter-term* (calculated in the second order in the charge e) has to be included in the Hamiltonian.

(iii) By introducing a *new interaction picture*, in which the experimentally unobservable interaction of the free electron with the vacuum radiation field is eliminated, an *unambiguous* and self-consistent *renormalization* is automatically performed.

In the present paper, by introducing this new physical interaction picture (without making dipole, Markov and two-level-atom approximations), including the spin–radiation-field interaction term and the term $(e^2/2mc^2)\boldsymbol{A}^2(\boldsymbol{R})$ in the Hamiltonian (both of which were neglected in [1–7]) and, finally, taking into account the effect of a unitary gauge transformation (which was not done in [1–7]), a gauge-invariant, rigorous analytic result for non-Markovian spontaneous Lyman-α decay is obtained. This result, containing the exponentially decaying term and a term describing the deviations from exponential decay, gives a complete description of the time evolution of the probability amplitude for finding the atom in an unstable state.

Unlike all treatments in the literature [1–6] (and references quoted therein), our exponentially decaying term contains the *complete* $2P \rightarrow 1S$ *frequency (Lamb) shift*, which is finite (cutoff-independent) and can be explicitly evaluated by taking into account the whole discrete and continuous atomic spectrum [9].

The paper is organized as follows. In section 2, by applying the SCPOM, a gauge-invariant treatment of the whole time evolution of a spontaneously decaying atomic state is presented. In the new physical interaction picture, without using the mass-renormalization concept, finite expressions for energy-level (Lamb) shifts are obtained. In section 3 the obtained results are applied for obtaining a complete description of the time evolution in the case of the Lyman-α spontaneous emission. In section 4 a conclusion is drawn. In

appendix A the gauge invariance of a level-shift expression, appearing in the calculations in section 2, is proved.

2. A gauge-invariant treatment of the spontaneous emission based on the SCPOM

The Hamiltonians for the unperturbed hydrogenic atom and the quantized radiation field (in the Coulomb gauge) read as

$$H^{\mathrm{A}} = \left[\frac{P^2}{2m} + V(R)\right] \otimes I_{sp} \qquad H^{\mathrm{R}} = \hbar c \sum_{\lambda=1}^{2} \int \mathrm{d}^3k\, k a^+(\boldsymbol{k}, \lambda) a^-(\boldsymbol{k}, \lambda) \tag{2.1}$$

where I_{sp} is the unit operator in the spin Hilbert space, $a^{\pm}(\boldsymbol{k}, \lambda)$ are the photon creation and annihilation operators for the mode $(\boldsymbol{k}, \lambda)$ and λ is the polarization index. The total Hamiltonian H reads as

$$H = H^0 + H^{\mathrm{I}} \qquad H^0 = H^{\mathrm{A}} + H^{\mathrm{R}} \tag{2.2}$$

and the interaction Hamiltonian between the atom and the radiation field is given by

$$H^{\mathrm{I}} = \sum_{l=1}^{3} H_l^{\mathrm{I}} \tag{2.3}$$

$$H_1^{\mathrm{I}} = \frac{e^2}{2mc^2} A^2(R) \qquad H_2^{\mathrm{I}} = -\frac{e}{mc} A(R) \cdot P \qquad H_3^{\mathrm{I}} = -\frac{e\hbar}{2mc} \sigma \cdot B(R) \tag{2.4}$$

where

$$A(R) = \frac{\sqrt{\hbar c}}{2\pi} \sum_{\lambda=1}^{2} \int \mathrm{d}^3k (k)^{-1/2} e_{k,\lambda} [a^-(\boldsymbol{k}, \lambda) \mathrm{e}^{\mathrm{i}kR} + a^+(\boldsymbol{k}, \lambda) \mathrm{e}^{-\mathrm{i}kR}] \tag{2.5}$$

is the vector potential, σ is the Pauli spin operator, $e_{k,\lambda}$ is the polarization vector and $B = \nabla \times A$ is the magnetic field.

We now introduce the interaction picture and separate the interaction Hamiltonian into two parts:

$$\begin{aligned} H^{\mathrm{I}}(t) &= \mathrm{e}^{(\mathrm{i}/\hbar)H^0 t} H^{\mathrm{I}} \mathrm{e}^{-(\mathrm{i}/\hbar)H^0 t} = \mathrm{e}^{(\mathrm{i}/\hbar)H^0 t} [H^{\mathrm{RT}} + H^{\mathrm{VT}}] \mathrm{e}^{-(\mathrm{i}/\hbar)H^0 t} \\ &= H^{\mathrm{RT}}(t) + H^{\mathrm{VT}}(t) \end{aligned} \tag{2.6}$$

where H^{RT} is the 'real-transition' part (containing the 'energy-conserving' transitions to lower-lying atomic levels) leading to the decay of a state, and H^{VT} is the 'virtual-transition' part (containing the 'energy-non-conserving' transitions) leading to the energy-level (Lamb) shifts.

The proof of the gauge invariance will be performed in a manner similar to that of Aharonov and Au [13], who have proven a general theorem, whose results are also applicable to the present case. Therefore, in order to prove the gauge independence of our results, instead of H, analogously as in [13], we will use the unitary gauge-transformed Hamiltonian $\tilde{H}$ in what follows:

$$\tilde{H} = \mathrm{e}^{(\mathrm{i}/c\hbar)e\chi} H \mathrm{e}^{-(\mathrm{i}/c\hbar)e\chi} = H^0 + \tilde{H}^{\mathrm{I}} \tag{2.7}$$

where

$$\tilde{H}^{\mathrm{I}} = H^{\mathrm{I}} + \frac{\mathrm{i}}{c\hbar} e[\chi, H^0] + \frac{\mathrm{i}}{c\hbar} e[\chi, H^{\mathrm{I}}] - \frac{1}{2c^2\hbar^2} e^2[\chi, [\chi, H^0]] \tag{2.8}$$

with a time-independent gauge function $\chi(R)$. Such gauge functions are very often used in quantum optics (see e.g. [14, 15] and references quoted therein).

In the interaction picture we again decompose the gauge-transformed interaction Hamiltonian into two parts:

$$\tilde{H}^{\mathrm{I}}(t) = \mathrm{e}^{(\mathrm{i}/\hbar)H^0 t} \tilde{H}^{\mathrm{I}} \mathrm{e}^{-(\mathrm{i}/\hbar)H^0 t} = \tilde{H}^{\mathrm{RT}}(t) + \tilde{H}^{\mathrm{VT}}(t) \tag{2.9}$$

where

$$\tilde{H}^{\mathrm{RT}}(t) = \mathrm{e}^{(\mathrm{i}/\hbar)H^0 t} \left\{ H^{\mathrm{RT}} + \frac{\mathrm{i}}{c\hbar} e[\chi, H^{\mathrm{RT}}] \right\} \mathrm{e}^{-(\mathrm{i}/\hbar)H^0 t} \tag{2.10}$$

is the 'real-transition' Hamiltonian causing real transitions to lower-lying atomic levels, and

$$\tilde{H}^{\mathrm{VT}}(t) = \mathrm{e}^{(\mathrm{i}/\hbar)H^0 t} \left\{ H^{\mathrm{VT}} + \frac{\mathrm{i}}{c\hbar} e[\chi, H^0] + \frac{\mathrm{i}}{c\hbar} e[\chi, H^{\mathrm{VT}}] - \frac{1}{2c^2\hbar^2} e^2[\chi, [\chi, H^0]] \right\} \mathrm{e}^{-(\mathrm{i}/\hbar)H^0 t} \tag{2.11}$$

is the 'virtual-transition' Hamiltonian causing virtual transitions (including the 'energy-non-conserving' transitions to higher-lying levels).

To concentrate our investigations on the dynamics caused by the 'real-transition' Hamiltonian $\tilde{H}^{\mathrm{RT}}$, we have to introduce a new *'real-transition' interaction picture* (RTIP) in which the dynamics caused by the 'virtual-transition' Hamiltonian H^{VT} is eliminated. The dynamics caused by $\tilde{H}^{\mathrm{VT}}$ is described by the time-development operator $\tilde{U}_{\mathrm{VT}}(t, 0)$:

$$\frac{\mathrm{d}\tilde{U}_{\mathrm{VT}}(t, 0)}{\mathrm{d}t} = -\frac{\mathrm{i}}{\hbar} \tilde{H}^{\mathrm{VT}}(t) \tilde{U}_{\mathrm{VT}}(t, 0). \tag{2.12}$$

In the RTIP the Hamiltonian H^{RT} reads as

$$\tilde{H}^{\mathrm{RT}}(t, t) \equiv \tilde{U}^{+}_{\mathrm{VT}}(t, 0) \tilde{H}^{\mathrm{RT}}(t) \tilde{U}_{\mathrm{VT}}(t, 0). \tag{2.13}$$

Furthermore, the Schrödinger equation in the RTIP takes the form

$$\frac{\mathrm{d}|\psi(t)\rangle}{\mathrm{d}t} = -\frac{\mathrm{i}}{\hbar} \tilde{H}^{\mathrm{RT}}(t, t) |\psi(t)\rangle \tag{2.14}$$

where

$$|\psi(0)\rangle = |nlm, m_s, v\rangle = |nlm\rangle \otimes |m_s\rangle \otimes |v\rangle \tag{2.15}$$

is the decaying state, with the energy eigenvalue E_n (n, l, m, m_s are the principal, angular momentum, magnetic and spin-magnetic quantum numbers, respectively, and v denotes the vacuum state of the radiation field). Then, the application of the SCPOM [10, 11] to the

above Schrödinger equation leads to an *exact closed equation of motion for the reduced state vector* $\mathcal{P}|\psi(t)\rangle$:

$$\frac{\mathrm{d}\mathcal{P}|\psi(t)\rangle}{\mathrm{d}t} = -\frac{\mathrm{i}}{\hbar}\mathcal{P}\tilde{H}^{\mathrm{RT}}(t,t)\mathcal{P}|\psi(t)\rangle - \frac{1}{\hbar^2}\int_0^t \mathrm{d}t'\,\mathcal{P}\tilde{H}^{\mathrm{RT}}(t,t)\tilde{T}(t,t')[I-\mathcal{P}]\tilde{H}^{\mathrm{RT}}(t',t')\mathcal{P}|\psi(t')\rangle \tag{2.16}$$

$$\tilde{T}(t,t') = \mathcal{T}\exp\left[-\frac{\mathrm{i}}{\hbar}\int_{t'}^{t}\mathrm{d}t''(I-\mathcal{P})\tilde{H}^{\mathrm{RT}}(t'',t'')\right] \tag{2.17}$$

where

$$\mathcal{P} = |nlm, m_s, v\rangle\langle nlm, m_s, v| \tag{2.18}$$

is our projection operator, $\mathcal{T}$ is the Dyson time-ordering operator, I is the unit operator in the product Hilbert space $\mathcal{H}_{\mathrm{A}} \otimes \mathcal{H}_{\mathrm{R}}$ of the atom and radiation field.

In equation (2.16) it can be easily shown that the application of the second-order approximation in the interaction (or in the charge e), the so-called Born approximation (BA), which is equivalent to the *self-consistent truncation condition* [10, 11],

$$(I-\mathcal{P})\tilde{H}^{\mathrm{RT}}(t,t)(I-\mathcal{P}) = 0 \tag{2.19}$$

leads to the restricted Hamiltonian $[\tilde{H}^{\mathrm{RT}}(t,t)]^{\mathrm{S}}$:

$$[\tilde{H}^{\mathrm{RT}}(t,t)]^{\mathrm{S}} = \mathcal{P}\tilde{U}_{\mathrm{VT}}^{+}(t,0)\mathcal{P}H^{\mathrm{RT}}(t)(I^{\mathrm{S}}-\mathcal{P})\tilde{U}_{\mathrm{VT}}(t,0)(I^{\mathrm{S}}-\mathcal{P}) + \mathrm{HC} \tag{2.20}$$

which acts in the subspace $\mathcal{H}^{\mathrm{S}}$ defined by the state vectors $\{(H^{\mathrm{RT}})^l|\psi(0)\rangle, l = 0, 1\}$. In obtaining the above equation we used the fact that as a consequence of the self-consistent truncation condition, the gauge-dependent term in $\tilde{H}^{\mathrm{RT}}(t)$ (cf equation (2.10)) vanishes.

The expression

$$\mathcal{P}\tilde{U}_{\mathrm{VT}}(t,0)\mathcal{P} = \mathcal{P}\tilde{U}_{\mathrm{VT}}(t,0)|nlm, m_s, v\rangle\langle nlm, m_s, v| \tag{2.21}$$

can be calculated from the equation of motion for the reduced state vector $\mathcal{P}|\psi^{\mathrm{VT}}(t)\rangle \equiv \mathcal{P}\tilde{U}_{\mathrm{VT}}(t,0)|nlm, m_s, v\rangle$ (which can be obtained by applying the SCPOM):

$$\frac{\mathrm{d}\mathcal{P}|\psi^{\mathrm{VT}}(t)\rangle}{\mathrm{d}t} = -\frac{\mathrm{i}}{\hbar}\mathcal{P}\tilde{H}^{\mathrm{VT}}(t)\mathcal{P}|\psi^{\mathrm{VT}}(t)\rangle - \left(\frac{1}{\hbar^2}\right)\int_0^t \mathrm{d}\tau\,\mathcal{P}\tilde{H}^{\mathrm{VT}}(t)\tilde{T}_{\mathrm{VT}}(t,t-\tau)(I-\mathcal{P})\tilde{H}^{\mathrm{VT}}(t-\tau)\mathcal{P}|\psi^{\mathrm{VT}}(t-\tau)\rangle \tag{2.22}$$

$$\tilde{T}_{\mathrm{VT}}(t,t-\tau) = \mathcal{T}\exp\left[-\frac{\mathrm{i}}{\hbar}\int_{t-\tau}^{t}\mathrm{d}t'(I-\mathcal{P})\tilde{H}^{\mathrm{VT}}(t')\right]. \tag{2.23}$$

The application of the BA in equation (2.22), which is equivalent to the self-consistent truncation condition [10, 11],

$$(I-\mathcal{P})\tilde{H}^{\mathrm{VT}}(t)(I-\mathcal{P}) = 0 \tag{2.24}$$

leads to the restricted Hamiltonian:

$$[\tilde{H}^{\mathrm{VT}}(t)]^{\mathrm{S}} = \tilde{H}_a^{\mathrm{VT}} + \tilde{H}_b^{\mathrm{VT}}(t) \tag{2.25}$$

$$\tilde{H}_a^{\mathrm{VT}} = \mathcal{P}\left\{\frac{e^2}{2mc^2}A^2 + \frac{\mathrm{i}}{c\hbar}e[\chi, H^{\mathrm{VT}}] - \frac{1}{2c^2\hbar^2}e^2[\chi,[\chi, H^0]]\right\}\mathcal{P} \tag{2.26}$$

$$\tilde{H}_b^{\mathrm{VT}}(t) = \mathrm{e}^{(\mathrm{i}/\hbar)H^0 t}\tilde{H}_b^{\mathrm{VT}}\mathrm{e}^{-(\mathrm{i}/\hbar)H^0 t} \tag{2.27}$$

$$\tilde{H}_b^{\mathrm{VT}} = \mathcal{P}\left\{H^{\mathrm{VT}} + \frac{\mathrm{i}}{c\hbar}e[\chi, H^0]\right\}(I_{\mathrm{VT}}^{\mathrm{S}} - \mathcal{P}) + \mathrm{HC} \tag{2.28}$$

with the unit operator $I_{\mathrm{VT}}^{\mathrm{S}}$ acting in the subspace $\mathcal{H}_{\mathrm{VT}}^{\mathrm{S}}$ defined by the state vectors $\{(\tilde{H}^{\mathrm{VT}})^k|\psi(0)\rangle, k = 0, 1\}$.

The above equation (2.22) in the BA and so-called Markov approximation (which consists of neglecting memory effects and replacing the upper limit of the time integration with ∞ by introducing a damping factor $\mathrm{e}^{-\eta\tau}$ in the integral ($\eta \to 0$ after carrying out the integration)) [9–11] reduces to

$$\frac{\mathrm{d}\mathcal{P}|\psi^{\mathrm{VT}}(t)\rangle}{\mathrm{d}t} = (-\mathrm{i}/\hbar)\Delta\tilde{E}_{nlm}^{\mathrm{VT(unr)}}\mathcal{P}|\psi^{\mathrm{VT}}(t)\rangle \tag{2.29}$$

$$\Delta\tilde{E}_{nlm}^{\mathrm{VT(unr)}} = \Delta\tilde{E}_{nlm}^{\mathrm{VT},1} + \Delta\tilde{E}_{nlm}^{\mathrm{VT},2} \tag{2.30}$$

$$\Delta\tilde{E}_{nlm}^{\mathrm{VT},1} = \langle nlm, m_s, v|\tilde{H}_a^{\mathrm{VT}}|nlm, m_s, v\rangle \tag{2.31}$$

$$\Delta\tilde{E}_{nlm}^{\mathrm{VT},2} = \lim_{\eta\to 0}\langle nlm, m_s, v|\tilde{H}_b^{\mathrm{VT}}\frac{1}{E_n - H_0 + i\eta}\tilde{H}_b^{\mathrm{VT}}|nlm, m_s, v\rangle \tag{2.32}$$

where $\Delta E_{nlm}^{\mathrm{VT(unr)}}$ (subscript m_s is dropped) is the unrenormalized radiative energy-level (Lamb) shift stemming from virtual-transition Hamiltonian H^{VT}. As will be shown in the appendix the expression $\Delta\tilde{E}_{nlm}^{\mathrm{VT(unr)}} = \Delta E_{nlm}^{\mathrm{VT(unr)}}$, appearing in equation (2.29), is gauge-independent. Therefore, the expression $\mathcal{P}\tilde{U}_{\mathrm{VT}}^{+}(t, 0)\mathcal{P}$ is also gauge-independent and reduces to

$$\mathcal{P}\tilde{U}_{\mathrm{VT}}^{+}(t, 0)\mathcal{P} = \mathcal{P}U_{\mathrm{VT}}^{+}(t, 0)\mathcal{P} = \mathrm{e}^{(\mathrm{i}/\hbar)t\Delta E_{nlm}^{\mathrm{VT(unr)}}}\mathcal{P}. \tag{2.33}$$

However, since $U_{\mathrm{VT}}(t, 0)$ contains the physically unobservable dynamics of the free electron interacting with the radiation field, a new *'physical' (bound-electron) interaction picture*, in which this dynamics is eliminated, has to be introduced. This, as has been shown in [9], leads to a quite *natural solution of the renormalization problem*. That is to say, *without using the conventional mass-renormalization concept*, which is not unambiguous [12], *finite Lamb-shift results* can be obtained. The time-development operator describing the unobservable dynamics of the free electron is given by

$$\frac{\mathrm{d}U_{\mathrm{free}}(t, 0)}{\mathrm{d}t} = -\frac{\mathrm{i}}{\hbar}H^{\mathrm{free}}(t)U_{\mathrm{free}}(t, 0) \tag{2.34}$$

where

$$H^{\mathrm{free}}(t) \equiv \exp\left[\frac{\mathrm{i}}{\hbar}\left(\frac{P^2}{2m} + H_{\mathrm{R}}\right)t\right]H^{\mathrm{I}}\exp\left[-\frac{\mathrm{i}}{\hbar}\left(\frac{P^2}{2m} + H_{\mathrm{R}}\right)t\right]. \tag{2.35}$$

In the BA and Markov approximation, the equation of motion for $\mathcal{P}|\psi^{\text{free}}(t)\rangle \equiv \mathcal{P}U_{\text{free}}(t,0)|nlm, m_s, v\rangle$ describing the time evolution of the reduced state vector under the influence of the free-electron–field interaction [9] reads as

$$\frac{\mathrm{d}\mathcal{P}|\psi^{\text{free}}(t)\rangle}{\mathrm{d}t} = (-\mathrm{i}/\hbar)\Delta E^{\text{free}}_{nlm}\mathcal{P}|\psi^{\text{free}}(t)\rangle \tag{2.36}$$

where $\Delta E^{\text{free}}_{nlm}$ is the self-energy of the free electron:

$$\Delta E^{\text{free}}_{nlm} = \Delta E^{\text{free},1}_{nlm} + \Delta E^{\text{free},2}_{nlm} \tag{2.37}$$

with

$$\Delta E^{\text{free},1}_{nlm} = \langle nlm|\frac{e^2}{2mc^2}A^2|nlm\rangle \tag{2.38}$$

$$\Delta E^{\text{free},2}_{nlm} = \frac{e^2\hbar}{(2\pi)^2m^2c}\lim_{\eta\to 0}\sum_{\lambda}\sum_{\epsilon=1,2}\int\frac{\mathrm{d}^3k}{k}\langle nlm|h_\epsilon G_{\text{free}}h_\epsilon|nlm\rangle \tag{2.39}$$

$$G_{\text{free}} = (-1)[K^2/(2m) - (\boldsymbol{K}\cdot\boldsymbol{P})/m + cK + \mathrm{i}\eta]^{-1} \tag{2.40}$$

$$\boldsymbol{K} \equiv \hbar\boldsymbol{k} \qquad h_1 \equiv \boldsymbol{e}_{k,\lambda}\cdot\boldsymbol{P} \qquad h_2 \equiv \tfrac{1}{2}\boldsymbol{\sigma}\cdot(\hbar\boldsymbol{k}\times\boldsymbol{e}_{k,\lambda}). \tag{2.41}$$

Now we can introduce the *physical interaction picture*:

$$\mathcal{P}|\psi^{\text{VT}}(t)\rangle_{\text{PI}} \equiv \mathrm{e}^{(\mathrm{i}/\hbar)t\Delta E^{\text{free}}_{nlm}}\mathcal{P}|\psi^{\text{VT}}(t)\rangle. \tag{2.42}$$

In this new picture equation (2.33) reduces to

$$\mathcal{P}U^{+}_{\text{VT}}(t,0)\mathcal{P} = \mathrm{e}^{(\mathrm{i}/\hbar)t\Delta E^{\text{VT}}_{nlm}}\mathcal{P} \tag{2.43}$$

where $\Delta E^{\text{VT}}_{nlm}$ is the 'renormalized' Lamb shift stemming from virtual transitions:

$$\Delta E^{\text{VT}}_{nlm} = \Delta E_{nlm} - \Delta E^{\text{RT}}_{nlm} \tag{2.44}$$

with ΔE_{nlm} as the total Lamb shift [9]:

$$\Delta E_{nlm} = \frac{e^2\hbar}{(2\pi)^2m^2c}\lim_{\eta\to 0}\sum_{\lambda}\sum_{\epsilon=1,2}\int\frac{\mathrm{d}^3k}{k}\langle nlm|h_\epsilon(G_{\text{bound}} - G_{\text{free}})h_\epsilon|nlm\rangle \tag{2.45}$$

where

$$G_{\text{bound}} = [E_n - (\boldsymbol{P}-\boldsymbol{K})^2/(2m) - V(R) - cK + \mathrm{i}\eta]^{-1} \tag{2.46}$$

and with $\Delta E^{\text{RT}}_{nlm}$ as the Lamb shift stemming from real transitions to lower-lying levels:

$$\Delta E^{\text{RT}}_{nlm} = \lim_{\eta\to 0}\langle nlm, m_s, v|H^{\text{RT}}\frac{1}{E_n - H_0 + \mathrm{i}\eta}H^{\text{RT}}|nlm, m_s, v\rangle. \tag{2.47}$$

In obtaining equation (2.45) we used the fact that because

$$\Delta E^{\text{VT},1}_{nlm} = \Delta E^{\text{free},1}_{nlm} \tag{2.48}$$

(see equations (2.31), (2.38) and (2.26)) the term A^2 does not give any contribution to the total Lamb shift.

Since it can be shown, quite similarly to the above, that the expression $(I^{\text{S}} - \mathcal{P})\tilde{U}_{\text{VT}}(t,0)(I^{\text{S}} - \mathcal{P})$ is also gauge-independent, it follows, finally, that the *restricted real-transition Hamiltonian* given by equation (2.20) is *gauge-independent* as well. Therefore, the equation of motion for the reduced state vector, equation (2.16), takes the following reduced gauge-invariant form in the BA:

$$\frac{\mathrm{d}\mathcal{P}|\psi(t)\rangle}{\mathrm{d}t} = -\frac{1}{\hbar^2}\int_0^t \mathrm{d}t'\,\mathcal{P}[H^{\text{RT}}(t,t)]^{\text{S}}[I-\mathcal{P}][H^{\text{RT}}(t',t')]^{\text{S}}\mathcal{P}|\psi(t')\rangle. \tag{2.49}$$

In the next section this equation will be applied to the case of spontaneous Lyman-α decay.

3. Complete solution for the time evolution in the case of Lyman-α spontaneous emission

Now, we will apply the equations and results derived in the preceding section to the case of spontaneous Lyman-α (2P $\rightarrow$ 1S) decay:

$$|\psi(0)) = |2\mathrm{P}\rangle \otimes |v\rangle = |n = 2, l = 1, m, m_s\rangle \otimes |v\rangle \tag{3.1}$$

$$|1\mathrm{S}\rangle = |n = 1, l = 0, m = 0, m_s\rangle. \tag{3.2}$$

In our case the projection operator takes the form

$$\mathcal{P} = |2\mathrm{P}\rangle\langle 2\mathrm{P}| \otimes |v\rangle\langle v| \tag{3.3}$$

and the unit operator acting in the subspace $\mathcal{H}^{\mathrm{S}}$ reads as

$$I^{\mathrm{S}} = \mathcal{P} + |1\mathrm{S}\rangle\langle 1\mathrm{S}| \otimes \int_0^\infty \mathrm{d}\omega |\omega\rangle\langle\omega| \tag{3.4}$$

where $|\omega\rangle$ are the one-photon radiation-field states for a photon with the frequency ω, and the quantum numbers $j = 1, m$ and $\tau = 0$ (electric multipole field). Then, by inserting the following equations (cf equation (2.43)):

$$\mathcal{P}U_{\mathrm{VT}}^{+}(t,0)\mathcal{P} = \mathcal{P}\langle 2\mathrm{P}, v|U_{\mathrm{VT}}^{+}(t,0)|2\mathrm{P}, v\rangle \approx \mathcal{P}\mathrm{e}^{(\mathrm{i}/\hbar)t\Delta E_{2\mathrm{P}}^{\mathrm{VT}}} \tag{3.5}$$

$$(I^{\mathrm{S}} - \mathcal{P})U_{\mathrm{VT}}(t,0)(I^{\mathrm{S}} - \mathcal{P}) \approx (I^{\mathrm{S}} - \mathcal{P})\mathrm{e}^{(-\mathrm{i}/\hbar)t\Delta E_{1\mathrm{S}}} \qquad \Delta E_{1\mathrm{S}}^{\mathrm{RT}} = 0 \tag{3.6}$$

into equation (2.20), the restricted Hamiltonian (which follows in the BA) takes the reduced form

$$[H^{\mathrm{RT}}(t,t)]^{\mathrm{S}} = \int_0^\infty \mathrm{d}\omega H_{2\mathrm{P},1\mathrm{S}}(\omega)\mathrm{e}^{\mathrm{i}(\bar{\omega}_0-\omega)t}|2\mathrm{P}\rangle\langle 1\mathrm{S}| \otimes |v\rangle\langle\omega| + \mathrm{HC} \tag{3.7}$$

where

$$\omega_0 \equiv (1/\hbar)(E_{2\mathrm{P}} - E_{1\mathrm{S}}) \qquad \bar{\omega}_0 \equiv \omega_0 + \Delta\omega_0^{\mathrm{VT}} \qquad \Delta\omega_0^{\mathrm{VT}} \equiv (1/\hbar)(\Delta E_{2\mathrm{P}}^{\mathrm{VT}} - \Delta E_{1\mathrm{S}}) \tag{3.8}$$

and the transition matrix element $H_{2\mathrm{P},1\mathrm{S}}(\omega)$ is given by [16, 6]

$$H_{2\mathrm{P},1\mathrm{S}}(\omega) = (\lambda/2\pi)^{1/2}\hbar(-\mathrm{i}\omega^{1/2})[1 + (\omega/\Omega)^2]^{-2} \tag{3.9}$$

with $\lambda = \gamma/\omega_0$ (γ is the Einstein coefficient for spontaneous Lyman-α transition) and $\Omega = 3c/(2a_0)$ (a_0 is the Bohr radius). Therefore, equation (2.49) reduces to a closed equation of motion for the probability amplitude

$$b_{2\mathrm{P}}(t) \equiv \langle 2\mathrm{P}| \otimes \langle v|\mathcal{P}|\psi(t)\rangle \tag{3.10}$$

of finding the atom in the initial state $|2\mathrm{P}\rangle$ and zero photons in the radiation field:

$$\frac{\mathrm{d}b_{2\mathrm{P}}(t)}{\mathrm{d}t} = (-\lambda/2\pi)\int_0^\infty \mathrm{d}\omega\, f(\omega)\int_0^t \mathrm{d}\tau\, \mathrm{e}^{\mathrm{i}(\bar{\omega}_0-\omega)\tau}b_{2\mathrm{P}}(t-\tau) \qquad b_{2\mathrm{P}}(0) = 1 \tag{3.11}$$

where

$$f(\omega) = \frac{2\pi}{\lambda\hbar^2}|H_{2P,1S}(\omega)|^2 \tag{3.12}$$

is the natural cutoff function. Since this equation is identical to equation (2.1) [6] (with the significant difference that instead of ω_0 the virtual-transition corrected frequency $\bar{\omega}_0$ has to be used throughout), all equations in section II of [6] can be used in the present case as well. This leads to the result

$$b_{2P}(t) = R(t) + D(t) \tag{3.13}$$

where

$$R(t) = (1 + \Delta_R)e^{i\bar{\omega}_0 t}e^{-iu_{-1}t} \qquad |\Delta_R| < 10\lambda \tag{3.14}$$

is the residue of the so-called Weisskopf–Wigner pole (lying on the Riemann sheet –1):

$$u_{-1} = \omega_0 + \Delta\omega_0 - i\gamma/2 + \Delta_u \qquad |\Delta_u/u_{-1}| < 4.1 \times 10^{-13} \tag{3.15}$$

$$\Delta\omega_0 = \Delta\omega_0^{VT} + \Delta\omega_0^{RT} \qquad \Delta\omega_0^{RT} = \frac{11\gamma}{24\pi} - \frac{5\lambda\Omega}{64} - \frac{\gamma}{2\pi}\ln\left[\frac{\Omega}{\bar{\omega}_0}\right] \tag{3.16}$$

describing the Markovian behaviour (exponential decay). Here $\Delta\omega_0$ is the complete frequency shift calculated in [9, 17]:

$$\Delta\omega_0/2\pi = \Delta E_{2P}/h - \Delta E_{1S}/h = -8132.8 \text{ MHz} \tag{3.17}$$

$$\Delta E_{1S}/h = 8136.9 \text{ MHz} \qquad \Delta E_{2P}/h = 4.1 \text{ MHz}. \tag{3.18}$$

(h is the Planck constant). In the above equation (3.16) $\Delta\omega_0^{RT}$ is the Lamb shift stemming from the real $2P \rightarrow 1S$ transition and can be calculated directly in the usual second-order perturbation theory as well:

$$\Delta E_{2P}^{RT} = \hbar\Delta\omega_0^{RT} = PV\int_0^\infty d\omega \frac{|H_{2P,1S}(\omega)|^2}{E_{2P} - E_{1S} - \hbar\omega} = -\frac{\lambda\hbar}{2\pi}I(\omega_0) \tag{3.19}$$

(*PV* is the Cauchy principal value). The solution of the integral:

$$I(u) \equiv \int_0^\infty d\omega \frac{f(\omega)}{\omega - u} \tag{3.20}$$

(which has been given in [6], equations (2.5)–(2.7)) yields precisely the result of equation (3.16) for $\Delta\omega_0^{RT}$ if $u = \omega_0$ is inserted. Further, $D(t)$ describes the non-Markovian behaviour (deviation from exponential decay):

$$D(t) = M(t) + \Delta_D(t) \qquad M(t) = -\frac{\lambda}{2\pi(\omega_0 + \Delta\omega_0)^2 t^2}e^{i\bar{\omega}_0 t} \tag{3.21}$$

with $M(t)$ as the asymptotic main term and the error estimate:

$$|\Delta_D(t)/M(t)| < 16.6 \times 10^{-2} \qquad t \geqslant 10^{-14} \text{ s}. \tag{3.22}$$

Finally, as in section 5 of [18], by suitably deforming the path of integration in the Laplace inversion expression for $b_{2P}(t)$ (cf equation (2.8) in [6]), the following integral representation for $b_{2P}(t)$ can be written down:

$$b_{2P}(t) = e^{i\tilde{\omega}_0 t} \int_0^\infty d\omega\, e^{-i\omega t} S(\omega) \tag{3.23}$$

with a *quasi-Lorentzian frequency spectrum*

$$S(\omega) = (\lambda/2\pi) f(\omega) / \{[\omega - (\omega_0 + \Delta\omega_0) + (\lambda/2\pi) J(\omega)]^2 + (\lambda/2)^2 f^2(\omega)\} \tag{3.24}$$

where both the *decay rate* $\lambda f(\omega)$ and the *frequency shift*

$$J(\omega) \equiv I(\omega) - i\pi f(\omega) - [I(\omega_0) - i\pi f(\omega_0)] \tag{3.25}$$

($I(\omega)$ is given by equation (3.20)) *depend on* the the *frequency* ω. However, since these frequency-dependent functions are multipled by a very small factor $\lambda \approx 10^{-8}$, the effect is weak and the usually used constant decay rate $\lambda f(\omega_0)$ and $J(\omega_0) = 0$ can be applied as good approximations in a relatively large frequency interval around ω_0.

4. Conclusion

The following conclusions may be drawn. Here, we have presented a *complete self-consistent gauge-invariant treatment of the single-atom spontaneous emission problem* (including unambiguous renormalization) within the scope of non-relativistic QED. For the first time to our knowledge, by taking into account *virtual transitions* and without ignoring *non-Markovian, retardation* and *electron-spin effects, a gauge-invariant analytic result for the whole time evolution* of the probability amplitude $b_{2P}(t)$ for finding the atom in the excited state $|2P\rangle$ has been derived. Moreover, in the BA it has been shown that, after introducing the physical bound-electron interaction picture, the term A^2 does not contribute at all, since, in the BA, its contribution is the same for both bound- and free-electron cases.

The comparison of our results with those of previous papers treating the same problem, without investigating the gauge invariance (see [1–6] and references quoted therein), shows that, instead of the *incomplete real-transition frequency shift* $\Delta\omega_0^{RT}$ (appearing in these papers), our present expressions for $b_{2P}(t)$ (see equations (3.13)–(3.16)) and the radiative line shape (see equation (3.24)) contain, for the first time, *the correct (complete) finite non-relativistic frequency shift* $\Delta\omega_0$, which agrees fairly good with the experiment [19]. The comparison with the work of authors who treated only the radiative line shape [7, 8], without investigating the time evolution of the decaying state, shows that the *non-Markovian effects* (missing in [7, 8]) led to a *quasi-Lorentzian frequency spectrum* in our equation (3.24), where both the decay rate as well as the frequency shift depend on the frequency ω.

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Appendix. Proof of gauge independence of expression $\Delta\tilde{E}^{\mathrm{VT(unr)}}_{nlm}$

In this appendix we prove the gauge-independence of the expression $\Delta E^{\mathrm{VT(unr)}}_{nlm}$ appearing in equation (2.30). It holds that

$$\Delta E \equiv \Delta\tilde{E}^{\mathrm{VT(unr)}}_{nlm} - \Delta E^{\mathrm{VT(unr)}}_{nlm}$$
$$= (\Delta\tilde{E}^{\mathrm{VT},1}_{nlm} - \Delta E^{\mathrm{VT},1}_{nlm}) + (\Delta\tilde{E}^{\mathrm{VT},2}_{nlm} - \Delta E^{\mathrm{VT},2}_{nlm}) \tag{A1}$$

$$\Delta\tilde{E}^{\mathrm{VT},1}_{nlm} = \langle\psi(0)|\tilde{H}^{\mathrm{VT}}_a|\psi(0)\rangle \qquad |\psi(0)\rangle = |nlm, m_s, v\rangle \tag{A2}$$

$$\Delta E^{\mathrm{VT},1}_{nlm} = \langle\psi(0)|\frac{e^2}{2mc^2}A^2|\psi(0)\rangle \tag{A3}$$

$$\Delta\tilde{E}^{\mathrm{VT},2}_{nlm} = \lim_{\eta\to 0}\langle\psi(0)|\tilde{H}^{\mathrm{VT}}_b\frac{1}{E_n - H^0 + \mathrm{i}\eta}\tilde{H}^{\mathrm{VT}}_b|\psi(0)\rangle \tag{A4}$$

$$\Delta E^{\mathrm{VT},2}_{nlm} = \lim_{\eta\to 0}\langle\psi(0)|H^{\mathrm{VT}}_b\frac{1}{E_n - H^0 + \mathrm{i}\eta}H^{\mathrm{VT}}_b|\psi(0)\rangle \tag{A5}$$

$$\tilde{H}^{\mathrm{VT}}_a = \mathcal{P}\left\{\frac{e^2}{2mc^2}A^2 + \frac{\mathrm{i}}{c\hbar}e[\chi, H^{\mathrm{VT}}] - \frac{1}{2c^2\hbar^2}e^2[\chi,[\chi,H^0]]\right\}\mathcal{P} \tag{A6}$$

$$\tilde{H}^{\mathrm{VT}}_b = \mathcal{P}\{H^{\mathrm{VT}} + \frac{\mathrm{i}}{c\hbar}e[\chi, H^0]\}(I^S_{\mathrm{VT}} - \mathcal{P}) + \mathrm{HC} \tag{A7}$$

$$H^{\mathrm{VT}}_b = \mathcal{P}H^{\mathrm{VT}}(I^S_{\mathrm{VT}} - \mathcal{P}) + \mathrm{HC}. \tag{A8}$$

By inserting equations (A6)–(A8) into equations (A2) and (A4), equation (A1) reduces to

$$\Delta E = \lim_{\eta\to 0}\langle\psi(0)|\left\{\frac{\mathrm{i}}{c\hbar}e[\chi, H^{\mathrm{VT}}] - \frac{1}{2c^2\hbar^2}e^2[\chi,[\chi, H^0 - E_n - \mathrm{i}\eta]]\right\}|\psi(0)\rangle$$
$$+ \lim_{\eta\to 0}\langle\psi(0)|H^{\mathrm{VT}}\frac{1}{E_n - H^0 + \mathrm{i}\eta}\frac{\mathrm{i}}{c\hbar}e[\chi, H^0 - E_n - \mathrm{i}\eta]|\psi(0)\rangle$$
$$+ \lim_{\eta\to 0}\langle\psi(0)|\frac{\mathrm{i}}{c\hbar}e[\chi, H^0 - E_n - \mathrm{i}\eta]\frac{1}{E_n - H^0 + \mathrm{i}\eta}H^{\mathrm{VT}}|\psi(0)\rangle$$
$$+ \lim_{\eta\to 0}\langle\psi(0)|\frac{\mathrm{i}}{c\hbar}e[\chi, H^0 - E_n - \mathrm{i}\eta]\frac{1}{E_n - H^0 + \mathrm{i}\eta}\frac{\mathrm{i}}{c\hbar}e[\chi, H^0 - E_n - \mathrm{i}\eta]|\psi(0)\rangle \tag{A9}$$

where we used the identity

$$[\chi, H^0] = [\chi, H^0 - E_n - \mathrm{i}\eta] \tag{A10}$$

Since, in equation (A9), whenever $H^0 - E_n - \mathrm{i}\eta$ acts on $|\psi(0)\rangle$, we can replace it by zero in the limit $\eta \to 0$, it follows immediately that

$$\Delta E = 0. \tag{A11}$$

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