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Stabilization of Positronium by Laser Fields

by

Antonella Karlson

A dissertation submitted to the Graduate Faculty in Physics
in partial fulfillment of the requirements for the degree of
Doctor of Philosophy, The City University of New York.

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Abstract

Stabilization of Positronium by Laser Fields

by

Antonella Karlson

Advisor: Professor M.H. Mittleman

We consider the problem of a positronium atom interacting with laser fields with the goal to extend its lifetime. Ps is an unstable system. Singlet Ps annihilates mainly by emission of two γ quanta and the triplet state - mainly by 3γ emission. The lifetimes are respectively $\tau_{an}^{s}= 1.25\ 10^{-10}$ sec and $\tau_{an}^{tr}=1.4\ 10^{-7}$ sec. The annihilation reaction is caused by an interaction term in the Hamiltonian, which on the scale of the Ps atom is essentially a zero range operator. Thus, the annihilation rate is proportional to the

absolute value squared of the Ps wave function at the origin. Since the wave function of Ps vanishes at the origin for all but states with angular momentum zero, Ps annihilates for all practicle purposes only from S states. If lasers are used to keep Ps out of them, the annihilation lifetime can be increased. Singlet and triplet Ps are not coupled by the lasers and they are discussed separately. A Ps atom interacting with laser fields is in a dressed state. Its wave function can be represented as a linear superposition of the bare states and under certain conditions it does not contain the ground state. This is the phenomenon of coherent population trapping. The latter can be used to extend the lifetime of Ps when the atom is driven by two semiclassical lasers.

We consider a two and three state Ps atom interacting respectively with one and two near-resonant circularly polarized lasers with spontaneous radiative transitions, annihilation from the ground state and photoionization from the excited states. The last two processes lead to the destruction of Ps and they are included via an antihermitian (absorptive) term in the Hamiltonian. The new annihilation rate depends on the initial Ps state, on the choice of bare states coupled by the laser(s) and on the laser parameters (intensity and detuning).

We obtain that the lifetime of triplet Ps in the field of two lasers cannot

be extended more than two times its value without the interaction with the lasers. This is due to spontaneous radiative transitions, which are fast and the atom fluoresces many times before annihilation. Therefore the effect of coherent population trapping is elliminated. The lifetime of singlet Ps can be extended with a small probability. If the initial state was 1S - up to 45 times its value outside of the lasers. If the initial state was 2P - almost twice the lifetime of this state outside of the lasers.

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I would like to thank my adviser Prof. M.H. Mittleman

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Chapter 1

Introduction

1.1 The positronium atom

Positronium (Ps) is the bound state of an electron and a positron. The possibility of its existence was first suggested by Mohorovicic [1] in 1934 and was first observed experimentally by Deutsch [2] in 1951. Its Bohr spectroscopic structure is equivalent to that of the hydrogen atom, but with each energy level being half of the hydrogen one, because the reduced mass is $m = \frac{1}{2}m_e$. The latter also causes the Ps distance scale to be twice that in hydrogen $a_0 = 2a_B$, where a_B is the Bohr radius. The ground state of Ps is composed of three triplet spin states of total spin one and a spin-zero singlet state.

These states are split by the usual spin-spin hyperfine interaction, which in Ps is larger than in hydrogen. Although the similarities between Ps and hydrogen are interesting, the primary reasons for studying Ps are its differences from hydrogen rather than its similarities to it. The most important of these differences and their physical significance are outlined below [3,4].

a. Annihilation properties.

Since the electron and the positron are a particle-antiparticle pair, Ps is an unstable system. Singlet Ps annihilates mainly by emission of two γ quanta and the triplet state - mainly by 3γ emission. These selection rules are due to angular momentum conservation and charge conjugation invarience. The lifetimes are respectively $\tau_{an}^{s}= 1.25\ 10^{-10}$ sec and $\tau_{an}^{tr}=1.4\ 10^{-7}$ sec. The annihilation reaction is caused by a quantum electrodynamical interaction term in the Hamiltonian, whose range is of the order of the Compton wavelength λ_c. On the scale of the Ps atom this is essentially a zero range operator. Thus, the decay rate is proportional to the absolute value squared of the Ps wave function at the origin, where the two particles are in contact. Since the wave function of Ps vanishes at the origin for all but states with angular momentum zero, Ps annihilates for all practicle purposes only from S states. For them the annihilation rate depends on the principal quantum number as n^{-3}

[4,5]. For states with higher angular momentum l, the annihilation rate is smaller than the rate for the respective S state by a factor of $(\frac{a_B}{\lambda_c})^{-2l} \approx {\alpha_F}^{-2l}$, where α_F is the fine structure constant. Therefore annihilation from these states is negligible.

b. Purely leptonic system.

Consequently high precision measurements of the decay rates, fine and hyperfine structure splittings provide unambiguois and stringent tests on quantum electrodynamical predictions. The difficulties of atomic structure calculations present in normal atoms are not relevant for Ps.

c. Other particle-antiparticle properties.

Ps can be used to study various symmetry principles such as charge conjugation and intrinsic electron-positron parity. The search for the annihilation of singlet and triplet Ps into γ-ray decay schemes forbidden by some symmetry principles can be used to set upper limits on the validity of these symmetries. Alternatively, such limits can be used to search for types of decay not included in the usual QED framework. In addition, the measurements of conventional channel annihilation rates, as well as fine and hyperfine structure splittings indirectly test CPT, since it is assumed in all calculations that the masses $m_{e+} = m_{e-}$ and the magnetic moments $|\mu_{e+}| = |\mu_{e-}|$ are

equal. The particle-antiparticle nature of the system also makes possible the investigation of virtual self-annihilation effects.

d. Atom with a 'light' nucleus.

A number of interesting effects related to the two-body formulation of quantum electrodynamics (the Bethe-Salpeter [6] or the Lepage [7] approach) can be tested.

Some other Ps related experiments [4] are the measurement of the Compton wavelength [8], precision positron polarimetry [9], γ-ray astronomy [10] and Ps formation in optically active substances [11].

Ps is produced in three different ways []. The standard technique is formation of Ps in gases [12]. Positrons of typically several hundred kilovolts energy emitted from a radioactive source slow down in a gas and eventually pick up an electron to form Ps. As many as 25-50% of the incoming positrons form Ps atoms, typically within 10^{-10}s of positron emission. The need for Ps formation in a background gas presents a problem in high-precision Ps experiments. Another technique is to make a positron beam incident on a variety of small-grained, low-density powders [13]. Up to 30% of the positrons can form Ps. The deviation of the decay rates and hyperfine structure splittings of Ps produced in this way from their presumed vacuum values are much

smaller than in gases. A third technique [14,15] leads to the confinement of Ps in vacuum and is a great improvement over the formation of Ps in gases and in powders. When positrons of a radioactive source are incident on various types of surfaces, up to one in 10^7 emerges at an energy of 1 eV. The efficiency can be improved up to 10^{-3} slow positrons per incident fast particle. Slow positron beams of typically $10^4 - 10^6$ e^+/sec are now available. Then the positrons are accelerated to several hundred eV in order to achieve optimal cross-section and are made incident on various surfaces (Au, Ti, Cu, etc.). Up to 80% of them form Ps, which subsequently leaves the target and enters the surrounding vacuum region. In this way it is also possible to produce Ps atoms with thermal energies, which is very convenient for experiments.

Currently Ps atoms are produced predominantly in the n=1 ground state and subsequently annihilate fast. Only about one in $10^3 - 10^4$ atoms is found in the n=2 upper state [15-17] which has a longer lifetime. One way of populating the excited states of atoms is the method of optical pumping. Therefore lasers can be used to keep Ps out of S states, in particular the 1S ground state, which will result in a significant increase of the annihilation lifetime. This leads to the study of the excitation probability of Ps atoms by

resonant laser fields [18,19].

An atom in a laser field is distorted or dressed by the field. Lasers can couple the ground state of Ps to one or more excited states. The new wave function of Ps in the laser field is altered and can be represented as a linear combination of the bare atomic states. This results in a longer annihilation lifetime of the dressed state in comparison to the bare ground state. The lifetime depends on the laser parameters (intensity and detuning) on one hand and on spontaneous radiative transitions on the other hand. The latter repopulate the 1S state and lead to annihilation. The contribution of photoionization to Ps destruction becomes significant at high laser intensities and has to be considered as well. Since the laser will essentially not couple the singlet and the triplet manifolds, they can be considered separately.

Typical fluorescent lifetimes are of the order of 10^{-8} sec, which is much longer than the singlet annihilation lifetime outside of the laser and shorter than the triplet one. If, due to the interaction with the laser, the singlet lifetime is increased up to but less than the characteristic lifetimes of the spontaneous radiative transitions, the latter can be neglected. For triplet Ps fluorescence has to be always included in the considerations.

Our goal is to obtain the lifetime of singlet and triplet Ps in the field of one

and two weak near resonant circularly polarized lasers, taking into consideration in an appropriate way annihilation, spontaneous radiative transitions and photoionization. Due to coherent population trapping (see Section 1.3 below), we expect a longer lifetime for a Ps atom driven by two lasers. Since the problem with one laser is relatively simpler, we need to solve it first, and then with the experience gained consider the two laser case.

1.2 A two state Ps atom interacting with one near-resonant laser field

We briefly discuss the case of one circularly polarized laser resonant with the 1S - 2P transition[1] neglecting all Ps destruction mechanisms. The laser field is treated semiclassically. For a detailed analysis of the problem see [20] (Chapter 2 and 4).

The dressed states of Ps interacting with the laser field, obtained in the two state rotating wave approximation (RWA) are:

$$|\phi_{\pm}\rangle \;=\; (2ch\mu)^{-1/2}\,(e^{\pm\mu/2}\,|u_0\rangle\,e^{i\omega t/2} \;\pm\; e^{\mp\mu/2}\,|u_1\rangle\,e^{-i\omega t/2})$$

[1]See Figure 1.1.

$$\times exp[-i(W_0 + W_1 \pm \varepsilon)\frac{t}{2\hbar}], \qquad (1.1)$$

where $|u_0\rangle$ and $|u_1\rangle$ are the 1S and 2P bare atomic states respectively. W_0 and W_1 are their eigenenergies. The laser frequency is ω, and the detuning parameter μ is defined as:

$$sh\mu = \frac{\hbar\Delta\omega}{\Lambda}. \qquad (1.2)$$

Here

$$\Delta\omega = \omega - W_{10}/\hbar \quad \text{and} \quad W_{10} = W_1 - W_0. \qquad (1.3)$$

Λ is the coupling matrix element for the one photon transition:

$$\Lambda = \frac{eE}{m\omega}\,\hat{\varepsilon}\cdot\langle u_1|\vec{p}|u_0\rangle \qquad (1.4)$$

where E is the amplitude of the laser electric field, $\hat{\varepsilon}$ is its polarization[2] and $m = 0.5m_e$ is the reduced mass. The Rabi frequency is

$$\varepsilon = \sqrt{\Delta\omega^2\hbar^2 + |\Lambda|^2}. \qquad (1.5)$$

Corrections to the dressed states in eq. (1.1) are due to the neglected states and to the RWA. They are of the same order of magnitude and are small.

In the rest frame of the atom the laser will be switched on adiabatically so that only one of the dressed states will be realized. Since Ps is usually in

[2]The laser is circularly polarized.

its ground state before the laser is switched on, this will be $|\Phi_+\rangle$ for $\Delta\omega > 0$ and $|\Phi_-\rangle$ for $\Delta\omega < 0$. For singlet Ps the annihilation will occur from one of these states and the rate is proportional to $|\Phi_\pm(\vec{r}=0)|^2$. The result is:

$$\frac{\Gamma}{\Gamma_0} = \frac{e^{|\mu|}}{2ch\mu} = \frac{\varepsilon + |\Delta\omega|\hbar}{2\varepsilon} \rightarrow \frac{1}{2}, \tag{1.6}$$

where the limit is taken for $\frac{\Delta\omega}{\varepsilon} \rightarrow 0$. Here $\Gamma_0 = \hbar/\tau_{an}$ is the decay rate in the absence of the laser. Therefore we obtain that the annihilation lifetime can be increased up to a factor of two, which occurs at resonance. For large detunings (compared to Λ) it is not affected by the laser.

It is necessary to note that the Rabi frequency must be larger than the annihilation rate in order to populate the excited state before annihilation, i.e. $\varepsilon \approx \Lambda > \Gamma_0$, where

$$\begin{aligned} \Gamma_0 &= 1.7\ 10^{-10}\ [\text{Ht}] \quad \text{for triplet Ps} \\ \Gamma_0 &= 1.9\ 10^{-7}\ [\text{Ht}] \quad \text{for singlet Ps.} \end{aligned} \tag{1.7}$$

From here we can estimate the laser intensities. We obtain for the triplet $I > 2.0\ \ W/cm^2$ and for the singlet $I > 3.2\ 10^2\ W/cm^2$. In order to compare we use a convenient atomic unit for the electric field:

$$E_0 = \frac{e}{a_B^2} \approx 5.14\ 10^9\ V/cm \quad \text{and} \quad I_0 = \frac{cE_0^2}{8\pi} = 3.52\ 10^{16}\ W/cm^2 \tag{1.8}$$

Here e is the electron charge and a_B is the Bohr radius. This shows that we need lasers, whose fields are weak compared to the ones present in the atom. Therefore the approximations and the formalism needed to solve this problem should be appropriate for weak near resonant lasers.

For triplet Ps the state 2P decays radiatively to 1S. In the absence of the laser the lifetime[3] is $\tau_{10} = 1.236\ 10^{-9}s$. This is much less than the triplet annihilation lifetime. Therefore fluorescence has to be included into the problem. The atom is no longer described by the state $|\Phi_{\pm}\rangle$, but by a superposition determined by the fluorescence process. We use a procedure originally given by Mollow [21] to describe this process. [4] The decay rate is proportional to $\delta(\vec{r})\,\langle\Psi|P_0|\Psi\rangle$, where $|\Psi\rangle$ is the wave function of the system and P_0 projects onto the state $|u_0\rangle$. The formalism used to obtain this expectation value is discussed in some detail for the three level atom in Chapter 2. The result for the two state case as given in [18] is:

$$\frac{\Gamma}{\Gamma_0} = \frac{1}{2}\left(1 + \frac{2(\gamma^2 + \Delta\omega^2)}{2\gamma^2 + |\Lambda|^2 + 2\Delta\omega^2}\right) \tag{1.9}$$

where γ is half of the natural radiative decay rate for the 2P$\rightarrow$1S transition.

[3]See Appendix A.

[4]See also [20], Chapter 4.

When the laser frequency is near the 1S - 2P transition, the parameters $\Delta\omega$ and Λ are defined as in the singlet case. At resonance the annihilation rate is reduced by a factor:

$$\frac{1}{2}\,\frac{4\gamma^2+|\Lambda|^2}{2\gamma^2+|\Lambda|^2} \approx \frac{1}{2}\left(1+\frac{2\gamma^2}{|\Lambda|^2}\right) + O\left(\frac{\gamma^3}{|\Lambda|^3}\right) \tag{1.10}$$

and is unaffected for large detunings $\Delta\omega$. For a strong enough laser so that the inequality $\Lambda >> \gamma$ holds, we obtain that the lifetime of triplet Ps is increased by a factor of two, just as in the singlet case.

1.3 Coherent population trapping in a three state positronium atom

The problem of coherent population trapping and more generally of dynamic symmetry when a multilevel quantum system is irradiated by lasers has been studied extensively in recent years [22-27] (and references therein). Dynamic symmetry implies, in addition to the conservation of the total atomic population, one or more other constants of the evolution in the dynamics of the system. The problem is to find such invariants and the conditions under which the system would have them. Most of the existing studies discuss co-

herent population trapping in the context of a three level 'Λ' system driven by two semiclassical lasers. The trapping states of a 'Λ' system driven by quantized fields have been very recently discovered [28]. Two-level systems are not known to exibit trapping states, except when the two-level atom is interacting with a quantized field [29] or with a frequency modulated semiclassical field [30].

In view of the above, coherent population trapping can be used to prolong the lifetime of Ps in the case of a three-level system. We consider Ps atoms in the field of two different circularly polarized lasers. Each of them is nearly resonant with a given transition between the ground state and an excited Ps state. In principle any two excited states which obey the selection rules can be chosen. But spontaneous radiative transitions can occur between them and other Ps states that are not coupled by the lasers. This would lead to a multilevel problem, which is considerably more difficult (and probably gives less enhancement of the lifetime), without introducing any new physics. Therefore it is necessary to select a simple three state system wich is closed with respect to fluorescence. This can be achieved by the following configuration: laser 'a' couples $1S(m_l = 0)$ with $3D(m_l = \pm 2)$ via a two photon transition and laser 'b' couples $1S(m_l = 0)$ with $2P(m_l = \pm 1)$ via a single

photon transition (see Figure 1.2). The laser detunings are respectively:

$$\Delta\omega_a = 2\omega_a - W_{20} \quad \text{where} \quad W_{20} = W_2 - W_0 \tag{1.11}$$

and

$$\Delta\omega_b = \omega_b - W_{10} \quad \text{where} \quad W_{10} = W_1 - W_0 \tag{1.12}$$

This configuration of Ps states and laser fields resembles an inverted 'Λ' system (or a 'V' system). It has the property of coherent population trapping at resonance, which means in this case that $\Delta\omega_a = \Delta\omega_b$. The three dressed Ps states close to resonance are found in Chapter 2, section 1.[5] One of them has no 1S component. This state would not annihilate at all were it not for the fact that spontaneous radiative transitions 3D→2P→1S occur.[6] They repopulate the ground state and permit annihilation. The controlling lifetime will be that of the 2P→1S transition, which is $\tau_{10} = 3.2\ 10^{-9}$sec. The 3D→2P transition is much slower with a lifetime of $\tau_{21} = 3.1\ 10^{-8}$sec.[7] The ratio $\tau_{10}/\tau_{an}^{s} \approx 25$ and therefore we anticipate an extension of the lifetime of singlet Ps of about 25 times. On the other hand the lifetime of triplet Ps is

[5] See eq. (2.18) and (2.20).

[6] Radiative transitions to other Ps states are forbidden by the selection rules.

[7] These lifetimes are derived in Appendix A.

longer than these fluorescent decay times and it is not clear beforehand to what extend it is influenced by coherent trapping.

1.4 Research program

The Hamiltonian of a Ps atom interacting with laser fields is as follows:

$$H = H_0 + H_A + H_R + H_D \tag{1.13}$$

where H_0 is the Hamiltonian of the Ps atom outside of the laser (a hydrogen atom Hamiltonian with a reduced mass $m = \frac{m_e}{2}$). H_A gives the interaction between the Ps atom and the laser field. Since photoionization by the laser is also possible and this is another mechanism for Ps destruction, here we need to include this process as well. H_R is the interaction Hamiltonian of Ps with the quantized electromagnetic field resulting from spontaneous radiation. H_D gives the annihilation of Ps from the ground state into high energy photons. Due to angular momentum conservation, annihilation of singlet Ps results in the emission of two gamma quanta and of triplet Ps - in three gamma quanta [3].

We need to estimate the relative magnitudes of the fluorescent and the

annihilation interactions. For the 2P→1S radiative transition we obtain that $\frac{\hbar}{\tau_{10}} \equiv 2\gamma_{10} = 7.586\ 10^{-9}$ [Ht], and for the 3D→2P transition - $\frac{\hbar}{\tau_{21}} \equiv 2\gamma_{21} = 7.832\ 10^{-10}$ [Ht]. We compare with the annihilation rate of Ps in eq. (1.7). In order to treat the annihilation interaction perturbatively, it must be smaller than the interaction of the atom with the radiation field resulting from fluorescence. In other words, we must have that $\Gamma_0 < \frac{\hbar}{\tau_{21}}$. This condition holds only for the triplet. Therefore we have to consider H_D nonperturbatively for singlet Ps and we can consider it up to first order in perturbation theory for triplet Ps.

It is necessary to note that the detunings $\Delta\omega_a$ and $\Delta\omega_b$ must be smaller than the width of the corresponding excited states, so that the lasers are resonant. In addition to the natural width of the states there are also induced widths due to the interaction with the lasers. This is known as power broadening.

In Chapter 2 we consider Ps atoms in the field of two near-resonant laser when photoionization is not present. In the first section we find the dressed states and discuss in detail the case very close to resonance. We investigate the influence of the initial Ps state on the wave function of the system in the lasers. We comment on the approximations used. In section 2

we include spontaneous radiative transitions. We find the expectation values of the basis set of matrices with respect to the complete wave function of the system. The occupation probabilities of the bare states for triplet Ps are found numerically for different values of the laser intensities. Considering the annihilation interaction perturbatively up to first order, we find the decay rate of triplet Ps.

In Chapter 3 we discuss singlet Ps in the field of one near-resonant laser. The Hilbert space of the system is extended to include the continuum states resulting from photoionization and the high energy gamma quanta from annihilation. We obtain a system of differential equation, which is solved with the Laplace transformation in the pole approximation. We seek the optimal laser intensity, for which the destruction of Ps is minimized.

In Chapter 4 we consider singlet Ps in the field of two near-resonant laser without including spontaneous radiative transitions. Photoionization and annihilation are considered nonperturbatively by introducing a complex potential in the Hamiltonian. We derive an expression for the probability to find singlet Ps for different initial conditions. We obtain an effective time-dependent decay rate as a function of the parameters in the problem.

In Chapter 5 we discuss triplet Ps in the field of one near-resonant laser.

Ionization and annihilation are included again via a complex potential. We investigate the influence of these terms on the Rabi frequency. The probability to find Ps is found analytically at resonance and in powers of γ/Λ.

In Chapter 6 we consider the complete physical problem of Ps in the field of two near-resonant lasers. Photoionization and annihilation are included by a nonhermitian term in the Hamiltonian. Spontaneous radiative transitions are treated with the formalism introduced in Chapter 2, section 1. This method allows to consider singlet and triplet Ps in a unified way taking into account all the relevant interactions. We find a system of nine coupled differential equations for the expectation values of the basis set of matrices with respect to the complete wave function of the system. One of them gives the probability to find Ps as a function of time. In section 2 we solve this system numerically for triplet Ps. We find the probability and the decay rate for different values of the laser intensities and detunings. In section 3 we solve for the probability of singlet Ps. We also solve some limiting cases in order to investigate the influence of photoionization and fluorescence. The probability and the lifetime are found for different values of the laser intensities and for different initial state.

In Appendix A we derive the fluorescence and single-photon ionization

rates. The matrix element of the two-photon laser induced transition $1S \leftrightarrow 3D$ and the two-photon ionization rate from the ground state are derived in Appendix B. The constraint on the wave function of the system used to include fluorescence is derived in Appendix C.

1.5 Numerical estimate of the critical laser intensities and of photoionization rates

In what follows, if not stated explicitely, all numerical values and most of the formulas are given in units $\hbar = c = 1$. In this system of units the dimension of all physical quantities is energy or inverse energy. We set the energy scale to be $1\,Ht = 27.2\,eV$.

We mentioned in section 2, while considering the two state problem that the Rabi frequency must be larger than the annihilation rate (1.7) in order to populate the excited state before annihilation. We denote with Λ_a the coupling matrix element for the two photon transition from the ground state to the 3D excited state. Λ_b is the coupling matrix element (1.4) for the one photon transition from 1S to 2P. The critical values of the laser fields $E_{crit}^{a,b}$

are determined by the condition:

$$\Lambda_{a,b}(E^{a,b} = E^{a,b}_{crit}) \;=\; \Gamma_0. \tag{1.14}$$

In order to proceed, the numerical values of the coupling matrix elements have to be computed. From eq. (1.4) we obtain[8]

$$\Lambda_b = 1.492\ \frac{E_b}{E_0}\ [\mathrm{Ht}], \tag{1.15}$$

where E_0 is the atomic electric field defined in eq. (1.8). From the condition (1.14) we obtain the critical value of the 'b' laser field:

$$\begin{aligned} E^b_{crit} &= E_0\ 1.14\ 10^{-10} \quad \text{for triplet Ps} \\ E^b_{crit} &= E_0\ 1.27\ 10^{-7} \quad \text{for singlet Ps.} \end{aligned} \tag{1.16}$$

These values are given in section 1.2. We can now express Λ_b in terms of the critical fields, which is convenient for further calculations:

$$\begin{aligned} \Lambda_b &= 1.7\ 10^{-10} \left(\frac{E^b}{E^b_{crit}}\right) [\mathrm{Ht}] \quad \text{for triplet Ps} \\ \Lambda_b &= 1.9\ 10^{-7} \left(\frac{E^b}{E^b_{crit}}\right) [\mathrm{Ht}] \quad \text{for singlet Ps.} \end{aligned} \tag{1.17}$$

Λ_a is more difficult to handle since it is a two photon matrix element:

$$\Lambda_a \;=\; e^2 E_a^2\ \langle u_2 | \hat{\epsilon}^* \cdot \vec{r} | \phi(\vec{r}) \rangle \tag{1.18}$$

[8] A detailed calculation is given in Appendix A.

and

$$|\phi(\vec{r})\rangle = \sum_n \frac{|u_n\rangle\langle u_n|\hat{\varepsilon}^* \cdot \vec{r}|u_0\rangle}{W_n - \omega_a - W_0}, \tag{1.19}$$

where the sum runs over all possible intermediate states $|u_n\rangle$ except 2P.[9] We can perform this sum without approximations by using the method of inhomogeneous differential equations which is described in [31-33] (Chapter 4).[10] A detailed calculation of this matrix element is given in Appendix B. We obtain:

$$\Lambda_a \;=\; 28.81 \left(\frac{E_a}{E_0}\right)^2 \text{[Ht]}. \tag{1.20}$$

Therefore the critical values of the laser electric field in this case are:

$$\begin{aligned} E^a_{crit} &= E_0\; 2.43\, 10^{-6} \quad \text{for triplet Ps} \\ E^a_{crit} &= E_0\; 8.12\, 10^{-5} \quad \text{for singlet Ps.} \end{aligned} \tag{1.21}$$

They are considerably larger than the ones for laser 'b' in eq. (1.16), because Λ_a is quadratic in the laser electric field. It is again convenient for further calculations to express Λ_a in terms of the critical fields:

$$\Lambda_a \;=\; 1.7\, 10^{-10} \left(\frac{E^a}{E^a_{crit}}\right)^2 \text{[Ht]} \quad \text{for triplet Ps}$$

[9] If we were to use opposite circular polarizations for laser 'a' and 'b', then the state 2P is also included in the sum.

[10] See also references given in [31].

$$\Lambda_a = 1.9\,10^{-7}\left(\frac{E^a}{E^a_{crit}}\right)^2 \text{[Ht]} \quad \text{for singlet Ps.} \tag{1.22}$$

This demonstrates again that the interaction energy of the Ps atom with the lasers, i.e. Λ_b in (1.17) and Λ_a in (1.22) is much smaller in comparison to the interaction energy in the bare atom. In comparison, the energy levels in Ps are:

$$\begin{aligned} W_{10} &= \frac{3}{8}\frac{e^2}{2a_B} = 0.1875\ \text{[Ht]} \\ W_{20} &= \frac{4}{9}\frac{e^2}{2a_B} = 0.2222\ \text{[Ht]} \\ W_{21} &= \frac{5}{72}\frac{e^2}{2a_B} = 0.0347\ \text{[Ht].} \end{aligned} \tag{1.23}$$

With the critical fields (1.16) and (1.21) we can now estimate the values of photoionization rates. In principle there are five possible ionization processes. Laser 'a' can ionize the Ps atom either from state 2P or from 3D. The same holds for laser 'b'.[11] It is also possible to induce a nonresonant transition from 1S into continuum states by a two photon process first with laser 'a' and then with 'b'. The two photon ionization from the ground state when the order of the lasers is reversed is resonant and is included in our previous considerations. We compute the ionization rates in perturbation theory to

[11] See Figure 1.2.

lowest order. For the radial part of the continuum wave function we take the Coulomb function with ingoing wave boundary conditions [34] (Chapter 14), [35] (Chapter 1.3) and [36]. The integrals in the one photon ionization rates can be performed analytically. A detailed derivation of all ionization rates is given in Appendix A. We obtain the following numerical values:

- ionization from 2P with laser 'a':

$$\begin{aligned} w^a_{2P} &= \left(\frac{E^a}{E^a_{crit}}\right)^2 7.637\,10^{-12}\ [\mathrm{Ht}] \quad \text{for triplet Ps} \\ w^a_{2P} &= \left(\frac{E^a}{E^a_{crit}}\right)^2 8.528\,10^{-9}\ [\mathrm{Ht}] \quad \text{for singlet Ps.} \end{aligned} \tag{1.24}$$

- from 3D with laser 'a':

$$\begin{aligned} w^a_{3D} &= \left(\frac{E^a}{E^a_{crit}}\right)^2 2.915\,10^{-12}\ [\mathrm{Ht}] \quad \text{for triplet Ps} \\ w^a_{3D} &= \left(\frac{E^a}{E^a_{crit}}\right)^2 3.255\,10^{-9}\ [\mathrm{Ht}] \quad \text{for singlet Ps.} \end{aligned} \tag{1.25}$$

- from 2P with laser 'b':

$$\begin{aligned} w^b_{2P} &= \left(\frac{E^b}{E^b_{crit}}\right)^2 2.961\,10^{-21}\ [\mathrm{Ht}] \quad \text{for triplet Ps} \\ w^b_{2P} &= \left(\frac{E^b}{E^b_{crit}}\right)^2 3.673\,10^{-15}\ [\mathrm{Ht}] \quad \text{for singlet Ps.} \end{aligned} \tag{1.26}$$

- from 3D with laser 'b':

$$w^b_{3D} = \left(\frac{E^b}{E^b_{crit}}\right)^2 1.355\,10^{-21}\ [\mathrm{Ht}] \quad \text{for triplet Ps}$$
$$w^b_{3D} = \left(\frac{E^b}{E^b_{crit}}\right)^2 1.681\,10^{-15}\ [\mathrm{Ht}] \quad \text{for singlet Ps.} \qquad (1.27)$$

We have represented the ionization rates in terms of the ratio of the laser field and the respective critical field. This is why the singlet and triplet ionization rates appear to be different. They are not, but the respective values of the critical fields are. This way of writing allows us to compare easily the above rates with the ones for annihilation in eq. (1.7) and estimate which of the photoionization processes are relevant. The intensity of laser 'a' (1.21) is much higher than the one of laser 'b' (1.16). This leads to relatively large ionization rates (1.24) and (1.25) for laser 'a', which are about 20-30 times smaller than those for annihilation, when the laser fields are equal to the critical ones. Therefore they can lead to an observable Ps destruction under certain conditions. On the other hand the ionization rates (1.26) and (1.27) for laser 'b' are orders of magnitude smaller than the respective annihilation rates and can be neglected.

The last possible ionization is the nonresonant two photon one from the ground state [37, 38]. Up to first order in perturbation theory the matrix

element for this transition is:

$$\Lambda_q \;=\; e^2 E_a E_b \,\langle u^-_{q,2}|\hat{\varepsilon}^* \cdot \vec{r}|\phi'(\vec{r})\rangle \tag{1.28}$$

and

$$|\phi'(\vec{r})\rangle = \sum_n \frac{|u_n\rangle\langle u_n|\hat{\varepsilon}^* \cdot \vec{r}|u_0\rangle}{W_n - \omega_a - W_0}, \tag{1.29}$$

where $|u^-_{q,2}\rangle$ is the energy normalized, with incoming wave boundary conditions, regular at the origin Coulomb function for unbounded states as defined in eq. (A.29). The sum runs over all possible intermediate states $|u_n\rangle$ except 2P.[12] It can be solved without any approximations with the inhomogeneous differential equation method [31-33] in a similar way as for Λ_a (see Appendix B). We obtain the following ionization rates:

$$\begin{aligned} w^{ab}_{1S} &= \left(\frac{E^a}{E^a_{crit}}\right)^2 \left(\frac{E^b}{E^b_{crit}}\right)^2 7.257\,10^{-33}\,[\mathrm{Ht}] \quad \text{for triplet Ps} \\ w^{ab}_{1S} &= \left(\frac{E^a}{E^a_{crit}}\right)^2 \left(\frac{E^b}{E^b_{crit}}\right)^2 1.010\,10^{-23}\,[\mathrm{Ht}] \quad \text{for singlet Ps.} \end{aligned} \tag{1.30}$$

Obviously these rates are too small in comparison to the annihilation rates to contribute to the destruction of Ps at the laser intensities of interest and this ionization process can be also neglected.

[12] Also in this case, if lasers 'a' and 'b' were to have opposite circular polarization, the state 2P has to be included as well.

To summarize the results of this section, we obtain that the relevant ionization rates are from the states 2P and 3D induced by laser 'a'. The rates for all other transitions to the continuum are too weak to result in an observable Ps destruction and need not be taken into consideration.

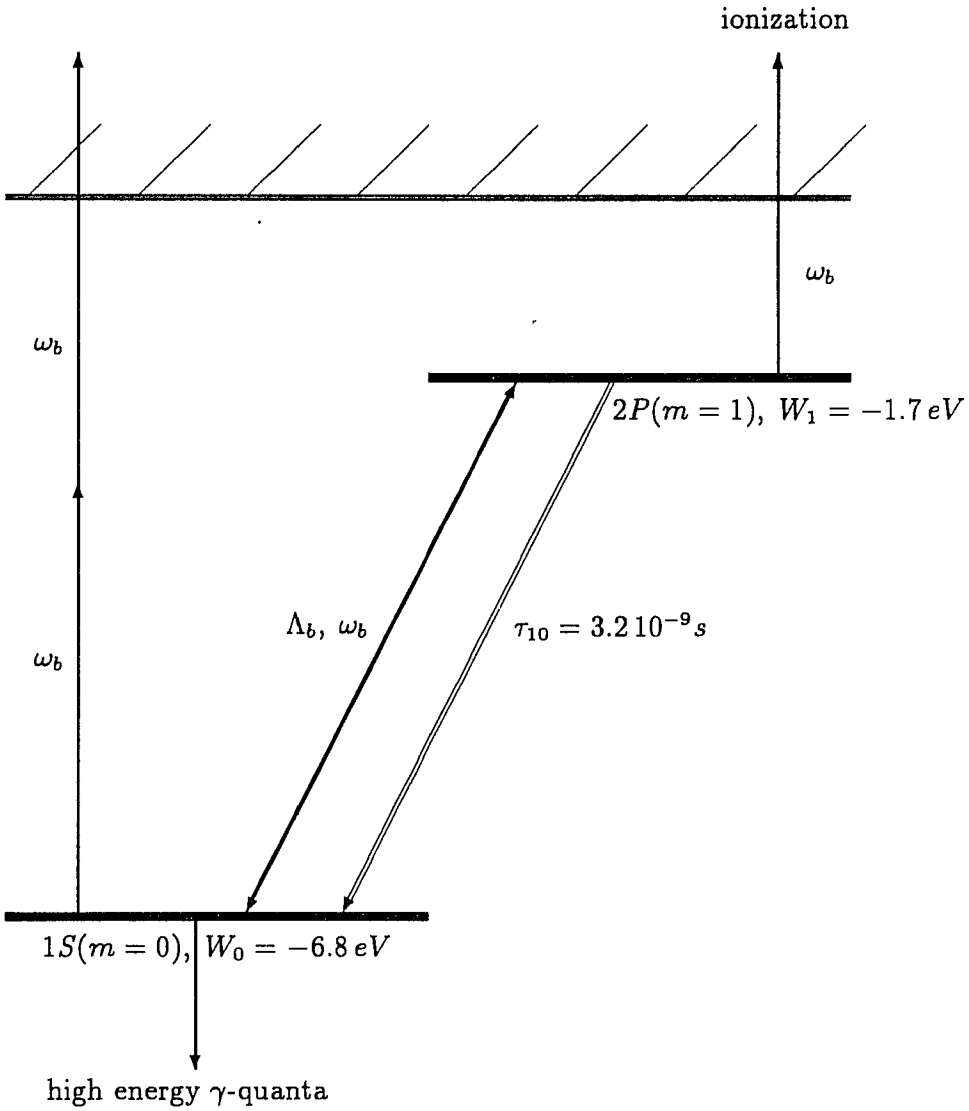

Figure 1.1: Energy level diagram of a Ps atom in the field of one near-resonant circularly polarized laser. The annihilation lifetimes are respectively $\tau_{an}^{s} = 1.25\,10^{-10}s$ and $\tau_{an}^{tr} = 1.4\,10^{-7}s$.

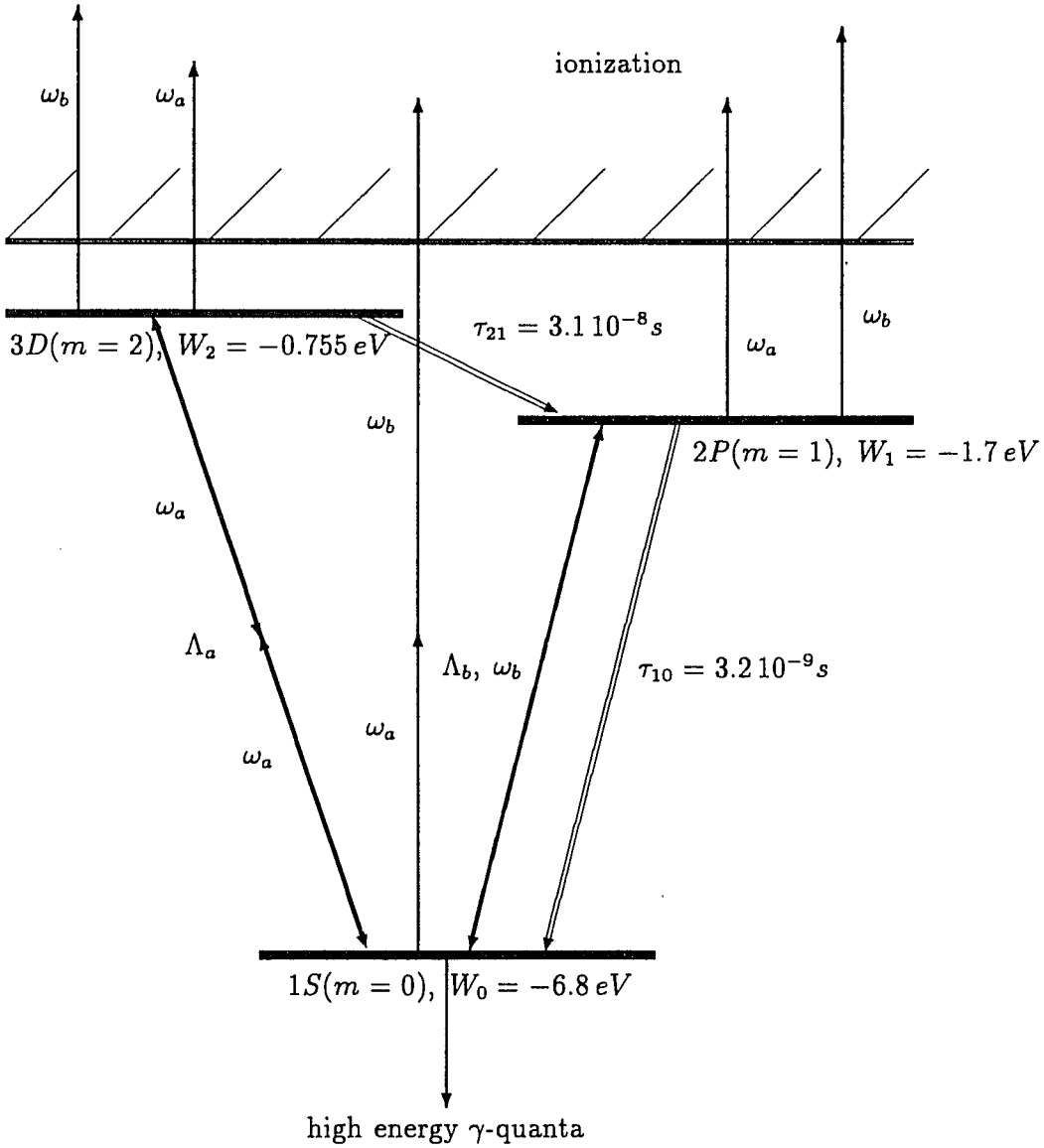

Figure 1.2: Energy level diagram of a Ps atom in the field of two near-resonant circularly polarized laser. The annihilation lifetimes are respectively $\tau_{an}^{s}=1.25\,10^{-10}s$ and $\tau_{an}^{tr}=1.4\,10^{-7}s$.

Chapter 2

Positronium atom in the field of two near-resonant lasers without photoionization

Photoionization is not included in this chapter. First we consider only the laser-atom interaction and find the wavefunctions which are the solutions of the Schrodinger equation. The results hold for singlet as well as for triplet Ps atoms. They will be used in Chapter 4 to find the lifetime of singlet Ps. Then we include fluorescence in the system by using a method first introduced by

Mollow in [21]. This enables us to find an expression for the lifetime of triplet Ps in the field of two lasers.

2.1 Dressed states

Since the laser is weak, it would seem that a straightforward perturbation theory in the interaction H_A is adequate. This is true except for the situation in which the laser is nearly resonant with a transition between a pair of bare atomic states. For example if the state $|u_0\rangle$ times some laser state is nearly degenerate with the state $|u_1\rangle$ with the same laser state less one photon, which is the case we have here, then it is necessary to use degenerate perturbation theory.

Our starting point is the Schrodinger equation:

$$i\frac{\partial}{\partial t}|\Psi(t)\rangle \;=\; (H_0 + H_A)|\Psi(t)\rangle. \tag{2.1}$$

The lasers are treated semiclassically and

$$H_A \;=\; \sum_{j=a,b} \frac{e}{m}\,\vec{A}_j(t)\cdot\vec{p} \tag{2.2}$$

is the interaction of the Ps atom with the two single mode lasers in the dipole approximation. The index j refers to laser 'a' and 'b' respectively.

See level diagram in Figure 1.2. We are working in the velocity gauge. The term quadratic in the vector potential is small and is not included in the Hamoltonian. Moreover it contributes nothing to matrix elements between different states of Ps in the dipole approximation. We have also performed a transformation to an interaction representation in which the time evolution due to the free Hamiltonian of the laser fields is absorbed into the wave function:

$$|\psi(t)\rangle \;=\; e^{-iH_{rad}\,t}\;|\Psi(t)\rangle \tag{2.3}$$

The vector potentials are:

$$\vec{A}_j(t) \;=\; \frac{E_j}{2\omega_j}\left(\hat{\varepsilon}_j\; e^{i\omega_j t} \;+\; \hat{\varepsilon}_j^{*}\; e^{-i\omega_j t}\right). \tag{2.4}$$

Both lasers are circularly polarized with polarization:

$$\hat{\varepsilon}_j \;=\; \frac{1}{2}(\hat{x}_j \pm i\hat{y}_j). \tag{2.5}$$

We assume that the wave function can be represented as a linear combination of the three orthonormal bare states:

$$\begin{aligned}|\Psi(t)\rangle \;=\;& \bar{\alpha}(t)\;|u_0\rangle\; e^{-iW_0 t} + \;\bar{\beta}(t)\;|u_1\rangle\; e^{-iW_1 t/} \\ &+ \bar{\gamma}(t)\;|u_2\rangle\; e^{-iW_2 t},\end{aligned} \tag{2.6}$$

with unknown time dependent amplitudes $\bar{\alpha}(t)$, $\bar{\beta}(t)$ and $\bar{\gamma}(t)$. The wave function $|\Psi(t)\rangle$ is substituted back into the Schrodinger equation. Then the latter is projected successively onto $|u_0\rangle$, $|u_1\rangle$ and $|u_2\rangle$ to obtain three coupled equations for the three unknown parameters.

$$\begin{aligned} i\bar{\alpha}'(t) &= \bar{\beta}(t)\,\frac{\Lambda_b}{2}e^{i\Delta\omega_b t} + \bar{\gamma}(t)\,\frac{\Lambda_a}{2}e^{i\Delta\omega_a t} \\ i\bar{\beta}'(t) &= \bar{\alpha}(t)\,\frac{\Lambda_b^*}{2}e^{-i\Delta\omega_b t} \\ i\bar{\gamma}'(t) &= \bar{\alpha}(t)\,\frac{\Lambda_a^*}{2}e^{-i\Delta\omega_a t} \end{aligned} \tag{2.7}$$

where $\Delta\omega_a$ and $\Delta\omega_b$ are defined in eq. (1.11) and (1.12) respectively. The one photon coupling matrix element Λ_b is definined in eq. (1.4) and calculated in Appendix A, while Λ_a is defined in eq. (1.18) and (1.19) and calculated in Appendix B. As described there, it is a second order matrix element because the transition $1S \rightarrow 3D$ is a two photon one via all possible intermediate states.

On the right hand side of this system of equations we have dropped some terms. Originally there are two kinds of exponential time dependencies, a slow one:

$$e^{\pm i\Delta\omega_a t} \quad \text{and} \quad e^{\pm i\Delta\omega_b t}, \tag{2.8}$$

and a fast one:

$$e^{\pm i(\Delta\omega_a - 2W_{20})t} \quad \text{and} \quad e^{\pm i(\Delta\omega_b - 2W_{10})t}. \tag{2.9}$$

The rapidly varying terms are assumed to average away and therefore make a small contribution to the equations and are dropped. This is the usual rotating wave approximation.

The system of differential equations (2.7) can be solved with the assumption:

$$\begin{aligned} \bar{\alpha}(t) &= \alpha\, e^{-i(\varepsilon - \Delta\omega_a)t/2} \\ \bar{\beta}(t) &= \beta\, e^{-i(\varepsilon - \Delta\omega_a + 2\Delta\omega_b)t/2} \\ \bar{\gamma}(t) &= \gamma\, e^{-i(\varepsilon + \Delta\omega_a)t/2} \end{aligned} \tag{2.10}$$

where α, β and γ are constants. This results in an eigenvalue problem for the Rabi frequency ε.

$$\begin{aligned} (\varepsilon - \Delta\omega_a)\alpha &= \beta\Lambda_b + \gamma\Lambda_a \\ (\varepsilon - \Delta\omega_a + 2\Delta\omega_b)\beta &= \alpha\Lambda_b^* \\ (\varepsilon + \Delta\omega_a)\gamma &= \alpha\Lambda_a^* \end{aligned} \tag{2.11}$$

The corresponding eigenvalue equation:

$$\varepsilon^3 + \varepsilon^2(-\Delta\omega_a + 2\Delta\omega_b) - \varepsilon(\Delta\omega_a^2 + |\Lambda_a|^2 + |\Lambda_b|^2) +$$

$$(\Delta\omega_a - 2\Delta\omega_b)(|\Lambda_a|^2 + \Delta\omega_a^2) - |\Lambda_b|^2\Delta\omega_a = 0 \qquad (2.12)$$

can be solved numerically to obtain the values of the three Rabi frequencies ε_i. They are substituted back in eq. (2.11) and using the normalization condition:

$$|\alpha|^2 + |\beta|^2 + |\gamma|^2 = 1, \qquad (2.13)$$

we obtain the corresponding eigenfunctions. They are the unknown parameters in the expansion of the wave function of the system:

$$\begin{aligned}
\alpha(\varepsilon_i) &= \frac{\varepsilon_i + \Delta\omega_a}{\Lambda_a^*}\,\gamma(\varepsilon_i) \\
\beta(\varepsilon_i) &= \frac{\varepsilon_i^2 - \Delta\omega_a^2 - |\Lambda_a|^2}{\Lambda_a^*\Lambda_b}\,\gamma(\varepsilon_i) \qquad (2.14) \\
\gamma(\varepsilon_i) &= \left[1 + \frac{(\varepsilon + \Delta\omega_a)^2}{|\Lambda_a|^2} + \left(\frac{\varepsilon^2 - \Delta\omega_a^2 - |\Lambda_a|^2}{\Lambda_a^*\Lambda_b}\right)^2\right]^{-\frac{1}{2}}
\end{aligned}$$

This gives us three orthonormal functions which are the solution of the Schrodinger equation (2.1). They are the dressed states of the Ps atom in the field of two single mode near resonant lasers and are the generalization of the states $|\phi_\pm\rangle$ in (1.1).

This eigenvalue problem can be solved analytically close to resonance.

We assume:

$$\begin{aligned} \Delta\omega_a &= \Delta\omega + \frac{\delta}{2} \\ \Delta\omega_b &= \Delta\omega - \frac{\delta}{2}, \end{aligned} \tag{2.15}$$

where δ is a small parameter. The Rabi frequencies are found to be:

$$\begin{aligned} \varepsilon_1 &= -\Delta\omega + \delta\left\{-\frac{1}{2} + \frac{2|\Lambda_a|^2}{|\Lambda_a|^2 + |\Lambda_b|^2}\right\} + O(\delta^2) \\ \varepsilon_{2,3} &= \pm s + \delta\left\{1 \mp \frac{\Delta\omega}{2s} - \frac{|\Lambda_a|^2}{s(s \pm \Delta\omega)}\right\} + O(\delta^2), \end{aligned} \tag{2.16}$$

where

$$s = \sqrt{\Delta\omega^2 + |\Lambda_a|^2 + |\Lambda_b|^2}. \tag{2.17}$$

The dressed state which corresponds to the eigenvalue ε_1 is:

$$\begin{aligned} |\Psi_1(t)\rangle = \frac{1}{\sigma} e^{-i\delta t\frac{|\Lambda_a|^2}{\sigma^2}} \Bigg\{ & \delta\frac{2\Lambda_a\Lambda_b}{\sigma^2}\,|u_0\rangle\, e^{i(\Delta\omega+\frac{\delta}{2})t} e^{-iW_0 t} \\ & - \Lambda_a\left(1 + \delta\frac{4|\Lambda_b|^2\Delta\omega}{\sigma^4}\right)|u_1\rangle\, e^{i\delta t} e^{-iW_1 t} \\ & + \Lambda_b\left(1 - \delta\frac{4|\Lambda_a|^2\Delta\omega}{\sigma^4}\right)|u_2\rangle\, e^{-iW_2 t}\Bigg\}, \end{aligned} \tag{2.18}$$

where

$$\sigma = \sqrt{|\Lambda_a|^2 + |\Lambda_b|^2}. \tag{2.19}$$

The most important feature of the state $|\Psi_1\rangle$ is that at resonance ($\delta \equiv 0$) it does not contain the ground state $|u_0\rangle$. This is the phenomenon of coherent population trapping which we discussed in Chapter 1, section 3. The bare states $|u_1\rangle$ and $|u_2\rangle$ are stable against annihilation, as discussed in Chapter 1, section 1. Therefore in the case considered here, i.e. resonance, no fluorescence and ionization, the Ps atom is stable while in the laser field, if it is in the state $|\Psi_1\rangle$. On the other hand the other two dressed states are a linear superposition of all three bare Ps states and even under the above mentioned conditions will annihilate. Up to zeroth order in δ they are:

$$|\Psi_{2,3}(t)\rangle = \frac{e^{-i(\Delta\omega \pm s)\frac{t}{2}}}{\sqrt{2s(s \pm \Delta\omega)}} \Big\{(\Delta\omega \pm s)\,|u_0\rangle\, e^{-i(W_0-\Delta\omega)t} + \Lambda_b^*\,|u_1\rangle\, e^{-iW_1 t} + \Lambda_a^*\,|u_2\rangle\, e^{-iW_2 t}\Big\}. \quad (2.20)$$

The actual state of the Ps atom interacting with the laser fields will be a superposition of the dressed states (2.6), (2.10) and (2.14):

$$|\Phi_l(t)\rangle = \sum_{i=1,2,3} C_{li}\,|\Psi_i(t)\rangle\,. \quad (2.21)$$

The index l refers to the initial conditions, which determine the unknown constants C_{li}:

$$|\Phi_l(t=0)\rangle = C_{l1}\,|\Psi_1(t=0)\rangle + C_{l2}\,|\Psi_2(t=0)\rangle + C_{l3}\,|\Psi_3(t=0)\rangle\,. \quad (2.22)$$

They can be found analytically at resonance $\Delta\omega_a = \Delta\omega_b = \Delta\omega$, when the dressed states are given by eq. (2.18) (for $\delta = 0$) and (2.20). Let us assume first that Ps was initially in its ground state, i.e. $|\Phi_0(t=0)\rangle = |u_0\rangle$. Consecutive projections of eq (2.22) to the left with the dressed states, which are orthonormal, yields:

$$C_{01} = 0 \tag{2.23}$$

$$C_{02} = \sqrt{\frac{s+\Delta\omega}{2s}} \rightarrow \frac{1}{\sqrt{2}}$$

$$C_{03} = -\sqrt{\frac{s-\Delta\omega}{2s}} \rightarrow -\frac{1}{\sqrt{2}},$$

where the arrow indicates the limit for $\Delta\omega \rightarrow 0$. Under the idealized circomstances considered here, i.e. resonance, no fluorescence and no ionization, the wave function (2.21) does not contain the coherently trapped dressed state (2.18). We obtain the important result that in this case Ps cannot be stabilized if it was initially in its ground state. Including the above mentioned processes into the problem will lead to some corrections for the coefficient C_{01}. To what extend this will influence the lifetime can be found only by a complete solution to the problem.

If Ps is in the state $|u_1\rangle$ before the lasers are turned on, the constants in the expansion of the wave function onto the dressed states are:

$$C_{11} = -\frac{q}{\sqrt{1+q^2}} \tag{2.24}$$

$$C_{12} = \frac{\Lambda_b}{\sqrt{2s(s+\Delta\omega)}} \rightarrow \frac{1}{\sqrt{2(1+q^2)}}$$

$$C_{13} = \frac{\Lambda_b}{\sqrt{2s(s-\Delta\omega)}} \rightarrow \frac{1}{\sqrt{2(1+q^2)}},$$

where

$$q = \frac{|\Lambda_a|}{|\Lambda_b|} = \left(\frac{E^a}{E^a_{crit}}\right)^2 / \left(\frac{E^b}{E^b_{crit}}\right). \tag{2.25}$$

Note that in this case the value of the parameter q influences significantly the wave function. For $q > 1$ we have that $C_{11} \approx 1$ and $C_{12} = C_{13} \approx 0$. The system is almost exclusively in the coherently trapped dressed state. This means that Ps can live infinitely long in the idealized circomstances considered here. For q=1 we obtain that $C_{11} = -1/\sqrt{2}$ and $C_{12} = C_{13} = 1/2$. Therefore the probability to find Ps in the coherently trapped dressed state is one half and there is a possibility for extension of the lifetime depending on additional factors in the problem. For $q < 1$ the constant $C_{11} < -q$ and there is a small contribution from the trapped state.

When Ps was initially in the state $|u_2\rangle$, we have:

$$C_{21} = \frac{1}{\sqrt{1+q^2}} \tag{2.26}$$

$$C_{22} = \frac{\Lambda_a}{\sqrt{2s(s+\Delta\omega)}} \rightarrow \frac{q}{\sqrt{2(1+q^2)}}$$

$$C_{23} = \frac{\Lambda_a}{\sqrt{2s(s-\Delta\omega)}} \rightarrow \frac{q}{\sqrt{2(1+q^2)}}.$$

Here we have the reverse situation than in the previous case. $C_{21} \approx 1$ and $C_{21} = C_{23} \approx 0$ for $q < 1$ and Ps can live infinitely long. For $q > 1$ the coefficient in eq.(2.21) in front of the coherently trapped state is small.

These considerations lead to important conclusion regarding the possibility to extend the lifetime of Ps in the field of two lasers. At resonance, when photoionization and fluorescence are not present, the Ps atom can be in the coherently trapped dressed state in the following two cases:

a. The atom was initially in the excited state 2P and the coupling matrix element Λ_a of laser 'a' is equal or larger than the one of laser 'b'.

b. The atom was initially in the excited state 3D and the coupling

matrix element Λ_a of laser 'a' is equal or smaller than the one of laser 'b'.

If these conditions are fulfilled, coherent population trapping is possible and it will result in the stabilization of Ps. When the initial condition is the ground state, the Ps wave function in the lasers will not contain the coherently trapped state, which means that the lifetime of Ps cannot be increased. The problem considered here is an idealisation of the real one. Ionization needs to be included, which will give an upper bound on the values of the laser intensities (especially for laser 'a') and therefore some restrictions on the possible values of the parameter q. Spontaneous radiative transitions lead to the repopulation of the ground state and alter the wave function of the system given by eq. (2.21). They are not very relevant for singlet Ps, because its annihilation lifetime is much shorter than the fluorescent decay times. We have the inverse situation for triplet Ps and it is not clear in advance to what extend radiative transitions will elliminate the effect of coherent population trapping. Finally, large values of the laser detunings will lead to a ground state component in the dressed state (2.18) and will decrease the effect of coherent population trapping.

We would like to comment on some limiting cases of the dressed states (2.18) and (2.20). The situation encountered here is similar to the one in the two state problem [20] (Chapter 2.) for the states in eq. (1.1). Namely for $\hbar\omega/|\Lambda| \to \pm\infty$, the dressed states evolve into:

$$\begin{aligned} |\Phi_{\pm}\rangle &\to |u_0\rangle\, e^{-i\frac{W_0}{\hbar}t} \\ |\Phi_{\mp}\rangle &\to |u_1\rangle\, e^{-i\frac{W_1}{\hbar}t}\,. \end{aligned} \tag{2.27}$$

For the problem considered here, let us assume that in the remote past laser 'a' is turned on first before laser 'b'. During the experiment the detuning is kept positive $\Delta\omega > 0$. We have as

$$t \to -\infty: \quad \frac{\Lambda_b}{\Lambda_a} \to 0 \quad \text{and} \quad s \to \Delta\omega \tag{2.28}$$

We obtain that in the remote past the three dressed states originate respectively from the following bare Ps states:

$$\begin{aligned} |\Psi_1\rangle &\to \frac{\Lambda_a}{|\Lambda_a|}\,|u_1\rangle\, e^{-iW_1 t}\,, \\ |\Psi_2\rangle &\to |u_0\rangle\, e^{-iW_0 t}\,, \\ |\Psi_3\rangle &\to \frac{\Lambda_a^*}{|\Lambda_a|}\,|u_2\rangle\, e^{-iW_2 t}\,. \end{aligned} \tag{2.29}$$

If the system is in the ground state before the lasers are turned on, $|\Psi_2\rangle$ is the initial dressed state. The system will stay in this state, if the time evolution

is truly adiabatic. Let us assume that in the remote future ($t \to \infty$) laser 'a' is turned off after laser 'b'. Following the line of the above discussion, we obtain that the system will return into its ground state.

Suppose now that in the remote future we have the reverse sequence of events: laser 'a' is turned off before laser 'b'. In this case $\frac{\Lambda_a}{\Lambda_b} \to 0$ and the bare Ps states, which will respectively evolve from the dressed states are:

$$\begin{aligned} |\Psi_1\rangle &\to -\frac{\Lambda_b}{|\Lambda_b|}\,|u_2\rangle\, e^{-iW_2 t}\,, \\ |\Psi_2\rangle &\to |u_0\rangle\, e^{-iW_0 t}\,, \\ |\Psi_3\rangle &\to \frac{\Lambda_b^*}{|\Lambda_b|}\,|u_1\rangle\, e^{-iW_1 t}\,. \end{aligned} \tag{2.30}$$

The system starting in the ground state will return to it again. Now let us assume that in addition we have chirping, i.e. the detuning $\Delta\omega$ changes sign while the lasers are turned on. Then depending on the initial conditions, the system will evolve in the remote future into the following bare states:

$$\begin{aligned} |\Psi_1\rangle &\to -\frac{\Lambda_b}{|\Lambda_b|}\,|u_2\rangle\, e^{-iW_2 t}\,, \\ |\Psi_2\rangle &\to \frac{\Lambda_b^*}{|\Lambda_b|}\,|u_1\rangle\, e^{-iW_1 t}\,, \\ |\Psi_3\rangle &\to |u_0\rangle\, e^{-iW_0 t}\,. \end{aligned} \tag{2.31}$$

Therefore, if the Ps atom has started originally in the ground state, it will

stay in the dressed state $|\Psi_2\rangle$ during the interaction with the lasers and it will end in the excited state $|u_1\rangle$ after they are turned off adiabatically in the above sequence.

Consider the reverse situation: In the remote past laser 'b' is turned on first before laser 'a'. Then there is chirping of the lasers. Finally in the remote future laser 'b' is turned off before laser 'a'. If the system was originally in the ground state, it will evolve into the dressed state $|\Psi_2\rangle$ and finally end into the excited bare state $|u_2\rangle$. In this way, the interaction of Ps with two nearly resonant lasers leads to its excitation either into $|u_1\rangle$ or $|u_2\rangle$ depending on the sequence of events in the experiment.

It is necessary to make some comments on the approximations used to find the dressed states. The rotating wave approximation amounts to the neglect of terms such as:

$$\frac{\Lambda_b}{2} e^{-i(\omega_b + W_{10})t}. \tag{2.32}$$

We may estimate the contribution of this term by treating it as small and replacing β by its zero order value. The resulting corrections are:

$$\frac{\delta\alpha}{\alpha} \approx \frac{\delta\beta}{\beta} \approx \frac{\delta\gamma}{\gamma} \approx O\left(\frac{\Lambda}{W_{10}}\right), \tag{2.33}$$

which has been assumed to be small.

The three state approximation can also be corrected in the same manner. $|\Psi_i(t)\rangle$ in (2.6) are assumed to be zero order solutions and the contributions from the states $|u_n\rangle$ for $n \neq 0, 1, 2$ are assumed small. The coefficients of $|u_n\rangle$ in the wave function are then found to be of the same order of magnitude as the corrections to α in (2.33). Therefore the three state approximation and the RWA are of comparable accuracy and it is usually not worth correcting one of them without also correcting the other.

2.2 Spontaneous radiative transitions

In this section we incorporate spontaneous radiative transitions in the problem. When an atom is in a resonant laser field for a time long compared with the natural decay time of an excited state, then the probability that the state decays by emissioin of spontaneous radiation into modes other than the laser mode must be included in the considerations. Since the allowed spontaneous decay times are $10^{-8} - 10^{-9}$s, this is a common phenomenon for triplet Ps. Therefore in eq.(2.1) we add the interaction between the radiation field and

the Ps atom, which is given by $H_R = e\vec{E}_R(t) \cdot \vec{r}$ and

$$\vec{E}_R(t) = \sum_{k\lambda} \sqrt{\frac{2\pi\omega_k}{V}} \left(\hat{\varepsilon}_{k\lambda}\, e^{-i\omega_k t}\, a_{k\lambda} + \hat{\varepsilon}^*_{k\lambda}\, e^{i\omega_k t}\, a^\dagger_{k\lambda} \right) \tag{2.34}$$

is the electric field. Here $a^\dagger_{k\lambda}$ ($a_{k\lambda}$) are the creation (annihilation) operators for radiation photons with the usual bosonic commutation relations:

$$\left[a_{k\lambda} \, , \, a^\dagger_{k'\lambda'} \right] = \delta_{kk'}\, \delta_{\lambda\lambda'} \tag{2.35}$$

The total wave function is now expandable in the dressed states (2.6), which we have already found. It is convenient to use a matrix notation for these states:

$$|\Psi_1\rangle = e^{-ie_1 t/2\hbar} \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}, |\Psi_2\rangle = e^{-ie_2 t/2\hbar} \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix}, |\Psi_3\rangle = e^{-ie_3 t/2\hbar} \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}. \tag{2.36}$$

The Hamiltonian can now be written as a 3×3 matrix for which the Gell-Mann matrices λ_a for a = 1....8 (i.e. the generators of the unitary group

SU(3)[1]) and the unit matrix form a complete set of operators.

$$\lambda_1 = \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \quad \lambda_2 = \begin{pmatrix} 0 & -i & 0 \\ i & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \quad \lambda_3 = \begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

$$\lambda_4 = \begin{pmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{pmatrix} \quad \lambda_5 = \begin{pmatrix} 0 & 0 & -i \\ 0 & 0 & 0 \\ i & 0 & 0 \end{pmatrix} \quad \lambda_6 = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}$$

$$\lambda_7 = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & -i \\ 0 & i & 0 \end{pmatrix} \quad \lambda_8 = \frac{1}{\sqrt{3}} \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -2 \end{pmatrix} \quad \lambda_9 = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix} \tag{2.37}$$

Any hermitian matrix can be expanded as follows:

$$A_{ij} = \sum_{b=1}^{9} a_b \, (\lambda_b)_{ij} \, , \tag{2.38}$$

where the coefficients a_b are given by:

[1]For more information on the SU(3) group see for example [40].

$$a_1 = \tfrac{1}{2}(A_{12} + A_{21}) \quad a_2 = \tfrac{i}{2}(A_{12} - A_{21}) \qquad a_3 = \tfrac{1}{2}(A_{11} - A_{22})$$

$$a_4 = \tfrac{1}{2}(A_{13} + A_{31}) \quad a_5 = \tfrac{i}{2}(A_{13} - A_{31}) \qquad a_6 = \tfrac{1}{2}(A_{23} + A_{32})$$

$$a_7 = \tfrac{i}{2}(A_{23} - A_{32}) \quad a_8 = \tfrac{1}{2\sqrt{3}}(A_{11} + A_{22} - 2A_{33}) \quad a_9 = \tfrac{1}{3}(A_{11} + A_{22} + A_{33}) \tag{2.39}$$

We need to obtain the matrix of the full Hamiltonian in this representation using the detailed form of the three orthonormal dressed states. Part of the Hamiltonian is diagonal in this representation:

$$H_0 + H_A \;\rightarrow\; \frac{1}{2}\varepsilon_i\,\delta_{ij} \;=\; \sum_{a=1}^{9} V(\varepsilon_i)_a\;\lambda_a\;. \tag{2.40}$$

The coefficients $V(\varepsilon_i)$ are defined by eq. (2.33) and (2.34). They are:

$$\begin{aligned} V_3 &= \frac{1}{4}(\varepsilon_1 - \varepsilon_2) \qquad & V_8 &= \frac{1}{4\sqrt{3}}(\varepsilon_1 + \varepsilon_2 - 2\varepsilon_3) \\ V_9 &= \frac{1}{6}(\varepsilon_1 + \varepsilon_2 + \varepsilon_3) \end{aligned} \tag{2.41}$$

and all other V_i are zero. Our major concern is the term H_R which gives the interaction with the radiation field. We have emission of spontaneous radiation from the transitions $|u_2\rangle \rightarrow |u_1\rangle$ and $|u_1\rangle \rightarrow |u_0\rangle$. Therefore we can split the radiation field operator into two components depending on the value of the photon wave vector:

$$\vec{E}_R \;=\; \vec{E}_R^{21} \;+\; \vec{E}_R^{10}. \tag{2.42}$$

$\vec{E}_R^{21}$ includes photons with energy within a narrow interval [2] around W_{21}, while $\vec{E}_R^{10}$ refers to photons resulting from the second radiative transition with energy within a narrow interval around W_{10}. These two operators commute, because of (2.35). Using the notation $\vec{d}_{ij} = e\langle u_i|\vec{r}|u_j\rangle$ for the dipole operator, we obtain:

$$H = \sum_{a=1}^{9} V_a\lambda_a + \left\{ \left(\sum_{a=1}^{9} X_a\lambda_a\right) \vec{E}_R^{10(+)}(t)\cdot\vec{d}_{01}\, e^{-i\omega_b t} + \left(\sum_{a=1}^{9} Z_a\lambda_a\right) \vec{E}_R^{21(+)}(t)\cdot\vec{d}_{12}\, e^{-i(2\omega_a-\omega_b)t} + h.c.\right\}. \quad (2.43)$$

Here $\vec{E}_R^{21(\pm)}(t)$ and $\vec{E}_R^{10(\pm)}(t)$ are the positive and negative frequency components of the above radiation operators. Originally there are two types of time dependencies in this representation of the dipole operator $e^{\pm i\omega_L t}$ and two types of time dependencies in the radiation field operator $e^{\pm i\omega_k t}$. Since $\omega_k \sim \omega_L$, the product of the two terms will contain both a slow varying factor $e^{\pm i(\omega_L-\omega_k)t}$ and a rapid one $e^{\pm i(\omega_L-\omega_k)t}$. A second level RWA [39] has been made by dropping the rapidly varying terms. Its justification and validity are the same as the first one. The new physics introduced by this approximation is the requirement that fluorescent emission follows only immediately after

[2] The width of the interval is of the order of the width of the states.

the absorption of a laser photon, thereby roughly conserving energy at each step.

The new parameters X_a and Z_a introduced in the above expression for the radiation Hamiltonian H_R are known functions of the amplitudes $\alpha(\varepsilon_i)$, $\beta(\varepsilon_i)$ and $\gamma(\varepsilon_i)$, in (2.14). They result from the expansion of the Hamiltonian in this representation onto the basis set of matrices (2.37):

$$\begin{aligned} \alpha_i \beta_j &= \sum_{a=1}^{9} X_a \, (\lambda_a)_{ij} \\ \beta_i \gamma_j &= \sum_{a=1}^{9} Z_a \, (\lambda_a)_{ij} \ , \end{aligned} \tag{2.44}$$

using eq. (2.38) and (2.39).

Physical observables can be expressed as expectation values of operators with respect to the total wave function of the system $|\Psi(t)\rangle$. We can obtain equations for these observables from:

$$i\frac{d}{dt}\langle\Psi|\mathcal{O}|\Psi\rangle \ = \ \langle\Psi|[\mathcal{O}, H]|\Psi\rangle. \tag{2.45}$$

If the operator $\mathcal{O}$ for a given observable is independent of the radiation operators and also it is not explicitly time-dependent, we obtain:

$$\begin{aligned} &i\frac{d}{dt}\langle\Psi|\mathcal{O}|\Psi\rangle = \\ &\sum_{a=1}^{9} V_a \, \langle\Psi| \, [\mathcal{O}, \lambda_a] \, |\Psi\rangle + \left\{ \sum_{a=1}^{9} X_a \, \langle\Psi|\vec{E}_R^{10(+)}(t) \cdot \vec{d}_{01} \ [\mathcal{O}, \lambda_a] \, |\Psi\rangle e^{-i\omega_b t} \right. \end{aligned} \tag{2.46}$$

$$+\sum_{a=1}^{9} Z_a \, \langle\Psi|\vec{E}_R^{21(+)}(t)\cdot\vec{d}_{12}\;[\mathcal{O},\lambda_a]\,|\Psi\rangle\; e^{-i(2\omega_a-\omega_b)t} \;+\; h.c.\Big\}.$$

It is apparent from the above expression that the equation for $\mathcal{O}$ couples to expectation values which contain radiation operators linearly and equations for those (expectation values of E_R) will couple to still others. This results in an infinite set of coupled equations which describe the infinite number of degrees of freedom of the radiation field. Mollow's contribution was the use of an approximate solution for the electric field as an approximate constraint on the wave function, which allows the truncation of the infinite set of equations. In our case the costraint is as follows:

$$\begin{aligned}
\vec{E}_R^{10(-)}(t)\,|\Psi(t)\rangle &= -\frac{2i}{3}\,W_{10}^3\,\vec{d}_{01}\,e^{-i\omega_b t}\,(\sum_{a=1}^{9} X_a\lambda_a)\,|\Psi(t)\rangle \\
\vec{E}_R^{21(-)}(t)\,|\Psi(t)\rangle &= -\frac{2i}{3}\,W_{21}^3\;\vec{d}_{12}\,e^{-i(2\omega_a-\omega_b)t}\,(\sum_{a=1}^{9} Z_a\lambda_a)\,|\Psi(t)\rangle. \qquad (2.47)
\end{aligned}$$

A detailed derivation is given in Appendix C.

Any hermitian matrix can be expanded over the set $\{\lambda^a\}$ for a=1...9. Therefore we can find the expectation value of any operator if we know:

$$S_a = \langle\Psi|\lambda_a|\Psi\rangle \qquad \text{for a=1...8.} \qquad (2.48)$$

In order to find an equation for S_a in eq. (2.48), we make use of the con-

straints (2.47) and of the algebra of the Gell-Mann matrices:

$$[\lambda_a,\ \lambda_b] = 2i\ f_{abc}\ \lambda_c \tag{2.49}$$

$$\{\lambda_a,\ \lambda_b\} = \frac{4}{3}\delta_{ab}I + 2d_{abc}\lambda_c.$$

Here f_{abc} are the structure constants of SU(3) [40], as defined by the commutation relations in eq. (2.49). They are completely antisymmetric. The constants d_{abc} are symmetric in all indeces [40]. We obtain after some lengthy derivations the following system of linear inhomogeneous differential equations:

$$\frac{d}{dt}S_a\ +\ M_{ab}S_b\ =\ Q_a \qquad \text{for a=1...8.} \tag{2.50}$$

For a=9 we have:

$$\frac{d}{dt}\,S_9\ =\ \frac{d}{dt}\,\langle\Psi|\Psi\rangle\ =\ 0\ , \tag{2.51}$$

which is just the normalization condition.[3] The matrix M is given by:

$$\begin{aligned} M_{ab}\ =\ & -2f_{abc}\,V_c\ - \\ & -4f_{alc}f_{cqb}\,[\gamma_{10}\,Re(X_l^*X_q)\ +\ \gamma_{21}\,Re(Z_l^*Z_q)] \\ & -4f_{alc}d_{cqb}\,[\gamma_{10}\,Im(X_l^*X_q)\ +\ \gamma_{21}\,Im(Z_l^*Z_q)] \\ & +4f_{acb}\,[\gamma_{10}\,X_9Im(X_c)\ +\ \gamma_{21}\,Z_9Im(Z_c)]\ , \end{aligned} \tag{2.52}$$

[3]We do not have absorption and the Hilbert space comprises only bound Ps states. Therefore the probability to find Ps is one .

and the inhomogeneous term Q is:

$$Q_a \;=\; \frac{8}{3}\,\gamma_{10}\, f_{abc}\, Im(X_b^* X_c) \;+\; \frac{8}{3}\,\gamma_{21}\, f_{abc}\, Im(Z_b^* Z_c). \qquad (2.53)$$

The parameters γ_{10} and γ_{21} are half of the natural decay rates of the states 2P and 3D respectively and are equal to:

$$\begin{aligned}
\gamma_{10} \;&=\; \frac{2}{3}\,\frac{W_{10}^3}{\hbar^4 c^3}\,|\vec{d}_{10}|^2 \;=\; \frac{\hbar}{2\tau_{10}} \;=\; 3.793\;10^{-9}\;[\mathrm{Ht}] \\
\gamma_{21} \;&=\; \frac{2}{3}\,\frac{W_{21}^3}{\hbar^4 c^3}\,|\vec{d}_{21}|^2 \;=\; \frac{\hbar}{2\tau_{21}} \;=\; 3.916\;10{-}10\;[\mathrm{Ht}]. \qquad (2.54)
\end{aligned}$$

The constraints are an expansion in:

$$\frac{\gamma_{10}}{W_{10}} \approx \frac{\gamma_{10}}{\omega_b} \qquad \text{and} \qquad \frac{\gamma_{21}}{W_{21}} \approx \frac{\gamma_{21}}{\omega_a}\,, \qquad (2.55)$$

so that subsequent results are accurate only to lowest order in these parameters.

We assume that there is no time dependence in the coefficients of eq. (2.56) and therefore they are constants. The solution is then given by:

$$S_a = (M^{-1})_{ab}\,(1 - e^{-Mt})_{bc}\,Q_c \;+\; (e^{-Mt})_{ab}\,S_b(t=0)\,. \qquad (2.56)$$

$S_a(t=0)$ is the initial condition. We denote with m_i the eigenvalues of the matrix M. Numerical calculations show that $Re\, m_i > 0$. Therefore all the terms in (2.46) that contain e^{-Mt} will decrease exponentially for large times.

The latter is known as Markovian behaviour. This means that all memory of the initial conditions vanishes exponentially at large times. In principle, this is not correct since it is known [41] that the memory of the initial conditions decays as a power of time for sufficiently large times. The origin of this discrepency is the Markovian approximation involved in the derivation of the constraint. In general, the coefficient in front of the power is too small for this term to be observed. Moreover, it has been shown recently [42, 43], that the existence of the power law behaviour depends on the method of preparation of the initial state in the experiment and may not be as universal as it was assumed. In any case, we shall neglect this behaviour.

Sufficiently large times in the case under consideration means $t >> \tau_{10}, \tau_{21}$. We are interested in times $t \geq \tau_{an}^{tr}$, which is considerably larger. Therefore an accurate description of the behaviour of the system is given by:

$$S_a = (M^{-1})_{ab} Q_b. \qquad \text{for a=1...8.} \tag{2.57}$$

Using the above results we can obtain the probability to find the bare atomic states when the system has a wave function $|\Psi(t)\rangle$. They are:

$$l_0 = |\langle\Psi|u_0\rangle|^2 = \langle\Psi|u_0\rangle\langle u_0|\Psi\rangle = \langle\Psi|\alpha_i\alpha_j|\Psi\rangle = \sum_{a=1}^{9}(U_0(\alpha))_a\, S_a$$

$$l_1 = |\langle\Psi|u_1\rangle|^2 = \langle\Psi|u_1\rangle\langle u_1|\Psi\rangle = \langle\Psi|\beta_i\beta_j|\Psi\rangle = \sum_{a=1}^{9}(U_1(\beta))_a\, S_a \quad (2.58)$$

$$l_2 = |\langle\Psi|u_2\rangle|^2 = \langle\Psi|u_2\rangle\langle u_2|\Psi\rangle = \langle\Psi|\gamma_i\gamma_j|\Psi\rangle = \sum_{a=1}^{9}(U_2(\gamma))_a\, S_a$$

The coefficients $(U_0)_a$, $(U_1)_a$ and $(U_2)_a$ are known functions of the parameters α_i, β_i and γ_i. For exaple $(U_0)_a$ are obtained from the expansion of the matrix $\alpha_i\alpha_j$ onto the Gell-Mann matrices using eq. (2.39):

$$(U_0)_1 = \alpha_1\alpha_2 \qquad (U_0)_2 = 0 \qquad (U_0)_3 = \frac{1}{2}(\alpha_1^2 - \alpha_2^2) \quad (2.59)$$

$$(U_0)_4 = \alpha_1\alpha_3 \qquad (U_0)_5 = 0 \qquad (U_0)_6 = \alpha_2\alpha_3 \qquad (U_0)_7 = 0$$

$$(U_0)_8 = \frac{1}{2\sqrt{3}}(\alpha_1^2 + \alpha_2^2 - 2\alpha_3^2) \qquad (U_0)_9 = \frac{1}{3}(\alpha_1^2 + \alpha_2^2 + \alpha_3^2)\,.$$

Finding the probabilities l_0, l_1 and l_2 is equivalent to finding the wave function $|\Psi(t)\rangle$ as a linear combination of the bare Ps states.

The occupation probabilities of the bare states are found numerically for different values of the laser intensities with a FORTRAN program. The results are given in Table 1.1. We find that the probability to find Ps in the excited state 2P is insignificant when the coupling with laser 'a' is stronger, while the probabilities for 1S and 3D are almost one half. This is logical because laser 'a' induces transitions from the ground state into 3D. In a similar way, the probability to find Ps in the state 3D is almost negligible,

$\|\Lambda_a\|$	$\|\Lambda_b\|$	l_0	l_1	l_2
$10\ \Gamma_0$	$10\ \Gamma_0$	0.488	0.256	0.256
$10\ \Gamma_0$	Γ_0	0.475	0.050	0.475
Γ_0	$10\ \Gamma_0$	0.499	0.499	0.002

Table 2.1: Probability to find the bare states in the wave function of triplet Ps, when the atom is in the field of two lasers. Photoionization is not considered. When the laser intensities are critical $|\Lambda_a| = |\Lambda_b| = \Gamma_0$.

when the coupling with laser 'b' is stronger. In this case the probabilities for 1S and 2P are almost one half. When the laser couplings are equal, the bare excited states have equal probability to be occupied. On the other hand the probabitity for the ground state remains almost the same and it is less than, but very close to one half. The figures given in Table 1.1 do not change significantly when the laser intensities are increased. They depend mainly on the ratio Λ_a/Λ_b and if it is smaller or greater than one. We find that the values of the detunings $\Delta\omega_a$ and $\Delta\omega_b$ within a range from zero up to a maximum of the natural widths of the states 3D and 2P respectively, practically do not influence the results.

The annihilation term in the Hamiltonian is given by:

$$H_D \;=\; -\frac{1}{2}\,\Gamma_0\,|u_c\rangle\langle u_0| \tag{2.60}$$

This is a phenomenological expression, without consideration for the mechanism of annihilation or for the inverse process, i.e. creation of an electron and positron pair from high energy γ-quanta. The latter are irrelevant for our considerations. Since τ_{an}^{tr} is much larger than the other characteristic times in the problem, H_D is much smaller than the laser-atom or the fluorescent interactions. Therefore it is justified to consider it in perturbation theory up to first order. In this case the annihilation rate of Ps in the laser fields is given by the following expression:

$$\Gamma \;=\; 2\,\langle\Psi|H_D|\Psi\rangle \;=\; \Gamma_0\,l_0\,. \tag{2.61}$$

It is equal to the annihilation rate outside of the laser fields times the probability to find Ps in the ground state when the atom is interacting with the lasers. This means we have neglected any correlations between the annihilation process on one hand and the laser induced or the spontaneous radiative transitions on the other hand. This is reasonable, since the latter are much faster than annihilation and the atom fluoresces many times. From

the numerical values for the probability given in Table 1.1 we obtain that the minimum value of the Ps decay rate is:

$$\Gamma = 0.475\,\Gamma_0 = 0.81\ 10^{-10}\ [\mathrm{Ht}] \tag{2.62}$$

Therefore the result is similar to the one obtained in the two state case. It is not possible to increase the lifetime of triplet Ps more than two times when the atom is in the laser fields. The reason for this are spontaneous radiative transitions, which are fast in comparison to the annihilation lifetime. In spite of coherent population trapping, they repopulate the ground state and lead to annihilation.

Chapter 3

Singlet positronium in the field of one near-resonant laser

According to the discussion in Chapter 1, we do not include spontaneous radiative transitions in this problem, but we have to consider the annihilation interaction nonperturbatively. Since annihilation and ionization are competing processes and both decrease the lifetime of Ps, they are considered simultaneously. We seek to find an optimal value for the laser intensity, so that the destruction of Ps is minimized.

We have the following Hamiltonian:

$$H = H_0' + H_A + H_D \tag{3.1}$$

The first step is to allow for annihilation of Ps in the ground state. The free part H_0' now includes also the free Hamiltonian of the high energy γ-quanta resulting from annihilation:

$$H_0' = (H_0 + 2m_e c^2)R_m + H_\gamma R_\gamma \, . \tag{3.2}$$

Since H_γ is relativistic, we have also added to H_0' the rest mass of the electron-positron pair. R_γ is a projection operator onto the subspace of the high energy photons with wave function $|\gamma_k\rangle$.

$$R_\gamma = \int \frac{d^3k}{(2\pi)^3} \, |\gamma_k\rangle\langle\gamma_k| \, . \tag{3.3}$$

The 'matter states' include all the bound states of Ps $|u_n\rangle$ and the continuum states $|u_q\rangle$ resulting from ionization. R_m is the projection operator onto matter subspace:

$$R_m = \sum_n |u_n\rangle\langle u_n| + \int \frac{d^3q}{(2\pi)^3} |u_q\rangle\langle u_q| \tag{3.4}$$

The Hilbert space is a direct sum of these two subspaces.[1] We have:

$$R_m \rightarrow \begin{cases} |u_n\rangle \\ \\ |u_q\rangle \end{cases} \qquad R_\gamma \rightarrow |\gamma_k\rangle \tag{3.5}$$

and $R_m + R_\gamma = 1$.

The annihilation Hamiltonian is:

$$H_D \;=\; -\frac{i}{2}\frac{\hbar}{\tau^s_{an}}\,|\gamma_k\rangle\langle u_0|\;. \tag{3.6}$$

The laser-atom interaction is chosen in the $\vec{r}\cdot\vec{E}$ gauge and in the dipole approximation it is:

$$H_A \;=\; e\vec{E}(t)\cdot\vec{r}\,R_m\;. \tag{3.7}$$

Note that the atom is interacting with one circularly polarized laser and due to the dipole selection rules the states have the following quantum numbers:

$$\begin{aligned} |u_0\rangle \equiv |1S\rangle \;&:\; l=0,\; m=0, \\ |u_1\rangle \equiv |2P\rangle \;&:\; l=1,\; m=1, \\ |u_q\rangle \;&:\; l=2,\; m=2. \end{aligned} \tag{3.8}$$

[1]The system cannot have states, which belong simultaneously to the matter and γ-quanta subspaces. There is either a bound Ps state, a ionized electron-positron pair or high energy γ-quanta. Therefore the Hilbert space is not the direct product of the two subspaces.

Energy considerations do not allow single photon transitions from the ground state into the continuum.

The second step is to include ionization in the problem. A detailed discussion of a two state atom, including ionization is given in [20] (Chapter 7). For a weak laser perturbation theory is an adequate description of a two photon ionization process from the ground state except in the case when an intermediate state $|u_1\rangle$ is nearly resonant with a single photon absorption by the ground state $|u_0\rangle$. Since this is exactly the case here, we must allow for strong coupling between these states in a manner analogous to the one already discussed in Chapter 2. This is accomplished by the projection operator formalism. We again partition the Hilbert space of the Hamiltonian, in a similar way as it was done above into matter and γ-quanta subspaces. This time one of the subspaces includes the states of interest: $|u_0\rangle$, $|u_1\rangle$, the ionization continuum $|u_q\rangle$ and the high energy photons $|\gamma_k\rangle$, while the remaining bound states of the Ps atom $|u_n\rangle$ for $n \neq 0, 1$ are included into the second subspace. The operator P projects onto the states of interest:

$$P = |u_0\rangle\langle u_0| + |u_1\rangle\langle u_1| + \int \frac{d^3q}{(2\pi)^3}|u_q\rangle\langle u_q| + \int \frac{d^3k}{(2\pi)^3}|\gamma_k\rangle\langle\gamma_k| \,. \quad (3.9)$$

Its compliment is Q = 1 - P. We have:

$$P \rightarrow \begin{cases} |u_0\rangle \\ |u_1\rangle \\ |u_q\rangle \\ |\gamma_k\rangle \end{cases} \qquad Q \rightarrow |u_n\rangle \text{ for n} \neq 0, 1 \tag{3.10}$$

The Ps atom can be ionized either by a resonant transition via the state $|u_1\rangle$ or by a nonresonant transition via an intermediate state $|u_n\rangle$ for $n \neq 0, 1$.

The resulting equation for $|P\Psi\rangle$ is:

$$P\,(i\hbar\frac{\partial}{\partial t} - \mathcal{H})|P\Psi\rangle \;=\; 0\;. \tag{3.11}$$

where:

$$\mathcal{H} \;=\; H \;+\; HQG_Q^{(+)}QH\;. \tag{3.12}$$

The Green's function $G_Q{}^{(+)}$ satisfies:

$$Q\,(i\hbar\frac{\partial}{\partial t} - H)\,QG_Q^{(+)} \;=\; Q\,\delta(t-t') \qquad . \tag{3.13}$$

with causal outgoing wave boundary conditions. The resonant pair of states is excluded by the Q operation, so we may safely use a perturbation method in powers of the laser field in the term containig this factor. In that case the

lowest order contribution suffices for this term. It is:

$$G_Q^{(+)}(t,t') \;=\; -\frac{i}{\hbar}\,\Theta(t-t')\,{\sum_n}'\,|u_n\rangle e^{-(\eta+iW_n)(t-t')/\hbar}\langle u_n|\,, \tag{3.14}$$

where $\eta \to 0^+$. The prime in the sum denotes that the states in the projection operator P are not included.

We now expand $|P\Psi\rangle$ in the bare states:

$$\begin{aligned} |P\Psi\rangle = \\ &\alpha(t)\,|u_0\rangle\,e^{-iW_0t/\hbar} \;+\; \beta(t)\,|u_1\rangle\,e^{-iW_1t/\hbar} \\ &+\int\frac{d^3q}{(2\pi)^3}\,\gamma_q(t)\,|u_q\rangle\,e^{-iW_qt/\hbar} \;+\; \int\frac{d^3k}{(2\pi)^3}\,\delta_k(t)\,|\gamma_k\rangle\,e^{-i\Delta E_kt/\hbar} \end{aligned} \tag{3.15}$$

where $E_k = \langle\gamma_k|H_\gamma|\gamma_k\rangle$ is the energy of the high energy photons and $\Delta E_k = E_k - 2mc^2$. We substitute this expression back into (3.11). Successive projections from the left with $|u_0\rangle, |u_1\rangle, |u_q\rangle$ and $|\gamma_k\rangle$ yield a set of equations for the amplitudes α, β, γ_q and δ_k. As in Chapter 2, the RWA is used and only slowly varying exponential terms are retained. We obtain:

$$\begin{aligned} i\dot\alpha &= \frac{\Lambda}{2\hbar}\,e^{i\Delta\omega t}\,\beta \;+\; \int\frac{d^3k}{(2\pi)^3}\,\frac{D_k}{\hbar}\,e^{i\Delta\omega t}\,\delta_k \;+\; \int\frac{d^3q}{(2\pi)^3}\,\frac{H_{0q}}{4\hbar}\,e^{i(\Delta\omega_q-\Delta\omega)t}\,\gamma_q \\ i\dot\beta &= \frac{\Lambda^*}{2\hbar}\,e^{-i\Delta\omega t}\,\alpha \;+\; \int\frac{d^3q}{(2\pi)^3}\,\frac{\Lambda_q}{2\hbar}\,e^{i\Delta\omega t}\,\gamma_q \\ i\dot\gamma_q &= \frac{\Lambda_q^*}{2\hbar}\,e^{-i\Delta\omega_q t}\,\beta \;+\; \frac{H_{0q}^*}{4\hbar}\,e^{-i(\Delta\omega_q+\Delta\omega)t}\,\alpha \end{aligned}$$

$$i\dot{\delta}_k = \frac{D_k^*}{\hbar} e^{-i\Delta\omega_k t}\,\alpha \tag{3.16}$$

Here the coupling matrix elements with the laser field are $\Lambda = \langle u_0|e\vec{r}\cdot\vec{E}|u_1\rangle$ and $\Lambda_q = \langle u_1|e\vec{r}\cdot\vec{E}|u_q\rangle$. The notations for the frequencies are similar to the ones used before: We have $\Delta\omega = \omega - W_{10}/\hbar$, $\Delta\omega_q = \omega - W_{q1}/\hbar$ and $\Delta\omega_k = (W_0 - \Delta E_k)/\hbar$. The matrix element of the annihilation interaction between the ground state of Ps and the free high energy photons is denoted by $D_k = \langle u_0|H_D|\gamma_k\rangle$, while H_{0q} is a second order matrix element arising from ionization of the ground state via all possible intermediate Ps states excluding the resonant one:

$$H_{0q} = \sum_n{}' \frac{\langle u_0|e\vec{r}\cdot\vec{E}|u_n\rangle\langle u_n|e\vec{r}\cdot\vec{E}|u_q\rangle}{W_{0n} + \hbar\omega} \tag{3.17}$$

Λ and Λ_q result from one photon transitions, while H_{0q} from non-resonant two photon transitions. Terms containing the latter are one power of $\frac{\Lambda}{\hbar\omega}$ smaller than the terms with Λ and Λ_q. The H_{0q} terms are the same order as the terms that are dropped in the RWA and therefore they should also be dropped.

In the equation for γ_q we have also neglected the coupling of continuum states to other continuum states by the laser. This is assumed to be a small effect, based on the fact that the so called free-free matrix elements which

couple these states are small compared to Λ and Λ_q.

Note that the right-hand side of the set of equations (3.16) is a hermitian matrix. Therefore the normalization of the wave function in P-subspace is preserved:

$$|\alpha|^2 + |\beta|^2 + \int \frac{d^3q}{(2\pi)^3}|\gamma_q|^2 + \int \frac{d^3k}{(2\pi)^3}|\delta_k|^2 = 1. \tag{3.18}$$

We solve the set of equations in the sudden approximation. In our case this is an unrealistic situation. However we shall be able to use the results obtained in this way to generate the adiabatic switching result, which is realistic. We impose the initial condition $\alpha(t = 0) = 1$ and all the other amplitudes are zero. Using the Laplace transformation, we obtain a system of coupled algebraic equations in s-space, which can be solved with the result:

$$\begin{aligned}
\tilde{\alpha}(s) &= \frac{s - i\Delta\omega + I(s - i\Delta\omega)}{D(s - i\Delta\omega)} \\
\tilde{\beta}(s) &= \frac{\Lambda^*}{2iD(s)} \qquad (3.19) \\
\tilde{\gamma}_q(s) &= \frac{\Lambda_q^*}{2\hbar}\,\frac{\tilde{\beta}(s + i\Delta\omega_q)}{is} \\
\tilde{\delta}_k(s) &= \frac{D_k^*}{\hbar}\,\frac{\tilde{\alpha}(s + i\Delta\omega_k)}{is}.
\end{aligned}$$

(3.20)

The denominator

$$D(s) \;=\; [s + i\Delta\omega + J(s + I\Delta\omega)]\;[s + I(s)] \;+\; \frac{|\Lambda|^2}{4\hbar^2} \tag{3.21}$$

is a function of two integrals I(s) and J(s). They can be expressed via known parameters:

$$\begin{aligned} I(s) &= \int \frac{d^3q}{(2\pi)^3}\,\frac{|\Lambda_q|^2}{4\hbar^2}\,\frac{1}{s - i\Delta\omega_q} \\ J(s) &= \int \frac{d^3k}{(2\pi)^3}\,\frac{|D_k|^2}{\hbar^2}\,\frac{1}{s - i\Delta\omega_k}, \end{aligned} \tag{3.22}$$

and are small compared to the other quantities in the problem.

We may now invert the Laplace transformation with:

$$\beta(t) \;=\; \frac{1}{2\pi i}\int_{-i\infty}^{i\infty} ds\, e^{st}\,\tilde{\beta}(s), \tag{3.23}$$

where the contour in the s-plane passes to the right of the singularities of $\tilde{\beta}(s)$. The integral can be performed by first finding the zeros of the denominator D(s). But strictly speaking there are no zeros [41], because of the s-dependence in the integrals I(s) and J(s). Instead there are branch cuts. Taking them into account would lead to a non-Markovian behaviour, indicating that the memory of the initial state is retained in the wave function. These effects are small to be of experimental interest. We shall limit ourselves to the 'pole approximation', which results in a Markovian behaviour

of the sysytem. We treat the denominator D(s) as a simple quadratic form in s:

$$D(s) \;=\; (s - s_+)(s - s_-)\,, \tag{3.24}$$

whose roots $s_\pm$ are functions of the two integrals I(s) and J(s). Then the roots $s_\pm$ are expanded in powers of I(s) and J(s), retaining only first order terms. Subsequently the s-dependence in the integrals is treated iteratively by substituting the zero order form of the roots $s_\pm$ into them. The latter are denoted by $I_\pm$ and $J_\pm$. We obtain:

$$s_\pm \;=\; \pm\frac{i}{2\hbar}|\Lambda|e^{\mp\mu} \;-\; \frac{e^{\pm\mu}}{2ch\mu}I_\pm \;-\; \frac{e^{\mp\mu}}{2ch\mu}J_\pm, \tag{3.25}$$

where:

$$\begin{aligned} I_\pm &= I\left(\pm\frac{i}{2}\frac{|\Lambda|}{\hbar}\,e^{\pm\mu} + \eta\right) \\ J_\pm &= I\left(\pm\frac{i}{2}\frac{|\Lambda|}{\hbar}\,e^{\pm\mu} + \eta\right)\,, \end{aligned} \tag{3.26}$$

for $\eta \to 0$. The same notations are used as in Chapter 1, section 2. The detuning parameter is $sh\mu = \frac{\hbar\Delta\omega}{|\Lambda|}$.

Now the contour integral in $\beta(t)$ can be carried out with the result:

$$\beta(t) \;=\; \frac{\Lambda^*}{2i}\,\frac{e^{ts_+} - e^{ts_-}}{s_+ - s_-} \tag{3.27}$$

The exponential decay at large times, instead of a power law, indicates the Markovian nature of the 'pole approximation'. The simplest way to find the other amplitudes is via the set of differential equations (3.16). The constants of integration are chosen in such a way as to satisfy the initial conditions. Terms of the order $\frac{I_\pm}{|\Lambda|}$ and $\frac{J_\pm}{|\Lambda|}$ are dropped consistently with the 'pole approximation' of the denominators. We obtain that each of the amplitudes is the difference of the following two terms:

$$\begin{aligned}
\alpha_\pm(t) &= \frac{s_\pm\, e^{(i\Delta\omega+s_\pm)t}}{s_+ - s_-}\,, \\
\gamma_{q\pm}(t) &= -\frac{\Lambda_q^*\Lambda^*}{4\hbar^2}\,\frac{e^{(-i\Delta\omega_q+s_\pm)t}}{(s_+ - s_-)(s_\pm - i\Delta\omega_q)}\,, \\
\delta_{k\pm}(t) &= -\frac{iD_k^*}{\hbar}\,\frac{s_\pm\, e^{(i\Delta\omega-i\Delta\omega_k+s_\pm)t}}{(s_+ - s_-)(s_\pm - i\Delta\omega_k + i\Delta\omega)}\,.
\end{aligned} \tag{3.28}$$

The calculation described up to this point applies to the situation in which the laser amplitude is zero for $t < 0$ and is constant for $t > 0$. The sudden switching is usually not a good description of the physical situation. A more realistic description is adiabatic switching of the laser. The results obtained so far can be adjusted to this case by allowing $s_\pm$ to be a slowly varying function of time through its dependence on the laser amplitude. The latter must be zero in the remote past and future and it should remain essentially

constant for a time duration T around t=0. Then the only change required is the replacement:

$$s_{\pm}t \;\rightarrow\; \int_{-\infty}^{t} dt'\, s_{\pm}(t') \tag{3.29}$$

in the expressions (3.27) and (3.28).

With this adjustment, the results obtained for the amplitudes can now be replaced in eq. (3.15) to find the wave function of the system in the adiabatic approximation:

$$|P\Psi\rangle \;=\; A_{+}\,|\Psi_{+}\rangle \;+\; A_{-}\,|\Psi_{-}\rangle, \tag{3.30}$$

where:

$$\begin{aligned} |\Psi_{\pm}\rangle \;=\; n_{\pm}\, e^{\int_{-\infty}^{t} dt'\, s_{\mp}(t')} \Bigg\{ & s_{\mp}\,|u_0\rangle\, e^{-iW_0 t/\hbar} \;+\; \frac{\Lambda^*}{2i\hbar}\,|u_1\rangle\, e^{-iW_1 t/\hbar} \\ & - \frac{\Lambda_q^*\Lambda}{4\hbar^2}\int \frac{d^3q}{(2\pi)^3}\,\frac{e^{-i\Delta\omega_q t}}{s_{\mp}-i\Delta\omega_q}\, e^{-iW_q t/\hbar}\,|u_q\rangle \\ & - \frac{i}{\hbar}\int \frac{d^3k}{(2\pi)^3}\,\frac{D_k^* s_{\mp}}{s_{\mp}+i\Delta\omega-i\Delta\omega_k}\, e^{i(\Delta\omega-\Delta\omega_k)t}\, e^{-i\Delta E_k t/\hbar}\,|\gamma_k\rangle \Bigg\} \end{aligned} \tag{3.31}$$

Up to lowest order in the integrals $I_{\pm}$ and $J_{\pm}$ the normalization constant is:

$$n_{\pm} \;=\; \frac{i\hbar}{|\Lambda|}\, e^{\mp\mu/2}\,\sqrt{\frac{2}{ch\mu}}. \tag{3.32}$$

The state $|P\Psi\rangle$ in eq. (3.30) was found via the sudden approximation wave function with the initial condition that $|P\Psi\rangle = |u_0\rangle$ at t=0. It satisfies this

condition for $t \to -\infty$ in the adiabatic approximation with the appropriate choice of the constants $A_\pm$. Note that the states $|\Psi_\pm\rangle$ are analogous to $|\phi_\pm\rangle$ in eq. (1.1), which are the wave functions of a two state atom in an adiabatically varying laser in the RWA. Indeed when the coupling to the continuum and the annihilation interaction are turned off ($\Lambda_q \to 0$ and $D_k \to 0$), we have $|\Psi_\pm\rangle \to |\phi_\pm\rangle$. When the laser is also switched off ($\Lambda \to 0$) in the remote past, we have:

$$\begin{aligned} |\Psi_+\rangle &\to |u_0\rangle e^{-iW_0 t/\hbar} \\ |\Psi_-\rangle &\to |u_1\rangle e^{iW_1 t/\hbar}, \end{aligned} \tag{3.33}$$

if $\Delta\omega > 0$ and the reverse for $\Delta\omega < 0$.

Now let us consider the case $\Delta\omega > 0$. The system starts in the state $|u_0\rangle$ which at $t \to -\infty$ is $|\Psi_+\rangle$. The adiabatic theorem tells us that for slow changes in the laser amplitude the system remains in this state. Therefore instead of the wave function $|P\Psi\rangle$ from eq. (3.30) and (3.31) we should take:

$$\begin{aligned} |P\Psi\rangle &\equiv |\Psi_+\rangle \quad \text{for } \Delta\omega > 0 \text{ or} \\ |P\Psi\rangle &\equiv |\Psi_-\rangle \quad \text{for } \Delta\omega < 0 \end{aligned} \tag{3.34}$$

as the dressed state of the system.

The probability to find a stable state of the Ps atom can be derived from eq. (3.31). Up to lowest order in the integrals $I_\pm$ and $J_\pm$ it is:

$$\begin{aligned} L_\pm &= |\langle \Psi_\pm | u_0 \rangle|^2 + |\langle \Psi_\pm | u_1 \rangle|^2 \\ &= exp\left\{ 2Re \int_{-\infty}^{\infty} dt\, s_\mp(t) \right\}. \end{aligned} \tag{3.35}$$

According to the above discussion, the subscript '$\pm$' refers to the cases when the detuning $\Delta\omega$ is positive or negative. The quantity:

$$-\,2\,Re \int_{-\infty}^{\infty} dt\, s_\mp(t) \;=\; \frac{\Gamma_\pm T}{\hbar} \;=\; \frac{T}{\tau_\pm^s} \tag{3.36}$$

can be considered as the transition parameter for ionization and annihilation of Ps in a near resonant laser. $\Gamma_\pm$ is the related width of the dressed state from which the transition to the continuum takes place, and $\tau_\pm$ is the lifetime of stable Ps. We can also interpret $Im\, s_\pm$ as the energy shift of the dressed state. Since we have already lost small energy shifts due to the RWA, it is inconsistent to keep small contributions arising from $Im\, s_\pm$ and they will be dropped. If the adiabatic switching of the laser takes place in a time which is short compared to the time T that the system spends in the laser field, during the remaining time the laser amplitude will be constant. In that case

we may replace the operator:

$$\frac{1}{T}\int_{-\infty}^{\infty} dt \;\rightarrow\; 1 \tag{3.37}$$

and evaluate $s_{\pm}$ by simply inserting that constant value of the laser amplitude.

In order to relate the width (3.36) to other characteristic quantities of the system, we find from (3.22) and (3.26):

$$\begin{aligned} Re\, I_{\pm} &= \pi \int \frac{d^3q}{(2\pi)^3}\, \frac{|\Lambda_q|^2}{4\hbar^2}\, \delta\left(\mp\frac{\Lambda}{2\hbar}e^{\mp\mu} + \Delta\omega_q\right) \\ Re\, J_{\pm} &= \pi \int \frac{d^3k}{(2\pi)^3}\, \frac{D_k^2}{\hbar^2}\, \delta\left(\mp\frac{\Lambda}{2\hbar}e^{\mp\mu} + \Delta\omega_k\right) \end{aligned} \tag{3.38}$$

In the above expressions there are energy conservation δ-functions respectively for ionization from $|u_1\rangle$ by the laser and for annihilation from $|u_0\rangle$. The first term in the argument of the δ-function is due to the $Im\, s_{\pm}$. As we mentioned, this is a small shift in the energy levels of the dressed states and has to be dropped. From eq. (3.25) and (3.38) follows:

$$\begin{aligned} Re\, s_{\pm} &= -\frac{\pi}{2ch\mu}\left\{e^{\pm\mu}\int \frac{d^3q}{(2\pi)^3}\, \frac{|\Lambda_q|^2}{4\hbar^2}\, \delta(\Delta\omega_q)\right. \\ &\quad \left. +\; e^{\mp\mu}\int \frac{d^3k}{(2\pi)^3}\, \frac{D_k^2}{\hbar^2}\, \delta(\Delta\omega_k)\right\} \end{aligned} \tag{3.39}$$

But note that the first term in this expression is proportional to the pho-

toionization rate from the state 2P:

$$w_{2P} = \frac{\pi e^2}{2\hbar^2} E^2 \int \frac{d^3q}{(2\pi)^3} \left|\hat{\varepsilon} \cdot \langle u_1|\vec{r}|u_q\rangle\right|^2 \delta(\Delta\omega_q) , \tag{3.40}$$

while the second term is proportional to the annihilation rate outside of the laser:

$$\Gamma_0 = 2\pi \int \frac{d^3k}{(2\pi)^3} \frac{|D_k|^2}{\hbar^2} \delta(\Delta\omega_k) . \tag{3.41}$$

With the above interpretation the meaning of the integrals is straightforward. Near resonance $I_\pm$ gives the width of the state $|u_1\rangle$ due to resonant ionization, while $J_\pm$ gives the width of the ground state due to annihilation. Now the width of the dressed Ps state can be given in terms of the ionization and annihilation rates:

$$\Gamma_\pm = \frac{1}{2ch\mu} \left(e^{\pm\mu} w_{2P} + e^{\mp\mu} \Gamma_0\right) . \tag{3.42}$$

From eq. (1.1) we obtain that the probability to find each of the bare atomic states in the dressed state $|\phi_\pm\rangle$ if annihilation and ionization are not present is respectively:

$$l_0^\pm = \frac{e^{\pm\mu}}{2ch\mu} \qquad \text{for } 1S, \tag{3.43}$$

and

$$l_1^\pm = \frac{e^{\mp\mu}}{2ch\mu} \qquad \text{for } 2P. \tag{3.44}$$

With this in mind, we can rewrite eq. (3.42) as:

$$\Gamma_{\pm} \;=\; l_1^{\pm}\, w_{2P} \;+\; l_0^{\pm}\, \Gamma_0. \tag{3.45}$$

Therefore the decay rate of Ps in the laser is equal to the probability to find the state 2P in a two state atom interacting with a laser field times the ionization rate from this bare state, added to the probability to find the state 1S times the annihilation rate outside of the laser.

The above expression for the decay rate depends on the amplitude of the laser electric field E via the ionization rate w_{2P} and the detuning parameter μ in the probabilities $l_0^{\pm}$ and $l_1^{\pm}$. We seek the optimal laser intensity for which the lifetime $\tau_{\pm}^{s}$ is maximal. Taking the derivative with respect to E^2, we obtain the condition for the extremum:

$$2x^3 \pm x^2 \pm 1 \pm \frac{t}{\Delta\omega^2} = 0\,, \tag{3.46}$$

where the dimensionless parameter x is the ratio of the Rabi frequency as defined in eq. (1.5) and the detuning $\Delta\omega$:

$$x \;=\; \frac{\varepsilon}{\Delta\omega} \;=\; \sqrt{1 + \frac{|\Lambda|^2}{\Delta\omega^2}}\,. \tag{3.47}$$

It depends on the laser intensity via Λ. On the other hand t is a constant:

$$t \;=\; \Gamma_0\, \frac{|\Lambda|^2}{w_{2P}}\,, \tag{3.48}$$

since both $|\Lambda|^2$ and w_{2P} are quadratic functions of the amplitude of the laser electric field. We solve eq. (3.46) numerically for different values of the detuning $\Delta\omega$. With the optimal laser intensity found in this way we evaluate the decay rate $\Gamma_\pm$ in eq. (3.45). We obtain that the lifetime of Ps in the laser increases when $\Delta\omega$ decreases. It reaches the value of twice the lifetime outside of the laser for a detuning of approximately 10^{-9} [Ht] and doesn't change further. We obtain the same result as in the two state problem discussed in Chapter 1, section 2. This means on one hand that the ionization rate from the state 2P is small and does not contribute to Ps destruction when the atom is interacting with one laser, as estimated roughly beforehand in Chapter 1, section 5. On the other hand, we have confirmed the assumption that the annihilation rate in the laser $\Gamma_\pm = l_0^\pm \Gamma_0$ is equal to the rate outside of the laser times the probability to find the ground state in the dressed state, when photoionization is not significant. This is a nonperturbative result.

Chapter 4

Singlet positronium in the field of two near-resonant lasers without fluorescence

Our goal is to obtain the lifetime of singlet Ps atoms interacting with two circularly polarized near-resonant lasers.[1] In Chapter 2, section 1 we studied this system without taking into consideration the possibility that the atom annihilates in the ground state and is ionized by the lasers in the excited

[1] See Figure 1.2.

states. These processes are incorporated into the problem, which is achieved by introducing a complex potential. This leads to the following nonhermitian Hamiltonian:

$$H = H_0 + H_A - iV \ , \tag{4.1}$$

where

$$V \ = \ \frac{1}{2} \left(R_0 \, |u_0\rangle\langle u_0| \ + \ R_1 \, |u_1\rangle\langle u_1| \ + \ R_2 \, |u_2\rangle\langle u_2| \right) \ . \tag{4.2}$$

The coefficient $R_0 = \Gamma$ is the annihilation rate given in (1.7), while $R_1 = w^a_{2P}$ is the ionization rate in (1.24) from the state $|2P\rangle$ induced by laser 'a' and $R_2 = w^a_{3D}$ is the ionization rate in (1.25) from the state $|3D\rangle$. As discussed in Chapter 1, section 5 and confirmed by the results in Chapter 3, ionization with laser 'b' is insignificant and it will not be considered here. As before, H_0 is the Hamiltonian of the Ps atom outside of the laser fields and H_A as in eq. (2.2) and (2.4) gives the interaction with the lasers.

We follow the approach of Chapter 2, section 1 and the formalism developed therein with the difference that the Hamiltonian contains an absorptive term. We want to obtain the solution of the Schrodinger equation with Hamiltonian (4.1), which we seek as a wave function of the form (2.6):

$$|\bar{\Psi}(t)\rangle \;=\; \bar{\alpha}(t)\,|u_0\rangle\, e^{-iW_0 t} \;+\; \bar{\beta}(t)\,|u_1\rangle\, e^{-iW_1 t} + \;\bar{\gamma}(t)\,|u_2\rangle\, e^{-iW_2 t}\,.$$

The unknown amplitudes depend on time as in (2.10):

$$\begin{aligned}
\bar{\alpha}(t) &= \alpha\, e^{-i(\varepsilon-\Delta\omega_a)t/2} \\
\bar{\beta}(t) &= \beta\, e^{-i(\varepsilon-\Delta\omega_a+2\Delta\omega_b)t/2} \\
\bar{\gamma}(t) &= \gamma\, e^{-i(\varepsilon+\Delta\omega_a)t/2}\,.
\end{aligned}$$

The difference with the problem considered before is that here the Rabi frequency ε can be complex. This is a consequence of the nonhermitian term in the Hamiltonian and reflects the loss of Ps due to annihilation and ionization. Substituting this wave function back into the Schrodinger equation and using the rotating wave approximation, yields a new eigenvalue problem for ε:

$$\begin{aligned}
(\varepsilon - \Delta\omega_a + iR_0)\,\alpha &= \beta\,\Lambda_b + \gamma\,\Lambda_a \\
(\varepsilon - \Delta\omega_a + 2\Delta\omega_b + iR_1)\,\beta &= \alpha\,\Lambda_b^* \qquad (4.3) \\
(\varepsilon + \Delta\omega_a + iR_2)\,\gamma &= \alpha\,\Lambda_a^*\,.
\end{aligned}$$

The corresponding eigenvalue equation has now additional terms, which contain the annihilation and ionization rates:

$$
\begin{aligned}
0 \;=\; & \varepsilon^3 + \varepsilon^2\{-\Delta\omega_a + 2\Delta\omega_b + i(R_0 + R_1 + R_2)\} - \varepsilon\{\Delta\omega_a^2 + |\Lambda_a|^2 + \\
& |\Lambda_b|^2 - R_0R_1 - R_0R_2 - R_1R_2 + 2i(R_0\Delta\omega_b + R_2\Delta\omega_b - R_2\Delta\omega_a)\} + \\
& (\Delta\omega_a - 2\Delta\omega_b)(|\Lambda_a|^2 + \Delta\omega_a^2) - |\Lambda_b|^2\Delta\omega_a + R_1R_2\Delta\omega_a - \\
& R_0R_1\Delta\omega_a + R_0R_2(\Delta\omega_a - 2\Delta\omega_b) + iR_0\Delta\omega_a(2\Delta\omega_b - \Delta\omega_a) - \\
& iR_1\Delta\omega_a^2 + iR_2\Delta\omega_a(\Delta\omega_a - 2\Delta\omega_b) - iR_0R_1R_2 - iR_1|\Lambda_a|^2 - iR_2|\Lambda_b|^2
\end{aligned}
$$

It is tractable only numerically (even at resonance). The solution gives three complex eigenvalues ε_i. They are substituted back in eq. (4.3) in order to find the corresponding eigenvectors:

$$
\begin{aligned}
\alpha(\varepsilon_i) &= \frac{\varepsilon_i + \Delta\omega_a + iR_2}{\Lambda_a^*}\,\gamma(\varepsilon_i) \qquad (4.4)\\
\beta(\varepsilon_i) &= \frac{\varepsilon_i^2 - \Delta\omega_a^2 - |\Lambda_a|^2 + i\varepsilon_i(R_2 + R_0) + i\Delta\omega_a(R_0 - R_2) - R_0R_2}{\Lambda_a^*\Lambda_b}\,\gamma(\varepsilon_i)
\end{aligned}
$$

Since the Hamiltonian is not hermitian, the eigenvectors of eq. (4.3) and therefore the wavefunctions $|\Psi_i(t)\rangle$ are not orthogonal. Nevertheless, the latter can be normalized at t=0, which leads to the condition:

$$|\alpha(\varepsilon_i)|^2 + |\beta(\varepsilon_i)|^2 + |\gamma(\varepsilon_i)|^2 = 1 \qquad (4.5)$$

for $i = 1, 2, 3$. Eq. (4.4) and (4.5) determine $|\gamma(\varepsilon_i)|$. We choose $\gamma(\varepsilon_i)$ to be real and obtain the following expression:

$$\gamma(\varepsilon_i) = \Big(1 + |\Lambda_a|^{-2}\left[(x_i+\Delta\omega_a)^2+(y_i+R_2)^2\right] + |\Lambda_a|^{-2}|\Lambda_b|^{-2}\left\{\left[x_i^2+y_i^2-\Delta\omega_a^2-|\Lambda_a|^2-y_i(R_0+R_2)-R_0R_2\right]^2 + \left[2x_iy_i+x_i(R_0+R_2)+\Delta\omega_a(R_0-R_2)\right]^2\right\}\Big)^{-\frac{1}{2}} . \quad (4.6)$$

We have denoted by x_i the real part of the complex Rabi frequency ε_i and by y_i - its imaginary part.

The total wave function of the system is a linear superposition of the dressed states $|\Psi_i(t)\rangle$:

$$|\Phi_l(t)\rangle = \sum_{i=1}^{3} C_{li}\,|\Psi_i(t)\rangle . \quad (4.7)$$

The constants C_{li} are determined by the initial conditions. The index l in (4.8) denotes in which bare state was the Ps atom at t=0. For example, if Ps was initially in its ground state, we have:

$$|\Phi_0(t=0)\rangle \equiv |u_0\rangle = \sum_{i=1}^{3} C_{0i}\,|\Psi_i(t=0)\rangle , \quad (4.8)$$

where

$$|\Psi_i(t=0)\rangle = \alpha(\varepsilon_i)\,|u_0\rangle + \beta(\varepsilon_i)\,|u_1\rangle + \gamma(\varepsilon_i)\,|u_2\rangle . \quad (4.9)$$

Since $|\Psi_i\rangle$ are not orthogonal, it is convenient to determine C_{0i} by multiplying eq. (4.8) from the left with the bare states $|u_j\rangle$ for j=0,1,2 and use the

orthonormality of the latter. We obtain a system of algebraic equations, which has the following solutions:

$$\begin{aligned} C_{02} &= -C_{01}\,\frac{\beta_1\gamma_3-\gamma_1\beta_3}{\beta_2\gamma_3-\gamma_2\beta_3} = -C_{01}D_{02} \\ C_{03} &= -C_{01}\,\frac{\beta_1\gamma_2-\gamma_1\beta_2}{\gamma_2\beta_3-\beta_2\gamma_3} = -C_{01}D_{03} \\ C_{01} &= (\alpha_1-\alpha_2D_{02}-\alpha_3D_{03})^{-1}\,. \end{aligned} \tag{4.10}$$

The first two equations define the parameters D_{02} and D_{03}. In the same way we can find the constants C_{1i} when the system had other initial conditions. When Ps was initially in the state $|u_1\rangle$, we have:

$$\begin{aligned} C_{12} &= -C_{11}\,\frac{\alpha_1\gamma_3-\gamma_1\alpha_3}{\alpha_2\gamma_3-\gamma_2\alpha_3} = -C_{11}D_{12} \\ C_{13} &= -C_{11}\,\frac{\alpha_1\gamma_2-\gamma_1\alpha_2}{\gamma_2\alpha_3-\alpha_2\gamma_3} = -C_{11}D_{13} \\ C_{11} &= (\beta_1-\beta_2D_{12}-\beta_3D_{13})^{-1}\,. \end{aligned} \tag{4.11}$$

If the initial state was $|u_2\rangle$, the coefficients are:

$$\begin{aligned} C_{22} &= -C_{21}\,\frac{\alpha_1\beta_3-bb_1\alpha_3}{\alpha_2\beta_3-\beta_2\alpha_3} = -C_{21}D_{22} \\ C_{23} &= -C_{21}\,\frac{\alpha_1\beta_2-\beta_1\alpha_2}{\beta_2\alpha_3-\alpha_2\beta_3} = -C_{21}D_{23} \\ C_{21} &= (\gamma_1-\gamma_2D_{22}-\gamma_3D_{23})^{-1}\,. \end{aligned} \tag{4.12}$$

If annihilation and ionization are not considered, i.e. $V \equiv 0$, then the coeficients in the expansion of the wave function (4.7) onto the dressed states $|\Psi_i(t)\rangle$ can be found by multilying eq. (4.8) to the left with another dressed state and use the orthonormality of the latter. We obtain:

$$\begin{aligned} \langle \Psi_l(t=0)|u_0\rangle \;&=\; \alpha_l^* \;= \\ &= \sum_{i=1}^{3} C_{0i}\, \langle \Psi_l(t=0)|\Psi_i(t=0)\rangle = \sum_{i=1}^{3} C_{0i}\, \delta_{li} = C_{0l} \end{aligned} \tag{4.13}$$

The coefficients are real and we have:

$$C_{0i} = \alpha(\varepsilon_i), \qquad C_{1i} = \beta(\varepsilon_i) \qquad \text{and} \qquad C_{2i} = \gamma(\varepsilon_i)\,. \tag{4.14}$$

We return to the problem with absorption. The probability to find a bound state of Ps at time $t > 0$ for initial condition $|u_0\rangle$ is:

$$\langle \Phi_0(t)|\Phi_0(t)\rangle \;=\; \sum_{ij} C_{0i}^*\, C_{0j}\; e^{-i(\epsilon_j - \epsilon_i^*)t/2}\, (\alpha_i^*\alpha_j + \beta_i^*\beta_j + \gamma_i^*\gamma_j)\,. \tag{4.15}$$

Similar expressions can be obtained for the other initial conditions by substituting the index '0' with '1' or '2'.

We define an effective time dependent decay rate $\Gamma_l(t)$ by the following equation:

$$\langle \Phi_l(t)|\Phi_l(t)\rangle \;=\; e^{-\int_0^t dt'\, \Gamma_l(t')}\,, \tag{4.16}$$

where the index l indicates the initial state. From the above definition we obtain:

$$\Gamma_l(t) \;=\; -\,\frac{\frac{d}{dt}\langle\Phi_l(t)|\Phi_l(t)\rangle}{\langle\Phi_l(t)|\Phi_l(t)\rangle} \;=\; -\frac{d}{dt}\,ln\,\langle\Phi_l(t)|\Phi_l(t)\rangle\,. \tag{4.17}$$

It is straightforward to find an expression for $\Gamma_l(t)$ by taking directly the time derivative of eq. (4.15). The effective time-dependent decay rate is equal to:

$$\frac{i}{2}\,\frac{\sum C^*_{li}\,C_{lj}\,(\varepsilon_j-\varepsilon^*_i)\,e^{-i(\varepsilon_j-\varepsilon^*_i)t/2}\,(\alpha^*_i\alpha_j+\beta^*_i\beta_j+\gamma^*_i\gamma_j)}{\langle\Phi_l(t)|\Phi_l(t)\rangle}\,. \tag{4.18}$$

It is easier to see the physical meaning of Γ_l by using eq. (2.45). For a system with a nonhermitian Hamiltonian as in eq. (4.1), the differential equation for physical observables is:

$$i\frac{d}{dt}\langle\Psi|\mathcal{O}|\Psi\rangle = \langle\Psi|(\mathcal{O}H - H^\dagger\mathcal{O})|\Psi\rangle. \tag{4.19}$$

If the operator $\mathcal{O}$ is given by the unit operator and the wave function - by eq. (4.7), we obtain:

$$i\frac{d}{dt}\langle\Phi_l(t)|\Phi_l(t)\rangle \;=\; \langle\Phi_l(t)|(H - H^\dagger)|\Phi_l(t)\rangle. \tag{4.20}$$

This leads to an expression for the effective decay rate in terms of the absorptive potential V:

$$\Gamma_l(t) \;=\; 2\,\frac{\langle\Phi_l(t)|V|\Phi_l(t)\rangle}{\langle\Phi_l(t)|\Phi_l(t)\rangle}\,. \tag{4.21}$$

Using the explicit form of the potential, given in eq. (4.2) we obtain:

$$\Gamma_l(t) \;=\; R_0\,\frac{|\langle\Phi_l(t)|u_0\rangle|^2}{\langle\Phi_l(t)|\Phi_l(t)\rangle} \;+\; R_1\,\frac{|\langle\Phi_l(t)|u_1\rangle|^2}{\langle\Phi_l(t)|\Phi_l(t)\rangle} \;+\; R_2\,\frac{|\langle\Phi_l(t)|u_2\rangle|^2}{\langle\Phi_l(t)|\Phi_l(t)\rangle}\,. \quad (4.22)$$

Therefore the effective decay rate defined in (4.17) is equal to the following sum: the annihilation rate of singlet Ps outside of the laser fields times the probability to find the ground state in the total wave function of the system for given initial conditions; plus the ionization rate from the bare state 2P, times the probability to find this state; plus the ionization rate from 3D, times the probability to find this state. It is the analogue of eq. (3.45) for singlet Ps interacting with one laser. Note that the latter expression was found by the projection operator method in a Hilbert space comprising bound Ps states as well as continuum states, while eq. (4.22) was found here by introducing a complex potential, which is a more straightforward method. By substituting eq. (4.7) for the wave function in (4.22), we obtain:

$$\Gamma_l(t) \;=\; \frac{\sum C_{li}^*\,C_{lj}\,e^{-i(\epsilon_j-\epsilon_i^*)t/2}\,(R_0\,\alpha_i^*\alpha_j \;+\; R_1\,\beta_i^*\beta_j \;+\; R_2\,\gamma_i^*\gamma_j)}{\langle\Phi_l(t)|\Phi_l(t)\rangle}\,, \quad (4.23)$$

where the denominator is given in eq. (4.15). This expression or eq. (4.18) can be used to numerically find the effective time-dependent decay rate.

Chapter 5

Triplet positronium in the field of one near-resonant laser

In this chapter we obtain the lifetime of triplet Ps in the field of one near-resonant circularly polarized laser. Annihilation and ionization are incorporated into the problem via a complex potential as in Chapter 4, which leads to a nonhermitian Hamiltonian. Spontaneous radiative transitions are included by the formalism introduced in Chapter 2, section 2. The results can be obtained analytically at resonance.

The system has the following Hamiltonian:

$$H = H_0 + H_A + H_R - iV\,. \tag{5.1}$$

As before, the time evolution due to the free Hamiltonian of the laser field is absorbed into the wave function and H_0 is the free Hamiltonian of the Ps atom outside of the laser. The interaction with the latter is given by the term:

$$H_A = \frac{e}{c}\,\vec{A}(t)\cdot\vec{p}, \tag{5.2}$$

which is in the dipole approximation and the velocity gauge as in Chapter 2. The vector potential is:

$$\vec{A}(t) \;=\; \frac{E}{2\omega}\left(\hat{\varepsilon}\;e^{i\omega t} \;+\; \hat{\varepsilon}^{*}\;e^{-i\omega t}\right). \tag{5.3}$$

The laser is circularly polarized with polarization vector:

$$\hat{\varepsilon} \;=\; \frac{1}{2}(\hat{x} \pm i\hat{y})\,. \tag{5.4}$$

$H_R = e\vec{E}_R(t)\cdot\vec{r}$ gives the interaction of the Ps atom with the quantized field of the radiation photons, which is defined in eq. (2.34). The Hamiltonian in eq. (5.1) is nonhermitian due to the last term, which gives the possibility for annihilation in the ground state and for ionization by the laser in the excited

state:

$$V = \frac{1}{2}\left(R_0\,|u_0\rangle\langle u_0| + R_1\,|u_1\rangle\langle u_1|\right) . \tag{5.5}$$

The coefficient $R_0 = \Gamma$ is the annihilation rate in eq. (1.7) and $R_1 = w^b_{2P}$ is the ionization rate in eq. (1.26) from the state $|2P\rangle$ induced by laser 'b'.

Instead of finding the dressed states analogous to eq. (2.6) and (2.10) and working in their representation, it is convenient to use the following time-dependent basis of the bare states:

$$\begin{aligned} |q_0\rangle &= |u_0\rangle\, e^{-iW_0 t}\, e^{i\Delta\omega t/2} \\ |q_1\rangle &= |u_1\rangle\, e^{-iW_1 t}\, e^{-i\Delta\omega t/2} , \end{aligned} \tag{5.6}$$

where $\Delta\omega$ is defined in eq. (1.3). We have:

$$|q_0\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix} \qquad \text{and} \qquad |q_1\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix} .$$

Within the rotating wave approximation[1], we obtain in this representation:

$$H_0 = \frac{\Delta\omega}{2}\,\sigma_3 \qquad \text{and} \qquad H_A = \frac{\Lambda}{2}\,\sigma_1 , \tag{5.7}$$

where σ_i for i=1,2,3 denotes the Pauli matrices. The potential V can be

[1]For a detailed discussion on the RWA see Chapter 2, section 1.

written as:

$$V = \frac{\sigma_4}{2}\bar{R} + \frac{\sigma_3}{2}\Delta R, \tag{5.8}$$

where σ_4 is the unit matrix. The above parameters can be expressed in terms of the annihilation and ionization rates:

$$\bar{R} = \frac{1}{2}(R_0 + R_1) \quad \text{and} \quad \Delta R = \frac{1}{2}(R_0 - R_1)\,. \tag{5.9}$$

It is interesting to investigate how the complex potential influences the behaviour of the system. Let us assume for a moment that we do not have spontaneous radiative transitions. In this case the Hamiltonian is $H_1 = H_0 + H_A - iV$ and it is time-independent in this representation:

$$H_1 = \frac{1}{2}\begin{pmatrix} \Delta\omega - i\Delta R - i\bar{R} & \Lambda \\ \Lambda & -\Delta\omega + i\Delta R - i\bar{R} \end{pmatrix}. \tag{5.10}$$

Its eigenvalues are:

$$\bar{\varepsilon}_{\pm} = \frac{1}{2}\left(-i\bar{R} \pm \sqrt{\Lambda^2 + (\Delta\omega - i\Delta R)^2}\right). \tag{5.11}$$

Comparing with the two state problem discussed in Chapter 1, section 2, we note that instead of the Rabi frequency ε in eq. (1.5), we have the complex quantity (5.11). The latter can be obtained from the Rabi frequency by the

substitution:

$$\bar{\varepsilon}_{\pm} \;=\; \frac{1}{2}\left[-i\bar{R} \,\pm\, \varepsilon\,(\Delta\omega \rightarrow \Delta\omega - i\Delta R)\right] \,. \tag{5.12}$$

$\bar{R}$ displaces the energy of the two dressed states $\Phi_{\pm}$ in (1.1) by a complex constant and therefore introduces an overall exponentially decaying factor in the wave function of the system. The new dressed states can be obtained from the old ones by multiplying with this factor and by substituting the detuning $\Delta\omega$ with $\Delta\omega - i\Delta R$. Note that at resonance ($\Delta\omega = 0$) the quantity under the square root in eq. (5.11) is real (if $|\Lambda| > |\Delta R|$) and the two dressed states have the same decay rate. The latter is equal to the average of the annihilation and ionization rates.

We now return to the problem with fluorescence. The radiative Hamiltonian is given by:

$$H_R \;=\; \vec{h}\cdot\vec{E}_R^{(+)}(t)\,\sigma_+ \;+\; h.c.\,, \tag{5.13}$$

where $\vec{E}_R^{(\pm)}$ are the frequency components of the quantized electric field of the radiation photons and

$$\vec{h} \;=\; \vec{d}_{01}\,e^{-i\omega t}\,. \tag{5.14}$$

We have denoted by:

$$\sigma_{\pm} \;=\; \frac{1}{2}(\sigma_1 \pm i\sigma_2) \tag{5.15}$$

and (as in Chapter 2) the dipole operator by $d_{01} = e\langle u_0|\vec{r}|u_1\rangle$. As before, we use an approximate solution for the electric field of the radiation photons, which is given as an approximate constraint on the wave function of the system. The latter is derived in Appendix C. In the representation considered here, it is given by the expression:

$$\vec{E}_R^{(-)}(t)|\Psi(t)\rangle \;=\; -i\frac{2}{3}\,W_{10}^3\,\vec{d_{01}}\,e^{-i\omega t}\,\sigma_+\,|\Psi(t)\rangle \tag{5.16}$$

A detailed discussion on the formalism introducing the constraint is given in Chapter 2, section 2.

Physical observables can be expressed as expectation values of operators with respect to the total wave function of the system $|\Psi(t)\rangle$. For a system with a nonhermitian Hamiltonian as in eq. (5.1), the differential equation for these observables is: [2]

$$\begin{aligned} i\frac{d}{dt}\langle\Psi|\mathcal{O}|\Psi\rangle \;&=\; \langle\Psi|(\mathcal{O}H - H^\dagger\mathcal{O})|\Psi\rangle \;= \\ &=\; \langle\Psi|[\mathcal{O},H']|\Psi\rangle - i\langle\Psi|\{\mathcal{O},V\}|\Psi\rangle\;, \end{aligned} \tag{5.17}$$

where $H' = H_0 + H_A + H_R$ is the hermitian part of the total Hamiltonian. If the operator $\mathcal{O}$ for a given observable is independent of the radiation

[2] And also the operator $\mathcal{O}$ does not depend explicitly on time.

operators, we obtain:

$$\begin{aligned} i\frac{d}{dt}\langle\Psi|\mathcal{O}|\Psi\rangle &= \frac{\Delta\omega}{2}\langle\Psi|[\mathcal{O},\sigma_3]|\Psi\rangle + \frac{\Lambda}{2}\langle\Psi|[\mathcal{O},\sigma_1]|\Psi\rangle \\ &+ \vec{h}\cdot\langle\Psi|\,\vec{E}_R^{(+)}(t)\,[\mathcal{O},\sigma_+]|\Psi\rangle \\ &+ \vec{h}^{\dagger}\cdot\langle\Psi|\,\vec{E}_R^{(-)}(t)\,[\mathcal{O},\sigma_-]|\Psi\rangle \\ &- i\bar{R}\,\langle\Psi|\mathcal{O}|\Psi\rangle - i\frac{\Delta R}{2}\langle\Psi|\{\mathcal{O},\sigma_3\}|\Psi\rangle\,. \end{aligned} \tag{5.18}$$

Note that the term in the third line of eq. (5.18) is equal to minus the hermitian conjugated of the preceding term.

Any two dimensional hermitian matrix can be expanded over the set $\{\sigma_a\}$ for a=1...4. Therefore we can find the expectation value of any operator if we know:

$$S_a = \langle\Psi|\sigma_a|\Psi\rangle \qquad \text{for a=1,...4.} \tag{5.19}$$

In order to find an equation for S_a from (5.19), we make use of the constraints (5.16) and of the algebra of the Pauli matrices:

$$\begin{aligned} [\sigma_i,\ \sigma_j] &= 2i\varepsilon_{ijk}\,\sigma_k\,, \\ \{\sigma_i,\ \sigma_j\} &= 2\delta_{ij}\,. \end{aligned} \tag{5.20}$$

We obtain the following homogeneous system of linear differential equations

with constant coefficients[3] for the vector S_a:

$$\frac{d}{dt}S_a + M_{ab}S_b = 0\,, \tag{5.21}$$

where the matrix M_{ab} is given by:

$$M = \begin{pmatrix} \gamma+\bar{R} & \Delta\omega & 0 & 0 \\ -\Delta\omega & \gamma+\bar{R} & \Lambda & 0 \\ 0 & -\Lambda & 2\gamma+\bar{R} & \Delta R-2\gamma \\ 0 & 0 & \Delta R & \bar{R} \end{pmatrix} \tag{5.22}$$

The parameter γ is half of the natural decay rate of the state 2P and is defined by the first eqation in (2.54). If the Ps atom is initially in its ground state, i.e. $|\Psi(t=0)\rangle = |u_0\rangle$, the initial condition for the above system of differential equations is:

$$S_a(t=0) = \begin{pmatrix} 0 \\ 0 \\ 1 \\ 1 \end{pmatrix}. \tag{5.23}$$

[3]The representation (5.6) was chosen so that the coefficients are constant.

In the case when there is no annihilation and ionization (V=0), the system (5.21) reduces to a set of three inhomogeneous equations. Since there is no decay, it is sufficient to find the solution for large times, i.e. $t >> \hbar/\gamma$ as was done in Chapter 2, section 2. The calculations for the two-state problem can be performed analytically. As expected, the result for the probability to find any of the bound states coincides with the expressions in [20] (Chapter 4), obtained in the representation of the dressed states.

When annihilation and ionization are included via a complex potential, the dressed states are decaying exponentially. Therefore we need to find the comlete time-dependent solution of the homogeneous system (5.21). If the number of the linearly independent eigenvectors of the matrix M in eq. (5.22) is equal to its dimension, the solution is given by [44]:

$$S_a(t) = \sum_{i=1}^{4} c_i \, v_a^{(i)} \, e^{-m_i t} , \tag{5.24}$$

where m_i are the eigenvalues and v^i are the corresponding eigenvectors of M. The constants c_i are determined from the initial conditions (5.23).

Note that at resonance ($\Delta\omega = 0$), the first equation decouples from the rest. This reduces the solution to the one of a three dimensional eigenvalue

problem. It is convenient to make the substitution:

$$m = m' + \gamma + \bar{R} \tag{5.25}$$

These simplifications lead to the following matrix:

$$M' = \begin{pmatrix} -m' & \Lambda & 0 \\ -\Lambda & \gamma - m' & \Delta R - 2\gamma \\ 0 & \Delta R & -m' - \gamma \end{pmatrix} \tag{5.26}$$

The corresponding eigenvalue equation is:

$$m'^3 + m' \left(|\Lambda|^2 + \Delta R^2 + 2\gamma \Delta R - \gamma^2 \right) + |\Lambda|^2 \gamma = 0 \tag{5.27}$$

The constraint is an expansion in the coefficient γ/W_{10} and subsequent results are accurate only to lowest order in this parameter. In principle, the eigenvalues of the matrix (5.26) can be found exactly. But it is sufficient to write them as a power series in γ/Λ. This is a high intensity expansion, which holds for $\gamma/\Lambda < 1$ or $E^b/E^b_{crit} > 22.3$. Let us assume first that $\gamma = 0$ and there are no spontaneous radiative transitions. In this case the eigenvalues are:

$$m'_2 = 0\,, \qquad m'_{3,4} = \pm i\,\varepsilon\,, \qquad \text{where} \qquad \varepsilon = \sqrt{|\Lambda|^2 - \Delta R^2}\,. \tag{5.28}$$

Next we seek the eigenvalues as an expansion in powers of γ/Λ with the zeroth order given by eq. (5.28). Up to second order we obtain:

$$\begin{aligned} m_2 &= \bar{R} + \gamma\left(1 - \frac{|\Lambda|^2}{\varepsilon^2}\right) + \gamma^2 \frac{2\Delta R\,|\Lambda|^2}{\varepsilon^4}\,, \\ m_{3,4} &= \pm i\varepsilon + \bar{R} + \gamma\left(1 + \frac{|\Lambda|^2}{2\varepsilon^2} \pm i\frac{\Delta R}{\varepsilon}\right) \\ &\quad - \gamma^2\left[\frac{|\Lambda|^2\,\Delta R}{\varepsilon^4} \pm \frac{i}{2\varepsilon}\left(1 + \frac{\Delta R^2}{\varepsilon^2} - \frac{3|\Lambda|^4}{4\varepsilon^4}\right)\right] \end{aligned} \tag{5.29}$$

It is important to note that $Re\, m_i > 0$, so that all terms in eq. (5.24) vanish exponentially at large time. Up to first order the corresponding eigenvectors are:

$$v^{(2)} = \frac{1}{\sqrt{\gamma^2 + \frac{\varepsilon^4}{|\Lambda|^2} + \frac{\Delta R^2}{(1-\frac{|\Lambda|^2}{\varepsilon^2})^2}}} \begin{pmatrix} -\frac{\varepsilon^2}{\Lambda} \\ \gamma \\ -\frac{\Delta R}{1-\frac{|\Lambda|^2}{\varepsilon^2}} \end{pmatrix}, \tag{5.30}$$

$$v^{(3,4)} = \frac{1}{\sqrt{\gamma^2 + \frac{|\Lambda|^2}{|p|^2} + \frac{\Delta R^2}{|1+p|^2}}} \begin{pmatrix} -\frac{\Lambda}{p} \\ \gamma \\ -\frac{\Delta R}{1+p} \end{pmatrix}, \quad \text{where} \quad p = \frac{|\Lambda|^2}{2\varepsilon^2} \pm i\frac{\Delta R}{\varepsilon}\,.$$

The probability to find a bound Ps state is:

$$S_4 \;=\; \langle\Psi(t)|\Psi(t)\rangle \;=\; c_2\, v_4^{(2)}\, e^{-m_2 t} \;+\; c_3\, v_4^{(3)}\, e^{-m_3 t} \;+\; c_4\, v_4^{(4)}\, e^{-m_4 t}\;. \quad (5.31)$$

The coefficients $c_i\, v_4^{(i)}$ are of the same order of magnitude as can be seen from the above expressions for the eigenvectors and the initial condition in eq. (5.23). Therefore the longest surviving term in eq. (5.31) will be the one for which $Re\, m_i$ is smallest. From eq. (5.29) we find that this is m_2:

$$Re\,(m_3 - m_2) \;=\; \gamma\,\frac{3|\Lambda|^2}{2\varepsilon^2} \;-\; \gamma^2\,\frac{3|\Lambda|^2\Delta R}{\varepsilon^4} \;>\; 0\;. \quad (5.32)$$

The difference between the two eigenvalues is approximatively equal to $\frac{3}{2}\gamma$ for laser fields $10 < E^b/E^b_{crit} < 10^4$, which are the intensities of interest.

The above analysis leads to the conclusion that the eigenvalue m_2 can be considered as the decay rate of Ps. Up to second order in $\gamma/|\Lambda|$ we have:

$$\Gamma \;=\; \frac{1}{2}\left(\Gamma_0 + w^b_{2P}\right) \;+\; \gamma\left(1 - \frac{|\Lambda|^2}{\varepsilon^2}\right) \;+\; \gamma^2\,\frac{(\Gamma_0 - w^b_{2P})\,|\Lambda|^2}{\varepsilon^4}\;. \quad (5.33)$$

This expression gives the decay rate of triplet Ps in the field of one laser, at resonance, up to second order in γ/Λ. It includes the possibility for ionization from the state 2P. We have found earlier[4] that in this case ionization is

[4]See Chapter 1, section 5.

not significant. Therefore the this result has more theoretical than practical importance. The method used here will be applied to the three state problem considered in Chapter 6 and will enable us to find the lifetime of triplet, as well as of singlet Ps.

Note that when ionization is not present, we have:

$$\bar{R} = \Delta R = \frac{\Gamma_0}{2} \qquad \text{and} \qquad \varepsilon \approx \Lambda \,. \tag{5.34}$$

Then eq. (5.32) reduces to:

$$\Gamma = \frac{\Gamma_0}{2}\left(1 + \frac{2\gamma^2}{|\Lambda|^2}\right) + O\left(\frac{\gamma^3}{|\Lambda|^3}\right), \tag{5.35}$$

which is exactly the result in eq. (1.10) up to second order in γ/Λ, obtained in [].

Chapter 6

The complete problem of positronium in the field of two near-resonant lasers

6.1 Formulation of the problem

In this chapter we obtain the lifetime of Ps atoms interacting with two circularly polarized near-resonant lasers[1]. In Chapter 2 we studied this system

[1]See Figure 1.2.

without taking into consideration that the atom can annihilate in the ground state and can be ionized by the lasers in the excited states. These processes are incorporated here into the problem by adding a complex potential to the Hamiltonian as it was done in Capter 4 and 5. Spontaneous radiative transitions are included by the formalism introduced in Chapter 2, section 2.

The system has the following Hamiltonian:

$$H = H_0 + H_A + H_R - iV \ , \tag{6.1}$$

where

$$V \ = \ \frac{1}{2} \left(R_0 \left| u_0 \right\rangle \left\langle u_0 \right| \ + \ R_1 \left| u_1 \right\rangle \left\langle u_1 \right| \ + \ R_2 \left| u_2 \right\rangle \left\langle u_2 \right| \right) . \tag{6.2}$$

It is nonhermitian due to the last term, which gives the possibility for decay of the bound Ps states into a free electron-positron pair or into high energy gamma quanta. The coefficient $R_0 = \Gamma_0$ is the annihilation rate given in eq. (1.7), while $R_1 = w^a_{2P}$ is the ionization rate in eq. (1.24) from the state $|2P\rangle$ induced by laser 'a' and $R_2 = w^a_{3D}$ is the ionization rate in eq. (1.25) from the state $|3D\rangle$. As discussed in Chapter 1, section 5 ionization with laser 'b' is insignificant and it will not be considered here.

As before, the time evolution due to the free Hamiltonian of the laser field is absorbed into the wave function and H_0 is the free Hamiltonian of

the Ps atom outside of the lasers. The interaction with the latter is given by the term:

$$H_A = \frac{e}{m} \sum_{j=a,b} \vec{A}_j(t) \cdot \vec{p}, \tag{6.3}$$

which is in the dipole approximation and the velocity gauge as in Chapter 2 and 5. The vector potentials are:

$$\vec{A}_j(t) = \frac{E_j}{2\omega_j} \left(\hat{\varepsilon}_j \, e^{i\omega_j t} + \hat{\varepsilon}_j^* \, e^{-i\omega_j t} \right), \tag{6.4}$$

where E_j are the amplitudes of the laser electric fields. The laser is circularly polarized and the polarization vectors are given in eq. (2.5). $H_R = e\vec{E}_R(t) \cdot \vec{r}$ gives the interaction of the Ps atom with the quantized field of the radiation photons, which is defined in eq. (2.34).

Instead of using the representation of the dressed states as was done in Chapter 2, we adopt here the approach of Chapter 5 and work in the following time-dependent basis of the bare states:

$$\begin{aligned} |q_0\rangle &= |u_0\rangle \, e^{-iW_0 t} \, e^{i\Delta\omega_a t/2} \\ |q_1\rangle &= |u_1\rangle \, e^{-iW_1 t} \, e^{i(\Delta\omega_a - 2\Delta\omega_b)t/2} \\ |q_2\rangle &= |u_2\rangle \, e^{-iW_2 t} \, e^{-i\Delta\omega_a t/2} \end{aligned} \tag{6.5}$$

where $\Delta\omega_a$ and $\Delta\omega_b$ are defined in eq. (1.11) and (1.12) respectively. We

have:

$$|q_0\rangle = \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}, \qquad |q_0\rangle = \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} \quad \text{and} \quad |q_1\rangle = \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}.$$

Using the rotating wave approximation[2] we obtain the matrices of the different terms in eq. (6.1) in this representation. The free Hamiltonian and the interaction with the lasers are given by:

$$H_0 + H_A = \frac{1}{2} \begin{pmatrix} \Delta\omega_a & \Lambda_b & \Lambda_a \\ \Lambda_b & \Delta\omega_a - 2\Delta\omega_b & 0 \\ \Lambda_a & 0 & -\Delta\omega_a \end{pmatrix}. \tag{6.6}$$

Using the Gell-Mann matrices from eq. (2.37) as a basis, we obtain:

$$H_0 + H_A = \sum_{a=1}^{9} X_a \lambda_a , \tag{6.7}$$

where

$$\begin{aligned} X_1 &= \frac{\Lambda_b}{2}, \qquad X_3 = \frac{\Delta\omega_b}{2}, \qquad X_4 = \frac{\Lambda_a}{2}, \\ X_8 &= \frac{1}{2\sqrt{3}}(2\Delta\omega_a - \Delta\omega_b), \qquad X_9 = \frac{1}{6}(\Delta\omega_a - 2\Delta\omega_b) \end{aligned} \tag{6.8}$$

[2]For a discussion on the RWA see Chapter 2, section 1.

and all other X_a are zero. The term giving the interaction with the radiation field is:

$$H_R = \begin{pmatrix} 0 & \vec{h}_1 \cdot \vec{E}_R^{10(+)}(t) & 0 \\ \vec{h}_1^{\dagger} \cdot \vec{E}_R^{10(-)}(t) & 0 & \vec{h}_2 \cdot \vec{E}_R^{21(+)}(t) \\ 0 & \vec{h}_2^{\dagger} \cdot \vec{E}_R^{21(-)}(t) & 0 \end{pmatrix} . \quad (6.9)$$

We have used a notation similar to the one in Chapter 2 and 5:

$$\vec{h}_1 = \vec{d}_{01}\, e^{-i\omega_b t} \quad \text{and} \quad \vec{h}_2 = \vec{d}_{12}\, e^{-i(2\omega_a - \omega_b)t} . \quad (6.10)$$

As in Capter 2, we have split the radiation field operator into two components depending on the value of the photon wave vector:

$$\vec{E}_R = \vec{E}_R^{21} + \vec{E}_R^{10}. \quad (6.11)$$

$\vec{E}_R^{21}$ includes photons with energy within a narrow interval around W_{21}, while $\vec{E}_R^{10}$ refers to photons resulting from the other radiative transition with energy within a narrow interval around W_{10}.[3] The superscript $(\pm)$ refers respectively to the positive and negative frequency components of these operators. Ex-

[3]The width of the interval is of the order of the width of the states.

panding on the basis set of matrices, we obtain:

$$\begin{aligned} H_R &= \vec{h}_1 \cdot \vec{E}_R^{10(+)}(t)\,\frac{1}{2}(\lambda_1 + i\lambda_2) \;+\; h.c. \\ &\quad + \vec{h}_2 \cdot \vec{E}_R^{21(+)}(t)\,\frac{1}{2}(\lambda_6 + i\lambda_7) \;+\; h.c. \end{aligned} \tag{6.12}$$

In this representation the decay term in the Hamiltonian is given by:

$$V = \frac{1}{2}\begin{pmatrix} R_0 & 0 & 0 \\ 0 & R_1 & 0 \\ 0 & 0 & R_2 \end{pmatrix}, \tag{6.13}$$

which can be expanded on the Gell-Mann matrices as follows:

$$V = \sum_{a=1}^{9} V_a\,\lambda_a\,. \tag{6.14}$$

Using eq. (2.39), we find the following expressions for the coefficients:

$$\begin{aligned} V_3 &= \frac{1}{2}\,\Delta R \qquad \text{where} \qquad \Delta R = \frac{1}{2}(R_0 - R_1) \\ V_8 &= \frac{1}{2\sqrt{3}}(\bar{R}_{01} - R_2)\,, \qquad \text{where} \qquad \bar{R}_{01} = \frac{1}{2}(R_0 + R_1) \\ V_9 &= \frac{1}{6}(R_0 + R_1 + R_2)\,. \end{aligned} \tag{6.15}$$

We proceed with the formalism introduced in Chapter 2, section 2, which is also used in Chapter 5 for the two state problem. Physical observables can

be expressed as expectation values of operators with respect to the total wave function of the system $|\Psi(t)\rangle$. For a system with a nonhermitian Hamiltonian as in eq. (6.1), the differential equation for these observables is:

$$\begin{aligned} i\frac{d}{dt}\langle\Psi|\mathcal{O}|\Psi\rangle &= \langle\Psi|(\mathcal{O}H - H^{\dagger}\mathcal{O})|\Psi\rangle = \\ &= \langle\Psi|[\mathcal{O}, H']|\Psi\rangle - i\langle\Psi|\{\mathcal{O}, V\}|\Psi\rangle \,, \end{aligned} \tag{6.16}$$

where $H' = H_0 + H_A + H_R$ is the hermitian part of the total Hamiltonian. Any three dimensional hermitian matrix can be expanded over the set $\{\lambda_a\}$ for a=1...9. Therefore we can find the expectation value of any operator if we know:

$$S_a = \langle\Psi|\lambda_a|\Psi\rangle \qquad \text{for a=1,...9.} \tag{6.17}$$

When the operator $\mathcal{O}$ is one of the Gell-Mann matrices λ_a,[4] we obtain from eq. (1.6):

$$\begin{aligned} i\frac{d}{dt}\langle\Psi|\lambda_a|\Psi\rangle &= \sum_{b=1}^{8} X_b \langle\Psi|[\lambda_a, \lambda_b]|\Psi\rangle \\ &+ \vec{h}_1 \cdot \langle\Psi|\, \vec{E}_R^{10(+)}(t) \left[\lambda_a, \frac{1}{2}(\lambda_1 + i\lambda_2)\right] |\Psi\rangle + h.c. \\ &+ \vec{h}_2 \cdot \langle\Psi|\, \vec{E}_R^{21(+)}(t) \left[\lambda_a, \frac{1}{2}(\lambda_6 + i\lambda_7)\right] |\Psi\rangle + h.c. \end{aligned}$$

[4]The matrices λ_a are independent of the radiation operators and also do not depend on time explicitely.

$$+ \sum_{b=1}^{9} V_b \langle\Psi| \{\lambda_a, \lambda_b\} |\Psi\rangle \ , \tag{6.18}$$

where a=1...9. In order to further simplify this system of differential equations, we must use the constraint on the wave function.[5] For the problem considered here, it consists of two equations as in Chapter 2, section 2. If the radiation operator contains photons with energies around W_{10}, we have:

$$\vec{E}_R^{10(-)}(t) \, |\Psi(t)\rangle \ = \ -\frac{2i}{3} \, W_{10}^3 \ \vec{h}_1 \, \frac{1}{2}(\lambda_1 + i\lambda_2)|\Psi(t)\rangle \ . \tag{6.19}$$

The constraint for radiation photons resulting from the $3D \rightarrow 2P$ transition is:

$$\vec{E}_R^{21(-)}(t) \, |\Psi(t)\rangle \ = \ -\frac{2i}{3} \, W_{21}^3 \ \vec{h}_1 \, \frac{1}{2}(\lambda_6 + i\lambda_7)|\Psi(t)\rangle \ . \tag{6.20}$$

Using the algebra of the Gell-Mann matrices given in eq. (2.49), we obtain from eq. (6.18) the following system of homogeneous linear differential equations:

$$\frac{d}{dt} S_a \ + \ M_{ab} S_b \ = \ 0 \ , \tag{6.21}$$

where the matrix M_{ab} is given on the next page. The notation $\bar{R}_{ij} = \frac{1}{2}(R_i + R_j)$ is used as a shorthand. The parameters γ_{10} and γ_{21} are half of the natural decay rates of the states 2P and 3D respectively. They are defined

[5] A derivation of the constraint is given in Appendix C.

$$
\begin{pmatrix}
\gamma_{10}+\bar{R}_{01} & \Delta\omega_b & 0 & 0 & 0 & 0 & -\Lambda_a/2 & 0 & 0 \\
-\Delta\omega_b & \gamma_{10}+\bar{R}_{01} & \Lambda_b & 0 & 0 & -\Lambda_a/2 & 0 & 0 & 0 \\
0 & -\Lambda_b & 2\gamma_{10}+\bar{R}_{01} & 0 & -\Lambda_a/2 & 0 & 0 & -\frac{2}{\sqrt{3}}(\gamma_{10}+\gamma_{21})+\frac{\Delta R}{\sqrt{3}} & \frac{2}{3}(-2\gamma_{10}+\gamma_{21}+\Delta R) \\
0 & 0 & 0 & \gamma_{21}+\bar{R}_{02} & \Delta\omega_a & 0 & \Lambda_b/2 & 0 & 0 \\
0 & 0 & \Lambda_a/2 & -\Delta\omega_a & \gamma_{21}+\bar{R}_{02} & -\Lambda_b/2 & 0 & \frac{\sqrt{3}}{2}\Lambda_a & 0 \\
0 & \Lambda_a/2 & 0 & 0 & \Lambda_b/2 & \gamma_{10}+\gamma_{21}+\bar{R}_{12} & \Delta\omega_a-\Delta\omega_b & 0 & 0 \\
\Lambda_a/2 & 0 & 0 & -\Lambda_b/2 & 0 & \Delta\omega_b-\Delta\omega_a & \gamma_{10}+\gamma_{21}+\bar{R}_{12} & 0 & 0 \\
0 & 0 & \frac{1}{\sqrt{3}}\Delta R & 0 & -\frac{\sqrt{3}}{2}\Lambda_a & 0 & 0 & 2\gamma_{21}+\frac{1}{3}(\bar{R}_{01}+2R_2) & \frac{2}{\sqrt{3}}[-\gamma_{21}+\frac{1}{3}(\bar{R}_{01}-R_2)] \\
0 & 0 & \Delta R & 0 & 0 & 0 & 0 & \frac{1}{\sqrt{3}}(\bar{R}_{01}-R_2) & \frac{1}{3}(2\bar{R}_{01}+R_2)
\end{pmatrix}
$$

The matrix M in eq. (6.21).

in eq. (2.54). Since the dressed states are decaying exponentially, we need to find the complete time-dependent solution of the homogeneous system (6.21). In Chapter 2, section 2 we found the solution only for large times, because the dressed states are stable without annihilation and ionization. In this case the memory of the initial conditions is lost. The solution has the same form [44] as eq. (5.24):

$$S_a(t) = \sum_{i=1}^{9} c_i \, v_a^{(i)} \, e^{-m_i t} \,, \tag{6.22}$$

where m_i are the eigenvalues of the matrix M, $v^{(i)}$ are its eigenvectors and the constants c_i are to be determined from the initial conditions. If the Ps atom is initially in its ground state, i.e. $|\Psi(t=0)\rangle = |u_0\rangle$, the initial condition for

the above system of differential equations is:

$$S_a^{(1S)}(t=0) = \begin{pmatrix} 0 \\ 0 \\ 1 \\ 0 \\ 0 \\ 0 \\ 0 \\ \frac{1}{\sqrt{3}} \\ 1 \end{pmatrix} . \tag{6.23}$$

If Ps was initially in one of the excited states, we have:

$$S_a^{(2P)}(t=0) = \begin{pmatrix} 0 \\ 0 \\ -1 \\ 0 \\ 0 \\ 0 \\ 0 \\ \frac{1}{\sqrt{3}} \\ 1 \end{pmatrix}, \qquad S_a^{(3D)}(t=0) = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ -\frac{2}{\sqrt{3}} \\ 1 \end{pmatrix}. \tag{6.24}$$

6.2 Numerical solution for triplet Ps

We first consider triplet Ps. The system (6.21) is solved with Mathematica analytically (with respect to the time t) for different values of the laser intensities. The numerical values of the annihilation rate (eq.(1.7)), the fluorescent decay rates (eq. (2.54)) and the ionization rates (eq. (1.24) and (1.25)) are substituted in the matrix M. It is not possible to obtain with

Mathematica a real analytical solution as a function of the above parameters, because of the large number of equations in the system. As in eq. (5.31), the time-dependent probability to find a bound Ps state is given by:

$$S_9(t) \;=\; \langle\Psi(t)|\Psi(t)\rangle \;=\; \sum_{i=1}^{9} c_i\, v_9^{(i)}\, e^{-m_i t} \;=\; \sum_{i=1}^{9} B_i\, e^{-m_i t}\,. \qquad (6.25)$$

For example, when the laser intensities are equal to the critical ones and the detunings $\Delta\omega_a = \Delta\omega_b = 0$, the matrix M has the following eigenvalues:

$$m_1 = 75.86\ 10^{-10}\ [\mathrm{Ht}] \qquad m_2 = m_3 = 41.61\ 10^{-10}\ [\mathrm{Ht}]$$

$$m_4 = m_5 = 39.08\ 10^{-10}\ [\mathrm{Ht}] \qquad m_6 = m_7^* = (6.39 + i\,1.11)\ 10^{-10}\ [\mathrm{Ht}]$$

$$m_8 = 4.80\ 10^{-10}\ [\mathrm{Ht}] \qquad m_9 = 1.58\ 10^{-10}\ [\mathrm{Ht}] \qquad (6.26)$$

The probability to find Ps is:

$$\begin{aligned} S_9(t) \;=\; & B_1\, e^{-m_1 t} \;+\; B_6\, e^{-m_6 t} \\ & + B_6^*\, e^{-m_7 t} \;+\; B_9\, e^{-m_9 t}\,, \end{aligned} \qquad (6.27)$$

where the coefficients $B_i = c_i\, v_9^{(i)}$ for different initial conditions are given in Table 6.1. All other coefficients are zero. The probability $S_9(t)$ practically does not depend on the initial Ps state. We found in Chapter 2, section 1, that in the idealized situation when no fluorescence and ionization are

initial state	B_1	B_6	B_9
1S	-1.6 10^{-5}	0.0224 + i0.0451	0.9551
2P	-0.0218	0.0236 + i0.0494	0.9745
3D	0.0020	-0.1005 + i0.3448	1.0150

Table 6.1: Coefficients multiplying the exponential terms in eq. (6.27) for the probability to find triplet Ps. The values of the laser fields are equal to the critical ones.

present in the problem, the atom is in a linear superposition of the dressed states (2.18) and (2.20). In this case the initial conditions determine the values of the coefficients in the expansion on the dressed states. Therefore they are expected to influence the results significantly. But spontaneous radiative transitions are included in the problem considered here. They are fast compared to the lifetime of triplet Ps and the atom fluoresces many times befor annihilation. This alters the wave function of the atom in the laser fields removing the dependence on the initial state.

The longest surviving term in the expression for the probability will be the one for which $Re\ m_i$ is smallest. This leads to the conclusion that the smallest real part of the eigenvalues of the matrix M is the decay rate of Ps.

The numerical values of the Ps decay rate for detunings $\Delta\omega_a = \Delta\omega_b = 0$ are given in Table 6.2. For comparison, the decay rates without ionization being present in the problem are given in the last column of the table. When laser 'a' or 'b' are switched off, we have a two state atom as discussed in Chapter 1, section 2 [18, 20]. Γ decreases gradually from its value Γ_0 to $\Gamma_0/2$ when the intensity of laser 'b' is increased. Since the ionization rates with laser 'b' are insignificant, increasing the intensity does not lead to Ps destruction and the results in the last two columns are identical. Increasing the intensity of laser 'a' above critical values leads to ionization and therefore to large values of the decay rate in the third column compared to the last one. Of course, this holds also when both lasers are turned on. In the latter case, even without ionization and laser intensities above critical, the decay rate does not decrease significantly from its two-state value of $\Gamma_0/2$. Though we have coherent population trapping[6], the rates of the radiative transitions $3D \rightarrow 2P \rightarrow 1S$ are larger than the annihilation rate of triplet Ps and they repopulate the ground state leading to annihilation. This is a confirmation of the result obtained in Chapter 2, section 2. Therefore the lifetime of triplet

[6]See Chapter 1, section 3.

$\frac{E^a}{E^a_{crit}}$	$\frac{E^b}{E^b_{crit}}$	$\Gamma\ 10^{10}\ [Ht]$	$\Gamma'\ 10^{10}\ [Ht]$
0.0	0.0	1.70	1.70
	0.1	1.70	1.70
	1.0	1.70	1.70
	10.0	1.62	1.62
	100.0	0.93	0.93
	1000.0	0.85	0.85
0.1	0.0	1.70	1.70
	0.1	1.70	1.70
	1.0	1.70	1.70
	10.0	1.62	1.62
	100.0	0.93	0.93
	1000.0	0.85	0.85
1.0	0.0	1.58	1.58
	0.1	1.58	1.58
	1.0	1.58	1.58
	10.0	1.56	1.55
	100.0	0.96	0.93
	1000.0	0.89	0.85
10.0	0.0	2.55	0.81
	0.1	2.55	0.81
	1.0	2.55	0.81
	10.0	2.57	0.81
	100.0	3.42	0.83
	1000.0	4.64	0.83
100.0	0.0	147.89	0.92
	0.1	147.89	0.92
	1.0	147.89	0.92
	10.0	147.89	0.92
	100.0	147.92	0.92
	1000.0	150.24	0.92

Table 6.2: Decay rates of triplet Ps in the field of two resonant circularly polarized lasers, when the atom was initially in its ground state. The annihilation rate outside of the lasers is $\Gamma_0 = 1.7\,10^{-10}\ [Ht]$. For comparison, the results without ionization are given in the last column.

Ps interacting with two laser fields can be increased two times with respect to its value outside of the lasers. In other words two lasers are not more effective than one.

The decay rate of Ps was also found off resonance for different values of the detunings $\Delta\omega_a$ and $\Delta\omega_b$. When ionization is present, changing the laser detunings from zero to 10^{-7} [Ht] does not influence the Ps decay rate, which is due to the increased width of the states. The values of the decay rate off resonance when ionization is not considered are given in Table 6.3.

As can be seen from the table, increasing the detuning of laser 'a' from zero to 10^{-6} [Ht] leads to a significant increase of the decay rate for all values of $\Delta\omega_b$. The influence of the detuning of laser 'b' on the decay rate is similar, but weaker than that of $\Delta\omega_a$ and is present mainly for detunings $\Delta\omega_a \geq 10^{-8}$ [Ht]. The stronger influence of $\Delta\omega_a$ on the decay rate can be explained with the fact that laser 'a' populates the state 3D. Increasing the detuning $\Delta\omega_a$ leads to a smaller probability for the atom to be in this state. But 3D fluoresces to 2P via a slow transition, compared to the fast one from 2P to 1S. Therefore the larger the probability that Ps is in the state 3D, the smaller the decay rate. The conclusion from the results in Table 6.3 is that the lifetime of Ps decreases when the lasers are off resonance, which confirms

$\Delta\omega_a\, 10^{10}$ [Ht]	$\Delta\omega_b\, 10^{10}$ [Ht]	$\Gamma\, 10^{10}$ $[Ht]$
0	0	0.81
1		0.81
10		0.81
10^2		1.08
10^3		1.55
10^4		1.56
0	1	0.81
1		0.81
10		0.81
10^2		1.08
10^3		1.55
10^4		1.56
0	10	0.81
1		0.81
10		0.81
10^2		1.08
10^3		1.56
10^4		1.57
0	100	0.81
1		0.81
10		0.82
10^2		1.12
10^3		1.62
10^4		1.63
0	1000	0.81
1		0.81
10		0.82
10^2		1.13
10^3		1.63
10^4		1.63

Table 6.3: Decay rates of triplet Ps in the field of two near-resonant circularly polarized lasers, when ionization is not considered. The values of the laser fields are $E^a = 10\, E^a_{crit}$ and $E^b = 10\, E^b_{crit}$. The annihilation rate outside of the lasers is $\Gamma_0 = 1.7\, 10^{-10}$ $[Ht]$.

the results obtained in the two state problem and the expectations based on physical reasoning.

6.3 Numerical solution for singlet Ps

The method used in the first section of this chapter can be applied also to singlet Ps. The system of differential equations (6.21), its solution given in eq. (6.22) and the initial conditions (6.23) and (6.24) hold for the singlet as well, provided all parameters are substituted with their appropriate values. The probability to find Ps is given again by eq. (6.25). This approach allows to include spontaneous radiative transitions also for singlet Ps, which was not done in Chapter 4. We solve the problem with Mathematica, as it was done in the case of the triplet. The eigenvalues of the matrix M for different values of the laser intensities and $\Delta\omega_a = \Delta\omega_b = 0$ are given in Table 6.4. The parameter q given in the third column was introduced at the end of Chapter 2, section 1 and is equal to:

$$q = \frac{|\Lambda_a|}{|\Lambda_b|} = \left(\frac{E^a}{E^a_{crit}}\right)^2 / \left(\frac{E^b}{E^b_{crit}}\right) . \qquad (6.28)$$

If the intensity of laser 'a' is equal to the critical one and for different values of the field of laser 'b', we obtain that the smallest eigenvalue is considerably less than the annihilation rate. This is an indication that the lifetime of singlet Ps can be extended in these circomstances. When the field of laser 'a' is ten times the critical one, photoionization is significant and this leads to larger eigenvalues. The smallest eigenvalue $m_9 > \Gamma_0$ and there is no possibility to extend the lifetime. In order to understand better the role of ionization and fluorescence, we have given the corresponding eigenvalues when these processes are not considered in the problem. For strong 'a' fields and no ionization present we obtain that $m_9 < \Gamma_0$ similar to the case when the values of the laser fields are reversed. This means that there would be a possibility for stabilization, because the atoms are mainly in the excited states. But ionization leads to Ps destruction and to a decrease in the lifetime. Note that when neither ionization no fluoresence are present there is always a zero eigenvalue. Therefore Ps can be exclusively in the coherently trapped dressed state and live infinitely long. This proves our considerations in Chapter 1, section 3. The smallest eigenvalue is an indication of the lifetime, but in the case of singlet Ps there are other things to be considered before making final conclusions. Namely the annihilation lifetime is smaller

than the characteristic times of the spontaneous radiative transitions. Therefore the latter cannot influence significantly the wave function of the system. In the idealized circomstances when photoionization and spontaneous radiative transitions are not present and at resonance it was found in Chapter 2, section 1 that the system is in a superposition of the dressed states. The constants in this expansion depend on the initial Ps state and on the parameter q. The following possibilities can be realized:

a. The initial state is 1S: The wave function of the system does not contain the coherently trapped dressed state. Therefore Ps cannot be stabilized. Considering the problem with ionization, fluorescence and off resonance will lead to a small contribution of the trapped state into the wave function.

b. The initial state is 2P:

b_1: For $q > 1$ the wave function contains almost exclusively the coherently trapped dressed state. Therefore the lifetime of Ps can be extended depending on fluorescence and ionization.

b_2: For q=1 the probability that the system is in the trapped state is exactly one half.

b_3: For $q < 1$ the wave function containes mainly the two non-trapped dressed states, which will lead to annihilation.

c. The initial state is 3D:

c_1: For $q > 1$ the wave function contains again mainly the two nontrapped dressed states and therefore Ps cannot be stabilized.

c_2: For q=1 the probability that the system is in the trapped state is exactly one half.

c_3: For $q < 1$ the wave function contains almost exclusively the coherently trapped dressed state and the lifetime can be increased.

Therefore we need to find the complete solution of the time-dependent probability S_9 and carefully compare the coefficients B_i in front of the exponential terms. They are found with Mathematica and for the complete physical problem are given in Table 6.5; in Table 6.6 - when photoinization is not present and in Table 6.7 - when both ionization and fluorescence are not present. Of course the last two tables do not describe the real physical situation, but the results obtained in these cases help us to understand better the influence of the different factors on the lifetime of singlet Ps.

$\frac{E^a}{E^a_{crit}}$	$\frac{E^b}{E^b_{crit}}$	q	m_1	m_2	m_3	m_4	m_5	m_6	m_7	m_8	m_9
1	1	1	$1.0112 \pm i2.5306$		1.0008	0.9781	$0.5523 \pm i1.2672$		$0.5503 \pm i1.2657$		**0.0978**
1	10	0.1	$1.0487 \pm i19.0749$		1.0299	0.9923	$0.5358 \pm i9.5375$		$0.5358 \pm i9.5375$		**0.0415**
1	100	0.01	$1.0495 \pm i190.00$		1.0306	0.9926	$0.5355 \pm i95.004$		$0.5355 \pm i190.00$		**0.0404**
1	1000	0.001	$1.0495 \pm i1900.$		1.0306	0.9781	$0.5355 \pm i950.0$		$0.5355 \pm i950.0$		**0.0404**
10	1	100	8.6035	$5.5925 \pm i95.004$		$5.5924 \pm i95.004$		2.5817	$2.5816 \pm i190.00$		2.5815
10	10	10	8.5512	$5.5794 \pm i9.4727$		$5.5794 \pm i95.4728$		$2.6079 \pm i190.94$		2.6078	2.6072
10	100	1	5,9318	$4.9270 \pm i134.34$		$4.9251 \pm i134.34$		$3.9254 \pm i268.67$		3.9164	3.8963

Table 6.4: Eigenvalues of the matrix M for singlet Ps arranged in decreasing value of the real part. All numerical values are given in 10^{-7} [Ht]. The annihilation rate outside of the lasers is 1.9 10^{-7} [Ht]. Complex numbers that span two columns of the table denote a pair of complex conjugated eigenvalues. The top sign refers to the first eigenvalue and the bottom one to the second.

$\frac{E^a}{E^a_{crit}}$	$\frac{E^b}{E^b_{crit}}$	q	m_1	m_2	m_3	m_4	m_5	m_6	m_7	m_8	m_9
			ionization not present								
10	1	100	0.9541	$0.9540 \pm i189.97$		0.9539	$0.5148 \pm i95.003$		$0.5148 \pm i95.003$		**0.0755**
10	10	10	$0.9545 \pm i190.91$		0.9543	0.9540	$0.5147 \pm i95.473$		$0.5147 \pm i95.473$		**0.0747**
10	100	1	$0.9799 \pm i268.68$		0.9709	0.9510	$0.5064 \pm i134.35$		$0.5064 \pm i134.35$		**0.0398**
			ionization and fluorescence not present								
1	1	1	0.9500	0.9500	$0.9500 \pm i2.5133$		$0.4750 \pm i1.2567$		$0.4750 \pm i1.2564$		**0.0000**
1	10	0.1	0.9500	0.9500	$0.9500 \pm i19.071$		$0.4750 \pm i9.5356$		$0.4750 \pm i9.5355$		**0.0000**
10	10	10	0.9500	0.9500	$0.9500 \pm i190.907$		$0.4750 \pm i95.473$		$0.4750 \pm i95.472$		**0.0000**

Table 6.4 continued

$\frac{E^a}{E^a_{crit}}$	$\frac{E^b}{E^b_{crit}}$	q	in. st.	B_1	B_2	B_3	B_4	B_5	B_6	B_7	B_8	B_9
1	1	1	1S	-0.0613 $\mp i$0.1833		0.	1.1217	-0.0004 $\pm i$0.0003		0.	0.	0.0017
			2P	-0.0242 $\pm i$0.0930		0.	0.5478	-0.0032 $\mp i$0.0099		0.	0.	0.5071
			3D	-0.0382 $\pm i$0.0866		0.	0.5302	0.0029 $\pm i$0.0102		0.	0.	0.5405
1	10	0.1	1S	-0.0007 $\mp i$0.0236		0.	1.0019	0.0000	0.0000	0.	0.	$2\ 10^{-6}$
			2P	-0.0010 $\mp i$0.0237		0.	0.9923	0.0000	0.0000	0.	0.	0.0100
			3D	$2\ 10^{-5} \mp i 2\ 10^{-4}$		0.	0.0011	0.0000	0.0000	0.	0.	0.9989
1	100	0.01	1S	$\mp i$ 0.0024		0.	1.0000	0.0000	0.0000	0.	0.	0.0000
			2P	0.0000	0.0000	0.	0.9999	0.0000	0.0000	0.	0.	0.0001
			3D	0.0000	0.0000	0.	-0.0081	0.0000	0.0000	0.	0.	1.0081
1	1000	0.001	1S	$\mp i$ 0.0002		0.	1.0000	0.0000	0.0000	0.	0.	0.0000
			2P	0.0000	0.0000	0.	1.0000	0.0000	0.0000	0.	0.	0.0000
			3D	0.0000	0.0000	0.	-0.0082	0.0000	0.0000	0.	0.	1.0082

Table 6.5: Coefficients multiplying the exponential terms in the time-dependent probability to find singlet Ps. This is the solution of the complete physical problem with photoionization and spontaneous radiation. Numbers that span two columns of the table denote a pair of complex conjugated coefficients. The top sign refers to the first coefficient and the bottom one to the second. The initial state of Ps is given in the fourth column.

$\frac{E^a}{E^a_{crit}}$	$\frac{E^b}{E^b_{crit}}$	q	in. st.	B_1	B_2	B_3	B_4	B_5	B_6	B_7	B_8	B_9
10	1	100	1S	-0.0006	0.0000	0.0000	0.	0.	0.	$\pm i$ 0.0018		1.0006
			2P	0.9872	0.0000	0.0000	0.	0.	0.	0.	0.	0.0128
			3D	-0.0005	0.0000	0.0000	0.	0.	0.	$\pm i$ 0.0015		1.0005
10	10	10	1S	-0.0006	0.0000	0.0000	0.	0.	$\pm i$ 0.0018		0.	1.0006
			2P	0.9774	0.0000	0.0000	0.	0.	$\pm i$ 2 10^{-5}		0.	0.0225
			3D	0.009	0.0000	0.0000	0.	0.	0.	0.	0.	0.9909
10	100	1	1S	-0.0004	-0.0001 $\mp i$0.0049		0.	0.	$\pm i$ 0.0037		0.	1.0007
			2P	0.4902	0.0000	0.0000	0.	0.	$\mp i$ 0.0018		0.	0.5101
			3D	0.1633	-0.0001 $\mp i$0.0085		0.	0.	$\pm i$ 0.0016		0.	0.8371

Table 6.5 continued

$\frac{E^a}{E^a_{crit}}$	$\frac{E^b}{E^b_{crit}}$	q	in. st.	B_1	B_2	B_3	B_4	B_5	B_6	B_7	B_8	B_9
10	1	100	1S	0.9952	0.	0.	0.0000	0.	0.	0.	0.	0.0048
			2P	-0.0857	0.	0.	0.0000	0.	0.	0.	0.	1.0857
	0		3D	0.9953	0.	0.	0.0000	0.	0.	0.	0.	0.0047
10	10	10	1S	0.0000	0.	0.	0.9953	0.	0.	0.	0.	0.0047
			2P	0.0000	0.	0.	-0.0742	0.	0.	0.	0.	1.0742
			3D	0.0000	0.	0.	0.9848	0.	0.	0.	0.	0.0152
10	100	1	1S	0.0000	0.	0.	0.9989	0.	0.	0.	0.	0.0011
			2P	0.0000	0.	0.	0.4776	0.	0.	0.	0.	0.5224
			3D	0.0000	0.	0.	0.4775	0.	0.	0.	0.	0.5225

Table 6.6: Coefficients mutiplying the exponential terms in the time-dependent probability to find singlet Ps. Photoionizatioin is **not** considered.

$\frac{E^a}{E^a_{crit}}$	$\frac{E^b}{E^b_{crit}}$	q	in. st.	B_1	B_2	B_3	B_4	B_5	B_6	B_7	B_8	B_9
1	1	1	1S	1.1429	0.0000	-0.0714 $\mp i$0.1890		0.	0.	0.	0.	0.0000
			2P	0.5714	0.0000	-0.0357 $\pm i$0.0945		0.	0.	0.	0.	0.5000
			3D	0.5713	0.0000	-0.035 $\pm i$0.0945		0.	0.	0.	0.	0.5001
1	10	0.1	1S	0.0000	1.0024	-0.0012 $\pm i$ 0.0249		0.	0.	0.	0.	0.0000
			2P	0.0000	0.9925	-0.0012 $\pm i$0.0247		0.	0.	0.	0.	0.0099
			3D	0.0000	0.0099	0.0000	0.0000	0.	0.	0.	0.	0.9901
10	10	10	1S	0.0000	1.0000	0.0000	0.0000	0.	0.	0.	0.	0.0000
			2P	0.0000	0.0100	0.0000	0.0000	0.	0.	0.	0.	0.9900
			3D	0.0000	0.9903	0.0000	0.0000	0.	0.	0.	0.	0.0097

Table 6.7: Coefficients mutiplying the exponential terms in the time-dependent probability to find singlet Ps. Photoionization and spontaneous radiative transitions are **not** considered.

Let us first consider the expression for the probability to find Ps given in eq. (6.25). We have found the eigenvalues m_i, which are either real or complex conjugared pairs. The coefficients B_i in front of the exponential terms are real when they correspond to real eigenvalues and comlex conjugated pairs otherwise. The terms in the probability corresponding to the latter ones are also real. Let $m_{j,j+1} = x \pm iy$ and $B_{j,j+1} = a \pm ib$. We have:

$$\begin{aligned} B_j\, e^{-m_j t} \;+\; B_{j+1}\, e^{-m_{j+1} t} \;&=\; 2\, Re\,\{B_j\, e^{-m_j t}\} \qquad (6.29) \\ &=\; e^{-xt}\, 2\,(a\, cos\, yt \;+\; b\, sin\, yt) \end{aligned}$$

Therefore the expression for the probability is real and is a sum of exponentially decaying terms, some of which also oscillate. It is normalised to one at t=0. The question is how to define an effective lifetime for a probability of this kind. Careful comparison of the coefficients B_i gives an obvious answer in some cases. If B_9 is close to one and all other coefficients are very small or zero, then we can safely consider m_9 to be the decay rate. If some other coefficient B_i is close to one and all other B_j for $j > i$ are zero, then we can take m_i to be the decay rate. There are a few cases when $B_9 \approx 0.5$ and also some other coefficient $B_i \approx 0.5$. If the difference between the corresponding eigenvalues is very large, the 'i' term will decay fast and the decay rate is

basically determined by m_9. In all other cases there is no straightforward way to define an effective decay rate. Most of these case fall in the following category: the coefficient B_9 is small, but different from zero and some B_j is very close to one for $j < 9$. In an experiment with a Ps beam most of the atoms will decay fast with a rate m_j and a very small number of atoms will live long with a lifetime $\tau = 1/m_9$. We have:

$$P(t) \;=\; S_9(t) \;=\; B_j\, e^{-m_j t} \;+\; B_9\, e^{-m_9 t}\,. \tag{6.30}$$

At time $t = t^*$ these two terms will become equal, where

$$t^* \;=\; -\frac{ln(B_9/B_j)}{m_j - m_9}\,. \tag{6.31}$$

For $t > t^*$ the probability will be essentially determined by the slowly decaying exponential term:

$$P(t) \;=\; B_9\, e^{-m_9 t} \qquad \text{for } \; t > t^*\,. \tag{6.32}$$

The lifetime of singlet Ps in the field of two resonant laser fields, divided by the annihilation lifetime τ_0 of the ground state outside of the lasers, for different values of the laser intensities and initial conditions is given in Table 6.8. Since we have used τ_0 as a unit of time, we give the lifetime of the state

2P:

$$\tau_{2P} = \tau_{10} + \tau_{an}^{s} = 26.6\,\tau_0 \tag{6.33}$$

and the state 3D:

$$\tau_{3D} = \tau_{21} + \tau_{10} + \tau_{an}^{s} = 274.6\,\tau_0 \tag{6.34}$$

outside of the laser fields in the last line of the table. In this way we can compare the results for these states when Ps is interacting with the lasers. When two numbers are given, the first one indicates the dominant term in the probability to find Ps and the second number in brackets refers to the smallest eigenvalue with a small but nonzero coefficient as discussed in the previous paragraph.

The first four lines of the table give the lifetime when the intensity of laser 'a' is critical and for different values of the field of laser 'b' above critical. Ionization is weak and the lifetime is determined mainly by the laser-atom interaction and fluorescence. When the value of the parameter $q < 1$, we have cases 'b_3' and 'c_3' considered above. If the initial Ps state was 3D, we obtain a lifetime of up to $47\,\tau_0 = 0.17\,\tau_{3D}$, which is smaller than the one without the interaction with the lasers. If Ps was initially in the bare state 2P, the dominent term in the probability gives a lifetime of almost twice τ_0, which

$\frac{E^a}{E^a_{crit}}$	$\frac{E^b}{E^b_{crit}}$	q	1S	2P	3D
1	1	1	1.94 (**19.43**)	19.43	19.43
1	10	0.1	1.91 (**45.78**)	1.91 (**45.78**)	45.78
1	100	0.01	1.91	1.91 (**47.03**)	47.03
1	1000	0.001	1.94	1.94	47.03
10	1	100	0.74	0.22 (0.74)	0.74
10	10	10	0.73	0.22 (0.73)	0.73
10	100	1	0.49	0.49	0.49
			ionization not present		
10	1	100	1.99 (25.17)	25.17	1.99
10	10	10	1.99 (25.43)	25.43	1.99 (25.43)
10	100	1	1.95 (47.74)	47.74	47.74
			ionization and fluorescence not present		
1	1	1	2	∞	∞
1	10	0.1	2	2	∞
10	10	10	2	∞	2
			no interaction with the lasers		
0	0		1	26.6	274.6

Table 6.8: Lifetime of singlet Ps in the field of two resonant laser fields, divided by the annihilation lifetime of the ground state outside of the lasers, for different values of the laser intensities and initial conditions.

is also smaller than its lifetime outside of the laser fields. The numbers in brackets for the initial state 2P refer to the lifetime given by m_9 in eq (6.32). For $q = 0.1$ and $t^* > 9.18\,\tau_0$ the lifetime is $45.78\,\tau_0 = 1.72\,\tau_{2P}$. For $q = 0.01$ and $t^* > 18.38\,\tau_0$ the lifetime is $47.03\,\tau_0 = 1.77\,\tau_{2P}$. When $q = 1$, we have cases 'b_2' and 'c_2' considered above and the lifetime for both 2P and 3D initial states is almost $20\,\tau_0$ (less than τ_{2P} and τ_{3D}). For Ps starting in its ground state and $q \leq 1$ we have case 'a' considered above. The probability to find Ps is the sum of a short lived exponential term plus a small contribution from a slowly decreasing one as in eq (6.30). For $q = 1$ and $t^* > 14.01\,\tau_0$ the second term will be dominant and will determine a lifetime of $19.43\,\tau_0$. For $q = 0.1$ and $t^* > 26.23\,\tau_0$, the lifetime is determined by the slow exponential and extended to $45.78\,\tau_0$. These two cases for initial state 1S and the two ones for 2P discussed above are the only possibilities to extend the lifetime of Ps in the lasers beyond the lifetime of the respective initial state without the laser fields.

When $E^a = 10\,E^a_{crit}$ and for all values of the field of laser 'b', ionization is strong and the lifetime is less than the one outside of the laser fields regrdless of the initial Ps state.

The following three lines in Table 6.8 give the lifetime for $E^a = 10\,E^a_{crit}$ and no photoionization present in the problem. As we mentioned before, this is not a realistic physical situation, but it helps us to gain a better understanding of the different factors influencing the lifetime. Here we have $q \geq 1$. If the initial state was 2P (cases 'b_1' and 'b_2'), the lifetime can be increased up to $47\,\tau_0 = 1.9\,\tau_{2P}$. If the initial states was 3D (cases 'c_1' and 'c_2'), the lifetime is always smaller than the one outside of the laser fields. When Ps was initially in its ground state, the lifetime cannot be increased more than twice. But for all values of the field of laser 'b', the coefficient B_9 has a small nonzero value. Therefore there is a small probability for an extended lifetime of up to 47 times.

Finally we have considered the problem when both ionization and fluorescence are not present. Here we can see clearly the role of the initial Ps state on the lifetime and the effect of coherent population trapping.

To summarise the results in this chapter, the lifetime of triplet Ps in the field of two lasers cannot be extended more than two times its value without the interaction with the lasers due to spontaneous radiative transitions. The lifetime of singlet Ps can be extended with a small probability. If the initial state was 1S - up to $45\,\tau_0$ and if the initial state was 2P - almost $2\,\tau_{2P}$.

Chapter 7

Conclusions

We have considered the problem of a positronium atom interacting with laser fields with the goal to extend its lifetime [18, 19]. Ps is an unstable system, since the electron and the positron are a particle-antiparticle pair. It annihilates for all practicle purposes only from S states. Singlet Ps annihilates mainly by emission of two γ quanta and the triplet state - mainly by 3γ emission. The lifetimes are respectively τ_{an}^{s}= 1.25 10^{-10} sec and τ_{an}^{tr}=1.4 10^{-7} sec. Singlet and triplet Ps are not coupled by the lasers and they are discussed separately, but we have found an approach, which can be applied to both of them.

A Ps atom interacting with laser fields is in a dressed state. Its wave

function can be represented as a linear superposition of the bare states and under certain conditions it does not contain the 1S ground state, which can lead to a longer lifetime. This is the phenomenon of coherent population trapping. Most of the existing studies discuss the latter in the context of a three level 'Λ' system driven by two semiclassical lasers. Two-level systems are not known to exibit trapping states, except when the two-level atom is interacting with a quantized field [29] or with a frequency modulated semi-classical field [30]. Therefore coherent population trapping can be used to prolong the lifetime of Ps in the case of a three-level system. We consider Ps atoms in the field of two different circularly polarized lasers. Each of them is nearly resonant with a given transition between the ground state and an excited Ps state. In principle any two excited states which obey the selection rules can be chosen. But spontaneous radiative transitions can occur between them and other Ps states that are not coupled by the lasers. This would lead to a multilevel problem, which is considerably more difficult (and probably gives less enhancement of the lifetime), without introducing any new physics. Therefore it is necessary to select a simple three state system wich is closed with respect to fluorescence. This can be achieved by the following configuration: laser 'a' couples $1S(m_l = 0)$ with $3D(m_l = \pm 2)$ via a two photon

transition and laser 'b' couples $1S(m_l = 0)$ with $2P(m_l = \pm 1)$ via a single photon transition (see Figure 1.2). This configuration of Ps states and laser fields resembles an inverted 'Λ' system (or a 'V' system). It has the property of coherent population trapping at resonance, which means in this case that the laser detunings are equal ($\Delta\omega_a = \Delta\omega_b$). The three dressed Ps states close to resonance are found in Chapter 2, section 1. One of them has no 1S component. This state would not annihilate at all were it not for the fact that spontaneous radiative transitions 3D→2P→1S occur.[1] They repopulate the ground state and permit annihilation. The controlling lifetime will be that of the 2P→1S transition, which is $\tau_{10} = 3.2\ 10^{-9}$sec. The 3D→2P transition is much slower with a lifetime of $\tau_{21} = 3.1\ 10^{-8}$sec.

Photoionization and annihilation are competing processes and they both lead to the destruction of Ps. We find that ionization with laser 'b' is too weak to be observed, but the one with laser 'a' is significant and for values of the field above critical is the dominant process. Nonresonant two-photon ionization from the ground state is also weak. Therefore we have considered the above configuration of a three state Ps atom interacting with two near-

[1]Radiative transitions to other Ps states are forbidden by the selection rules.

resonant lasers, including in the problem spontaneous radiative transitions, annihilation from the ground state and photoionization from the excited states.

In Chapter 3 and 5 we discuss the two state problem with one laser. Photoionization is included, which was not done in previous work. The approach in Chapter 3 is suitable for treating singlet Ps without fluorescence. We have extended the Hilbert space to include the continuum states resulting from annihilation and ionization and use the projection operator formalism. The approach of Chapter 5 is to include annihilation and ionization via an antihermitian (absorptive) term in the Hamiltonian. This allows to consider spontaneous radiative transions as well. The method is adopted in Chapter 6 to solve the complete physical problem for singlet and triplet Ps in the three state case. It can be also used for stable atoms (no annihilation present), when there is photoionization induced by the lasers. The results obtained in both chapters confirm the ones for the two state Ps atom in [18, 19].

In Chapter 2 we find the dressed states of the Ps atom in the field of two lasers. Their analytic form is given in eq. (2.18) and (2.20) in the case when the system is very close to resonance and no fluorescence and ionization are present. The actual state of the atom is a supperposition of the dressed

states with constants determined by the initial conditions. We obtain that the initial Ps state and the value of the parameter

$$q = \frac{|\Lambda_a|}{|\Lambda_b|} = \left(\frac{E^a}{E^a_{crit}}\right)^2 / \left(\frac{E^b}{E^b_{crit}}\right) \tag{7.1}$$

are the major factors, which determine to what extent the Ps wave function in the lasers contains the coherently trapped dressed state. In the idealized circumstances when photoionization and spontaneous radiative transitions are not present and at resonance the following possibilities can be realized:

a. The initial state is 1S: The wave function of the system does not contain the coherently trapped dressed state. Therefore Ps cannot be stabilized. Considering the problem with ionization, fluorescence and off resonance will lead to some contribution of the trapped state into the wave function.

b. The initial state is 2P:

b_1: For $q > 1$ the wave function contains almost exclusively the coherently trapped dressed state. Therefore the lifetime of Ps could be extended depending on fluorescence and ionization.

b_2: For q=1 the probability that the system is in the trapped

state is exactly one half.

b_3: For $q < 1$ the wave function containes mainly the two non-trapped dressed states, which will lead to annihilation.

c. The initial state is 3D:

c_1: For $q > 1$ the wave function contains again mainly the two nontrapped dressed states and therefore Ps cannot be stabilized.

c_2: For q=1 the probability that the system is in the trapped state is exactly one half.

c_3: For $q < 1$ the wave function contains almost exclusively the coherently trapped dressed state and there is a possibility to increase the lifetime.

In Chapter 2, section 2, we solve the problem for triplet Ps without ionization. We find the occupation probabilities of the three bare states. The initial Ps state is not a factor, since spontaneous radiative transitions are much faster than the annihilation rate and the atom fluoresces many times while in the laser fields. This alters the dressed wave function removing the dependence on the initial state. We find that the lifetime cannot be extended more than two times (as in the two state case). The reason is again

fluorescence, which in spite of coherent population trapping repopulates the ground state and leads to annihilation.

In Chapter 6, section 1 we consider the complete physical problem of Ps in the field of two near-resonant lasers. We obtain a system of homogeneous linear first order differential equations for the expectation values of the basis set of matrices (the generators of the SU(3) group) with respect to the total wave function of the system. One of them gives the probability to find Ps as a function of time:

$$P(t) \;=\; S_9(t) \;=\; \sum_{i=1}^{9} B_i \, e^{-m_i t} \,, \tag{7.2}$$

where the eigenvalues m_9 are in general complex and are arranged in decreasing order of their real part. Therefore it is a linear combination of exponentially decaying terms, some of which also oscillate. This probability was found numerically for triplet Ps in section 2 and for singlet Ps in section 3. For triplet Ps we find again that the initial state does not influence the results. The longest surviving turm in the expression for the probability has a coefficient B_9 close to one. Therefore the smallest eigenvalue can be considered as the decay rate. The latter is given for different values of the laser intensities and detunings. We obtain that the lifetime in the laser fields

cannot be extended more than twice. This confirms the results of Chapter 2 and shows that two lasers are not more effective than one. Photoionization proves to be significant for values of the field of laser 'a' above critical.

In the case of singlet Ps we obtain in Chapter 6, section 3 the complete expression for the probability for different values of the laser intensities and different initial Ps states. As expected, the latter influence strongly the lifetime, because spontaneous radiative transitions are slow in comparison to the annihilation rate. We find as well the probability when ionization is not present and also when ionization and fluorescence are not present. These are not a realistic physical situations, but they helps us to gain a better understanding of the different factors influencing the lifetime. The question is how to define an effective lifetime for a probability, which is such a complicated function of time. Careful comparison of the coefficients B_i gives an obvious answer in some cases. If B_9 is close to one and all other coefficients are very small or zero, then we can safely consider m_9 to be the decay rate. If some other coefficient B_j is close to one and all other B_i for $i > j$ are zero, then we can take m_j to be the decay rate. There are a few cases when $B_9 \approx 0.5$ and also some other coefficient $B_j \approx 0.5$. If the difference between the corresponding eigenvalues is very large, the 'j' term

will decay fast and the decay rate is basically determined by m_9. In all other cases there is no straightforward way to define an effective decay rate. They fall in the following category: the coefficient B_9 is small, but different from zero and some B_j is very close to one for $j < 9$. In an experiment with a Ps beam most of the atoms will decay fast with a rate m_j and a very small number of atoms will live long with a lifetime $\tau = 1/m_9$. We have:

$$P(t) \;=\; S_9(t) \;=\; B_j\, e^{-m_j t} \;+\; B_9\, e^{-m_9 t}\,. \tag{7.3}$$

At time $t = t^*$ these two terms will become equal, where

$$t^* \;=\; -\frac{ln(B_9/B_j)}{m_j - m_9}\,. \tag{7.4}$$

For $t > t^*$ the probability will be essentially determined by the slowly decaying exponential term:

$$P(t) \;=\; B_9\, e^{-m_9 t} \qquad \text{for } \; t > t^*\,. \tag{7.5}$$

Ionization is weak when the intensity of laser 'a' is critical and for different values of the field of laser 'b' above critical and the lifetime is determined mainly by the laser-atom interaction and fluorescence. When the value of the parameter $q < 1$, we have cases 'b_3' and 'c_3' considered above. If the initial

Ps state was 3D, we obtain a lifetime of up to $47\,\tau_0 = 0.17\,\tau_{3D}$. Therefore it is reduced by a factor of five with respect to the lifetime

$$\tau_{3D} \;=\; \tau_{21} + \tau_{10} + \tau_{an}^{s} \;=\; 274.6\,\tau_0 \tag{7.6}$$

without the interaction with the lasers. If Ps was initially in the bare state 2P, the dominent term in the probability gives a lifetime of almost twice τ_0, which is also smaller than its lifetime

$$\tau_{2P} \;=\; \tau_{10} + \tau_{an}^{s} \;=\; 26.6\,\tau_0 \tag{7.7}$$

outside of the laser fields. For $q = 0.1$ and $t^* > 9.18\,\tau_0$ the lifetime is determined by the smallest eigenvalue and it is $45.78\,\tau_0 = 1.72\,\tau_{2P}$. For $q = 0.01$ and $t^* > 18.38\,\tau_0$ the lifetime is $47.03\,\tau_0 = 1.77\,\tau_{2P}$. When $q = 1$, we have cases 'b_2' and 'c_2' considered above and the lifetime for both 2P and 3D initial states is almost $20\,\tau_0$ (less than τ_{2P} and τ_{3D}). For Ps starting in its ground state and $q \leq 1$ we have case 'a' considered above. The probability to find Ps is the sum of a short lived exponential term plus a small contribution from a slowly decreasing one as in eq (7.3). For $q = 1$ and $t^* > 14.01\,\tau_0$ the second term will be dominant and will determine a lifetime of $19.43\,\tau_0$. For $q = 0.1$ and $t^* > 26.23\,\tau_0$, the lifetime is determined again by the slow exponential and extended to $45.78\,\tau_0$. These two cases for initial state 1S

and the two ones for 2P discussed above are the only possibilities to extend the lifetime of Ps in the lasers beyond the lifetime of the respective initial state without the laser fields.

To summarise the results, the lifetime of triplet Ps in the field of two lasers cannot be extended more than two times its value without the interaction with the lasers due to spontaneous radiative transitions. The lifetime of singlet Ps can be extended with a small probability. If the initial state was 1S - up to $45\,\tau_0$ and if the initial state was 2P - almost $2\,\tau_{2P}$.

Appendix A

Calculation of radiative and one-photon ionization decay rates

In this and the following appendix the explicit form of the wave functions for 1S, 2P and 3D positronium states [45] are used extensively and it is convenient to write them here. The radial parts are:

$$\begin{aligned} R_{10}(r) &= \frac{2}{a_0^{3/2}} e^{-\frac{r}{a_0}} \\ R_{21}(r) &= \frac{1}{(2a_0)^{3/2}} \frac{r}{\sqrt{3}a_0} e^{-\frac{r}{2a_0}} \end{aligned} \tag{A.1}$$

$$R_{32}(r) = \frac{1}{(3a_0)^{3/2}} \frac{2\sqrt{2}}{27\sqrt{5}} \frac{r^2}{a_0^2} e^{-\frac{r}{3a_0}}$$

while the angular are:

$$\begin{aligned} Y_0^0(\theta,\varphi) &= \frac{1}{2\sqrt{\pi}} \\ Y_1^1(\theta,\varphi) &= -\frac{1}{2}\sqrt{\frac{3}{2\pi}}\, sin\theta\, e^{i\varphi} \\ Y_2^2(\theta,\varphi) &= \frac{1}{4}\sqrt{\frac{15}{2\pi}}\, sin^2\theta\, e^{2i\varphi} \end{aligned} \tag{A.2}$$

We shall also need:

$$Y_3^3(\theta,\varphi) = -\frac{1}{8}\sqrt{\frac{35}{\pi}}\, sin^3\theta\, e^{3i\varphi}\,. \tag{A.3}$$

Note that a_0 is twice the Bohr radius a_B.

There are two spontaneous radiative transitions that are relevant to the problem. Half of the respective natural decay rates are given by the expressions (2.54):

$$\gamma_{10} = \frac{2}{3}\frac{W_{10}^3}{c^3\hbar^4}|\vec{d}_{10}|^2 \quad \text{for } 2P \rightarrow 1S \tag{A.4}$$

$$\gamma_{21} = \frac{2}{3}\frac{W_{21}^3}{c^3\hbar^4}|\vec{d}_{21}|^2 \quad \text{for } 3D \rightarrow 2P \tag{A.5}$$

The calculation of the matrix elements in $\vec{d}_{10}$ and $\vec{d}_{21}$ is straightforward. The radial integrals can be done by a transformation to a dimensionless variable

$z = r/a_0$ and using the relation:

$$\int_0^\infty z^n e^{-az}\, dz \;=\; \frac{n!}{a^{n+1}} \tag{A.6}$$

where $a > 0$ and n is an integer. We obtain the following numerical values:

$$|\vec{d}_{10}|^2 \;=\; |\langle u_1|e\vec{r}|u_0\rangle|^2 \;=\; 0.5549\, e^2 a_0^2 \tag{A.7}$$

$$|\vec{d}_{21}|^2 \;=\; |\langle u_2|e\vec{r}|u_1\rangle|^2 \;=\; 9.017\, e^2 a_0^2\,, \tag{A.8}$$

which give the respective half widths:

$$\gamma_{10} \;=\; 3.793\ 10^{-9}\ [\mathrm{Ht}] \tag{A.9}$$

$$\gamma_{21} \;=\; 3.916\ 10^{-10}\ [\mathrm{Ht}] \tag{A.10}$$

The corresponding lifetimes $\tau_{ij} = 2\hbar/\gamma_{ij}$ of these states are:

$$\tau_{10} \;=\; 3.189\ 10^{-9}\ s \tag{A.11}$$

$$\tau_{21} \;=\; 3.089\ 10^{-8}\ s \tag{A.12}$$

Another relevant quantity is Λ_b, the coupling matrix element for the one photon transition $1S \leftrightarrow 2P$. Its numerical value is necessary to estimate the critical amplitude of the elecrtic field of laser 'b'. It is given in (1.4) by the expression:

$$\Lambda_b \;=\; \frac{eE}{m\omega}\hat{\varepsilon}\cdot\langle u_1|\vec{p}|u_0\rangle \tag{A.13}$$

Using the wave functions (A.1) and (A.2), we calculate the above integral to obtain:

$$\Lambda_b \;=\; 1.492\,\frac{E_b}{E_0}\,[\mathrm{Ht}] \tag{A.14}$$

where E_0 is the atomic electric field defined in (1.8).

In principle there are five possible ionization processes. Laser 'a' can ionize the Ps atom either from the state 2P or from 3D. The same holds for laser 'b' (see Figure 1.2). It is also possible to induce a nonresonant transition from 1S into continuum states by a two photon process first with laser 'a' and then with 'b'. The two photon ionization from the ground state when the order of the lasers is reversed is resonant and is included in our previous considerations. We compute the single-photon ionization rates in perturbation theory up to first order. The integrals can be performed analytically and this is outlined below. The two-photon ionization rate from the ground state will be calculated in Appendix B.

The wave function of the continuum states has to be determined carefully in regard to the boundary conditions and normalization. See [34] (Chapter 14), [35] (Chapter 1.3) and [36]. It is the solution for unbound states of the

Schrodinger equation with a Coulomb potential:

$$\left\{\frac{1}{r^2}\frac{\partial}{\partial r}(r^2\frac{\partial}{\partial r}) - \frac{\hat{L}^2}{r^2} + q^2 - \frac{2me^2}{r}\right\} |\Psi_q^{(\pm)}(\vec{r})\rangle \;=\; 0\,, \tag{A.15}$$

where $\hat{L}$ is the orbital angular momentum operator and q is the momentum after ionization in the center of mass frame. The superscript '$\pm$' of the wave function denotes outgoing or incoming wave boundary conditions. The latter one is needed for calculating ionization rates. The substitutions:

$$|\Psi_q^{(\pm)}(\vec{r})\rangle \;=\; \sum_{l=0}^{\infty}\sum_{m=-l}^{l} Y_l^m(\theta,\varphi)\,|\Psi_{ql}^{(\pm)}(r)\rangle \tag{A.16}$$

and

$$|\Psi_{ql}^{(\pm)}(r)\rangle \;=\; i^l\,\frac{e^{\pm i\delta_l(q)}}{r}\,w_l(qr) \tag{A.17}$$

lead to the following equation for the reduced radial wave function:

$$w_l''(z) - \left\{\frac{l(l+1)}{z^2} + \frac{2\eta}{z} - 1\right\} w_l(z) \;=\; 0\,. \tag{A.18}$$

Here $z = qr$ and $\eta = -(a_0 q)^{-1}$ is the Coulomb parameter. The Coulomb scattering phase shift [34] is denoted by $\delta_l(\eta)$ and it is equal to:

$$e^{2i\delta_l} \;=\; \frac{\Gamma(l+1+i\eta)}{\Gamma(l+1-i\eta)}\,, \tag{A.19}$$

where $\Gamma(x)$ is the gamma funtion. This equation has two linearly independent solutions. They are the regular (at the origin) Coulomb function $F_l(\eta, z)$ and

the irregular Coulomb function $G_l(\eta, z)$. Their assymptotic behaviour (for $z \rightarrow \infty$) is:

$$F_l(\eta, z) \rightarrow sin(z - \eta ln2z - \frac{\pi l}{2} + \delta_l) \tag{A.20}$$

$$G_l(\eta, z) \rightarrow cos(z - \eta ln2z - \frac{\pi l}{2} + \delta_l) \tag{A.21}$$

These two solutions are normalized to a delta-function:

$$\int_0^\infty w_l(q_1 r) w_l(q_2 r)\, dr = \frac{\pi}{2}\, \delta(q_1 - q_2) \tag{A.22}$$

It is more convenient to have the energy normalized solutions of the radial Schrodinger equation when calculating the ionization rate. The energy normalized regular Coulomb function is:

$$F_l^E(\eta, qr) = \sqrt{\frac{2m}{\pi \hbar^2 q}}\, F_l(\eta, qr)\,. \tag{A.23}$$

The regular Coulomb function can be expressed in terms of the confluent hypergeometric series [46]:

$$F_l(\eta, qr) = \tag{A.24}$$
$$2^l\, e^{-\frac{\pi\eta}{2}}\, \frac{|\Gamma(l+1+i\eta)|}{(2l+1)!}\, e^{-iqr}\, (qr)^{l+1}\, {}_1F_1(l+1-i\eta, 2l+2; 2iqr),$$

The latter is defined by:

$${}_1F_1(a, b; z) \equiv M(a, b, z) = \sum_{n=0}^{\infty} \frac{\Gamma(a+n)}{\Gamma(a)} \frac{\Gamma(b)}{\Gamma(b+n)} \frac{z^n}{n!}\,. \tag{A.25}$$

When calculating the ionization rates, we use an integral representation for the confluent hypergeometric function:

$$M(a,b,z) \;=\; \frac{2^{1-b}e^{z/2}}{B(a,b-a)}\int_{-1}^{1} dt\,(1+t)^{b-a-1}\,(1-t)^{a-1}\,e^{-\frac{zt}{2}}\;. \qquad \text{(A.26)}$$

As usual $B(x,y)$ denotes the beta function:

$$B(x,y) \;=\; \frac{\Gamma(x)\Gamma(y)}{\Gamma(x+y)}\;. \qquad \text{(A.27)}$$

Some properties of the gamma function that are used extensivelly in the calculations are:

$$\Gamma(n+1)=n! \qquad\qquad \Gamma(x+1)=x\Gamma(x)$$

$$\Gamma(1+ix)\Gamma(1-ix)=\frac{\pi x}{sh\pi x} \qquad\qquad \Gamma^{*}(z)=\Gamma(z^{*}) \qquad \text{(A.28)}$$

Putting together expressions (A.16, 17, 23, 24 and 26) we obtain the explicit form of the energy normalized, with incoming wave boundary conditions, regular at the origin wave function for unbound states in the presence of a Coulomb potential:

$$|u^{-}_{l,\eta}(qr)\rangle \;=\; \sqrt{\frac{2mq}{\pi\hbar^2}}\,\frac{i^l\,e^{-\frac{\pi\eta}{2}}\,2^{-l-1}\,\Gamma(l+1-i\eta)}{B(l+1-i\eta,l+1+i\eta)(2l+1)!}\,z^l\,I(l,\eta,z)\;, \qquad \text{(A.29)}$$

where

$$I(l,\eta,z) \;=\; \int_{-1}^{1} dt\,e^{-izt}\,(1+t)^{l+i\eta}(1-t)^{l-i\eta}\;. \qquad \text{(A.30)}$$

This wave function will be used to calculate all ionization rates. [1]

Using the energy conservation relation:

$$\frac{\hbar^2 q^2}{2m} = \hbar\omega_{ph} + W_i \,, \tag{A.31}$$

we obtain the numerical values of the the Coulomb parameter η. For the four possible single-photon ionization processes they are:

with laser:	'a'	'b'
from 3D:	$-\sqrt{3}$	$-\frac{6}{\sqrt{23}}$
from 2P:	$-\frac{6}{\sqrt{7}}$	$-\sqrt{2}$

(A.32)

The total one-photon ionization rate (probability for ionization per unit time) is given by the following expression [31] (Chapter 2):

$$w = \frac{\pi}{2\hbar}\, e^2 E^2 \int d\Omega_q \, |\langle u^-_{l,\eta}(\vec{r})|\hat{\varepsilon}_\lambda \cdot \vec{r}|u_i\rangle|^2 \, \rho(E_q) \,, \tag{A.33}$$

where $\rho(E_q)$ is the density of final states. It is equal to one when the final states are energy normalized as in (A.23). $\int d\Omega_q$ denotes integration over the angular variables of the final momentum q. With the given normalization of the initial and final state wave functions, we obtain that the dimension of

[1] Note that in the system of units $\hbar = c = 1$, it has a dimension of energy, which will be used to verify the dimension of the the final result.

the ionization rate (again in units $\hbar = c = 1$) is energy, which is the correct one. The explicit forms of the wave functions can be substituted in (A.33). Since the lasers are circularly polarized, the photon polarization vector is:

$$\hat{\varepsilon} \;=\; \frac{1}{\sqrt{2}}(\hat{x} + i\hat{y}) \qquad \text{and} \qquad \hat{\varepsilon}\cdot\vec{r} = \frac{r}{\sqrt{2}}\, sin\theta\, e^{i\varphi} \tag{A.34}$$

After some simplifications we obtain the following expression for the ionization rate:

$$w \;=\; \frac{\pi E^2 a_0^3\, e^{-\pi\eta}\, 2^{-2l'-1}}{|\eta|\,\Gamma(l'+1+i\eta)\,\Gamma(l'+1-i\eta)} \times \tag{A.35}$$

$$\left| \int_0^\infty dz\, z^{l'+3}\, I(l',\eta,z)^*\, \bar{R}_{nl}(z) \int d\Omega_r\, Y_{l'}^{m'}(\theta,\varphi)^*\, Y_l^m(\theta,\varphi)\, sin\theta\, e^{i\varphi} \right|^2 .$$

The primed quantum numbers refer to the final continuum state. Due to the circular polarization of the laser, we have:

$$l' = l+1 \qquad \text{and} \qquad m' = m+1 \tag{A.36}$$

The bar on the bound state radial wave function denotes that the dimensional factor $a_0^{-3/2}$ has been taken away.

We calculate first the ionization rate from the state 3D. Therefore $l = 2$, $m = 2$ and $l' = 3$, $m' = 3$. The angular integral in (A.35) is equal to $\sqrt{\frac{6}{7}}$. The radial integral can be reduced to the following expression:

$$I_r \;=\; 28!\,3^6\sqrt{\frac{2}{5}} \int_{-1}^{1} dt\, \frac{(1+t)^{3-i\eta}\,(1-t)^{3+i\eta}}{(1-3it)^9} \,. \tag{A.37}$$

It can be solved by changing the integration variable via the transformation:

$$t = \frac{s-1}{s+1} . \tag{A.38}$$

This leads to the following expression:

$$I_r = \sqrt{\frac{2}{5}} \frac{2^8 3^6 8!}{(1+3i)^9} \int_0^\infty ds \, \frac{(s+1)\, s^{3-i\eta}}{(1+\beta s)^9} , \qquad \text{where } \beta = \frac{1-3i}{1+3i} . \tag{A.39}$$

This type of integral can be solved analytically [47]:

$$\int_0^\infty ds \, \frac{s^{\mu-1}}{(1+\beta s)^\nu} = \beta^{-\mu} \, B(\mu, \nu-\mu) \tag{A.40}$$

for $\nu, \mu > 0$ and $|arg(\beta) < \pi|$. We obtain that the radial integral in (A.35) is equal to:

$$I_r = \sqrt{\frac{2}{5}} \frac{2^8 \, 3^6}{(1+3i)^9} \, \beta^{-4+i\eta} \, M_\eta , \tag{A.41}$$

where

$$M_\eta = \frac{1}{\beta} \Gamma(5-i\eta)\, \Gamma(4+i\eta) + \Gamma(4-i\eta)\, \Gamma(5+i\eta) . \tag{A.42}$$

The total ionization rate from the state 3D is obtained from (A.35) and (A.41):

$$w_{3D}^\eta = 0.2931 \, E^2 a_0^3 \, \frac{e^{-\pi\eta} \, e^{2\eta \, arg(\beta^{-1})}}{|\eta| \, \Gamma(4+i\eta)\, \Gamma(4-i\eta)} \, |M_\eta|^2 . \tag{A.43}$$

We simlify this expression by taking the absolute value squared of (A.42) and using the properies of the gamma function:

$$w_{3D}^{\eta} = \tag{A.44}$$
$$0.1172\pi\ E^2 a_0^3\ e^{2\eta\, arg(\beta^{-1})}\ \left(\frac{-e^{-\pi\eta}}{sh\pi\eta}\right)\ (9+\eta^2)(4+\eta^2)(1+\eta^2)(4+3\eta)^2\ ,$$

where

$$arg(\beta^{-1}) = 2.498\ . \tag{A.45}$$

Then we substitute the numerical values of the Coulomb parameter η from (A.32). We obtain that the ionization rate from the state 3D with laser 'a' is equal to:

$$w_{3D}^{a} = 0.06172\ (E^a)^2\ a_0^3\ . \tag{A.46}$$

The one with laser 'b' is:

$$w_{3D}^{b} = 0.01303\ (E^b)^2\ a_0^3\ . \tag{A.47}$$

The ionization rate from the state 2P is calculated in the same way from (A.35). We have $l = 1$, $m = 1$ and $l' = 2$, $m' = 2$. The numerical value of the ionization rate with laser 'a' is:

$$w_{2P}^{a} = 0.1617\ (E^a)^2\ a_0^3\ . \tag{A.48}$$

The one with laser 'b' is:

$$w_{2P}^{b} \;=\; 0.02847\,(E^{b})^{2}\,a_{0}^{3}\,. \tag{A.49}$$

It is convenient to write these rates in terms of the critical fields. We first consider singlet Ps:

$$E_{crit}^{a} = E_0\,8.12\,10^{-5} \qquad \text{and} \qquad E_{crit}^{b} = E_0\,1.27\,10^{-7}\,. \tag{A.50}$$

The Bohr radius is $a_B = 7.3\,10^{-3}\;[\mathrm{Ht}]^{-1}$, and we obtain:

$$\begin{aligned}
(E^{a})^{2}\,a_{0}^{3} &= \left(\frac{E^{a}}{E_{crit}^{a}}\right)^{2} 5.274\,10^{-8}\;[\mathrm{Ht}]\,,\\
(E^{b})^{2}\,a_{0}^{3} &= \left(\frac{E^{b}}{E_{crit}^{b}}\right)^{2} 1.290\,10^{-13}\;[\mathrm{Ht}]\,.
\end{aligned} \tag{A.51}$$

Therefore the respective ionization rates are:

$$\begin{aligned}
w_{3D}^{a} &= \left(\frac{E^{a}}{E_{crit}^{a}}\right)^{2} 3.255\,10^{-9}\;[\mathrm{Ht}]\,,\\
w_{2P}^{a} &= \left(\frac{E^{a}}{E_{crit}^{a}}\right)^{2} 8.528\,10^{-9}\;[\mathrm{Ht}]\,,\\
w_{3D}^{b} &= \left(\frac{E^{b}}{E_{crit}^{b}}\right)^{2} 1.681\,10^{-15}\;[\mathrm{Ht}]\,,\\
w_{2P}^{b} &= \left(\frac{E^{b}}{E_{crit}^{b}}\right)^{2} 3.673\,10^{-15}\;[\mathrm{Ht}]\,,
\end{aligned} \tag{A.52}$$

For triplet Ps we obtain in a similar way:

$$(E^{a})^{2}\,a_{0}^{3} \;=\; \left(\frac{E^{a}}{E_{crit}^{a}}\right)^{2} 4.723\,10^{-11}\;[\mathrm{Ht}]\,,$$

$$(E^b)^2\, a_0^3 \;=\; \left(\frac{E^b}{E^b_{crit}}\right)^2 1.040\,10^{-19}\ [\mathrm{Ht}]\,. \tag{A.53}$$

The respective ionization rates are:

$$\begin{aligned} w^a_{3D} &= \left(\frac{E^a}{E^a_{crit}}\right)^2 2.915\,10^{-12}\ [\mathrm{Ht}]\,, \\ w^a_{2P} &= \left(\frac{E^a}{E^a_{crit}}\right)^2 7.637\,10^{-12}\ [\mathrm{Ht}]\,, \\ w^b_{3D} &= \left(\frac{E^b}{E^b_{crit}}\right)^2 1.355\,10^{-21}\ [\mathrm{Ht}]\,, \\ w^b_{2P} &= \left(\frac{E^b}{E^b_{crit}}\right)^2 2.961\,10^{-21}\ [\mathrm{Ht}]\,, \end{aligned} \tag{A.54}$$

We have represented the ionization rates in terms of the ratio of the laser field and the respective critical field. This is why the singlet and triplet ionization rates appear to be different. They are not as can be seen from eq. (A.46-49), but the respective values of the critical fields are. This way of writing allows us to compare easily the above rates with the ones for annihilation in eq. (1.7) and estimate which of the photoionization processes are relevant.

Appendix B

Calculation of two-photon processes

In this appendix we calculate the matrix element of the two-photon laser induced transition 1S↔3D and the ionization rate from the ground state induced first with laser 'a' and then with laser 'b'. These are more difficult to handle since they occur via all posible intermediate Ps states.

The matrix element of the first transition is:

$$\Lambda_a = e^2 E_a^2 \langle u_2 | \hat{\epsilon}^* \cdot \vec{r} | \phi(\vec{r}) \rangle , \qquad \text{(B.1)}$$

where

$$|\phi(\vec{r})\rangle \;=\; \sum_n \frac{|u_n\rangle\langle u_n|\hat{\varepsilon}^*\cdot\vec{r}|u_0\rangle}{W_n-\omega_a-W_0}\,. \tag{B.2}$$

The sum runs over all possible intermediate states $|u_n\rangle$ except 2P. We can perform this sum without approximations by using the method of inhomogeneous differential equations which is described in [31] (Chapter 4), [32, 33] and references therein. The state $|\phi(\vec{r})\rangle$ satisfies the following equation:

$$(H_0-\omega_a-W_0)\;|\phi(\vec{r})\rangle \;=\; \hat{\varepsilon}^*\cdot\vec{r}|u_0\rangle \tag{B.3}$$

As before H_0 denotes the Hamiltonian of the Ps atom outside of the laser field. Eq. (B.3) can be reduced to a one dimensional equation by the substitution:

$$|\phi(\vec{r})\rangle \;=\; \frac{4}{\sqrt{3}}\,m a_0^{3/2}\,Y_1^1(\theta,\varphi)\,f(x)\,, \tag{B.4}$$

where $Y_1^1(\theta,\varphi)$ is the respective spherical harmonic function given in eq. (A.2). We have denoted with a_0 the Bohr radius multiplied by two and introduced a dimensionless variable $x=\frac{r}{a_0}$. The unknown function $f(x)$ satisfies an inhomogeneous differential equation:

$$\left(\frac{d^2}{dx^2}+\frac{2}{x}\frac{d}{dx}-\frac{2}{x^2}+\frac{2}{x}-\frac{5}{9}\right)\;f(x) \;=\; xe^{-x}, \tag{B.5}$$

which can be solved with the Green's function method. After some transfor-

mations we obtain the corresponding homogeneous differential equation:

$$F''(z) + (4-z)\,F'(z) - 0.6584\,F(z) = 0\,, \qquad z = \frac{2\sqrt{5}}{3}\,x\,. \tag{B.6}$$

This is the equation of the confluent hypergeometric function [20]. It has two solutions. One of them M(0.6584, 4, z) is regular at the origin and the other one - U(0.6584, 4, z) is irregular. We can now represent f(x) as an integral of known functions:

$$\begin{aligned} f(x) \;=\; & -0.7559 \int_0^\infty dy\; y^3 e^{-y}\, M(0.6584,\ 4,\ \frac{2\sqrt{5}}{3} y_s)\, y_s\, e^{-\sqrt{\frac{5}{9}} y_s} \times \\ & \times U(0.6584,\ 4,\ \frac{2\sqrt{5}}{3} y_l)\, y_l\, e^{-\sqrt{\frac{5}{9}} y_l}\,. \end{aligned} \tag{B.7}$$

Here y_s denotes the smaller of the two variables x and y, while y_l - the larger one. From eq. (B.4), (B.6) and (B.7) we can solve the integrals in the matrix element of the two photon transition numerically with Mathematica to obtain:

$$\Lambda_a \;=\; 28.81 \left(\frac{E_a}{E_0}\right)^2. \tag{B.8}$$

From this result and the definition of the critical electric field in eq. (1.14), we obtain:

$$E^a_{crit} \;=\; E_0\; 2.43\, 10^{-6} \quad \text{for triplet Ps} \tag{B.9}$$

$$E^a_{crit} \;=\; E_0\; 8.12\, 10^{-5} \quad \text{for singlet Ps} \tag{B.10}$$

They are considerably larger than the ones for laser 'b' in eq. (1.16), because Λ_a is quadratic in the laser electric field.

The last possible ionization is the nonresonant two photon one from the ground state [37, 38]. Up to first order in perturbation theory the matrix element for this transition is:

$$\Lambda_q \;=\; e^2 E_a E_b \,\langle \Psi^{(-)}_{q,l=2} | \hat{\varepsilon}^* \cdot \vec{r} | \phi\prime(\vec{r}) \rangle \;, \tag{B.11}$$

where

$$|\phi'(\vec{r})\rangle = \sum_n \frac{|u_n\rangle\langle u_n|\hat{\varepsilon}^* \cdot \vec{r}|u_0\rangle}{W_n - \omega_a - W_0} \;. \tag{B.12}$$

The wave function $|\Psi^{(-)}_{q,l=2}\rangle$ is defined in eq. (A.16), (A.17), (A.29) and (A.30). This is the energy normalized with incoming wave boundary conditions, regular at the origin wave function for unbound states in the presence of a Coulomb potential. The sum in (B.12) runs over all possible intermediate states $|u_n\rangle$ except 2P. The matrix element in eq. (B.11) can be solved without any approximations with the inhomogeneous differential equation method [31] in a similar way as it was done above for Λ_a. Using Mathematica, we obtain the following ionization rates:

$$w^{ab}_{1S} \;=\; \left(\frac{E^a}{E^a_{crit}}\right)^2 \left(\frac{E^b}{E^b_{crit}}\right)^2 7.257\,10^{-33}\,[\mathrm{Ht}] \quad \text{for triplet Ps}$$

$$w_{1S}^{ab} = \left(\frac{E^a}{E^a_{crit}}\right)^2 \left(\frac{E^b}{E^b_{crit}}\right)^2 1.010\,10^{-23}\,[\mathrm{Ht}] \quad \text{for singlet Ps.} \quad (B.13)$$

Appendix C

Derivation of the constraint

In this appendix we give a derivation of the constraint, used in Chapters 2, 5 and 6 to handle spontaneous radiative transitions in an atom interacting with laser fields. All notations used here are the same as the ones in the respective chapters.

We consider a system with an hermitian Hamiltonian and derive the constraint for the case in Chapter 2. As described there, physical observables can be expressed as expectation values of operators with respect to the total wave function of the system $|\Psi(t)\rangle$. We can obtain equations for these

observables from:

$$i\frac{d}{dt}\langle\Psi|\mathcal{O}|\Psi\rangle = \langle\Psi|[\mathcal{O},H]|\Psi\rangle. \tag{C.1}$$

If the operator $\mathcal{O}$ for a given observable is independent of the radiation operators and also does not depend explicitely on time, we obtain from (C.1) (eq. (2.46)):

$$\begin{aligned} i\frac{d}{dt}\langle\Psi|\mathcal{O}|\Psi\rangle = \qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad (C.2)\\ \sum_{a=1}^{9} V_a \,\langle\Psi|\,[\mathcal{O},\lambda_a]\,|\Psi\rangle + \Big\{\sum_{a=1}^{9} X_a \,\langle\Psi|\vec{E}_R^{10(+)}(t)\cdot\vec{d}_{01}\;\,[\mathcal{O},\lambda_a]\,|\Psi\rangle e^{-i\omega_b t}\\ + \sum_{a=1}^{9} Z_a \,\langle\Psi|\vec{E}_R^{21(+)}(t)\cdot\vec{d}_{12}\;\,[\mathcal{O},\lambda_a]\,|\Psi\rangle\; e^{-i(2\omega_a-\omega_b)t} \;\; + \;\; h.c.\Big\}. \end{aligned}$$

It is apparent from the above expression that the equation for $\mathcal{O}$ couples to expectation values which contain radiation operators linearly and equations for those (expectation values of E_R) will couple to still others. This results in an infinite set of coupled equations which describe the infinite number of degrees of freedom of the radiation field. Mollow's contribution [21] was the use of an approximate solution for the electric field as an approximate constraint on the wave function, which allows the truncation of the infinite set of equations. The physical reasoning is as follows: When a photon annihilation operator acts on a multiphoton wave function, the result is rather compli-

cated, because any one of the photons may be the one annihilated. But the annihilation must take place at the atom. Therefore the most likely photon to be absorbed will be the one that was last emitted, since the preceding ones have moved away at the velocity of light and are no longer near the atom.

The Hamiltonian in Chapter 2 is:

$$H = H' + H_R\,, \tag{C.3}$$

where H_R gives the interaction of the system with the quantized electromagnetic field resulting from spontaneous radiative transitions and H' contains all the other terms. We work in the interaction representation with respect to H':

$$|\Psi(t)\rangle = e^{-iH't}\,|\Psi_I(t)\rangle \tag{C.4}$$

and

$$H_I(t) = e^{iH't}\,H_R\,e^{-iH't}\,, \tag{C.5}$$

with the respective Schrodinger equation:

$$i\frac{\partial}{\partial t}|\Psi_I(t)\rangle = H_I(t)\,|\Psi_I(t)\rangle\,. \tag{C.6}$$

Also all operators are in the matrix representation of the dressed states (2.6, 10, 14) as in Chapter 2, section 2.

The time evolution operator $U(t_1, t_2)$ [48] will be used extensively in the derivation of the constraint and we would like to recall some of its properies. It is a unitary operator $U^\dagger = U^{-1}$, which describes the time evolution of the system and satisfies the following differential equation:

$$i\frac{\partial}{\partial t_1}U(t_1, t_2) \;=\; H_I(t_1)\,U(t_1, t_2)\,, \tag{C.7}$$

with the initial condition $U(t_1, t_1) = 1$. It can be represented as follows:

$$U(t_1, t_2) \;=\; T\,e^{-i\int_{t_2}^{t_1} H_I(t)\,dt}\,. \tag{C.8}$$

The symbol 'T' denotes the time ordered product.[1] The scattering matrix is equal to the evolution operator for infinite boundaries: $S = U(+\infty, -\infty)$. The wave function of the system at a given time can be obtained from the wave function at a previous time by:

$$|\Psi(t_2)\rangle \;=\; U(t_2, t_1)\,|\Psi(t_1)\rangle\,. \tag{C.9}$$

In an infinite dimensional Hilbert space $U(t_1, t_2)$ is mathematically well defined if H' and H_R are self-adjoint.

The negative frequency component of the electric field operator of the radiation photons acts on the wave function of the system resulting in a

[1] See for example [49] or [50].

state, which we denote with:

$$|\vec{\chi}(t,t_1)\rangle \;=\; \vec{E}_R^{(-)}(t_1)\,|\Psi_I(t)\rangle \tag{C.10}$$

Our goal is to find an expression for $|\vec{\chi}(t,t_1)\rangle$, which is proportional to the wave function of the system with a proportionality factor containing only quantities characteristic for the atom, but not involving the radiation field. The respective differential equation for this state is obtained using (C.6):

$$i\frac{\partial}{\partial t}|\vec{\chi}(t,t_1)\rangle \;=\; H_I(t)\,|\vec{\chi}(t,t_1)\rangle \;+\; [\vec{E}_R^{(-)}(t_1),H_I(t)]\,|\Psi_I(t)\rangle\;. \tag{C.11}$$

It is convenient to use the following notation for the interaction Hamiltonian:

$$H_I(t) \;=\; \vec{E}_R^{(+)}(t)\cdot\vec{h}(t) \;+\; \vec{E}_R^{(-)}(t)\cdot\vec{h}^{\dagger}(t)\;. \tag{C.12}$$

The commutator in (C.11) is found using the commutation relations (2.35) of the creation and annihilation operators for radiation photons:

$$\begin{aligned}\left[E_{Rj}^{(-)}(t_1),H_I(t)\right] \;&=\; \left[E_{Rj}^{(-)}(t_1),E_{Rl}^{(+)}(t)\right]\,h_l(t)\\ &=\; \frac{2\pi}{V}\sum_{k\lambda}\omega_k\,(\varepsilon_{\hat{k}\lambda})_j(\varepsilon_{\hat{k}\lambda}^{*})_l\;e^{i\omega_k(t-t_1)}\,h_l(t)\;.\end{aligned} \tag{C.13}$$

The sum over the photon polarization λ can be done using the following relation:

$$\sum_{\lambda}(\varepsilon_{\hat{k}\lambda})_j(\varepsilon_{\hat{k}\lambda}^{*})_l \;=\; \delta_{jl}-\frac{k_jk_l}{k^2}\;. \tag{C.14}$$

Then the sum over the photon wave vector is transformed into a three dimensional integral. The latter is subsequently reduced to a one dimensional one by integrating over the angles to obtain:

$$[E_{Rj}^{(-)}(t_1), H_I(t)] \;=\; \frac{2}{3\pi}\, h_j(t) \int_0^\infty dk\, k^2\, \omega_k\, e^{i\omega_k(t-t_1)}\,. \qquad \text{(C.15)}$$

We assume that the state $|\vec{\chi}(t,t_1)\rangle$ can be expressed using the time evolution operator as:

$$|\vec{\chi}(t,t_1)\rangle \;=\; -i\int_{-\infty}^{t} dt_2\, U(t,t_2)\, F(t_1-t_2)\, \vec{h}(t_2)\, U^{-1}(t,t_2)\, |\Psi_I(t)\rangle\,. \qquad \text{(C.16)}$$

In order to prove this and to find the unknown function F, we obtain the time derivative of eq. (C.17) using the properties of the evolution operator:

$$i\frac{\partial}{\partial t}|\vec{\chi}(t,t_1)\rangle \;=\; H_I(t)\,|\vec{\chi}(t,t_1)\rangle \;+\; F(t_1-t)\,\vec{h}(t)\,|\Psi_I(t)\rangle \qquad \text{(C.17)}$$

Comparing the above expression with eq. (C.11) and using the results in eq. (C.16), we find the unknown function $F(\tau)$:

$$F(\tau) \;=\; \frac{2}{3\pi c^3}\int_0^\infty d\nu\, \nu^3 C(\nu)\, e^{-i\nu\tau}\,. \qquad \text{(C.18)}$$

The function $C(\nu)$ has been inserted here to control the ultraviolet divergencies of QED. Its properties $C(\infty)=0$ and $C(\nu)\approx 1$ for small ν, ensure that

all integrals converge. This procedure can be formally justified only in covariant renormalization theory. Our calculations will be limited to quantities that are independent of the specific form of $C(\nu)$.

Now we can set $t = t_1$ in eq. (C.17). Since $F(\tau)$ is a sharply peaked function around $\tau = 0$, we can expand $U(t, t_2)$ and $\vec{h}(t_2)$ around $t_2 = t$ and keep only lowest order terms. Using again the properties of the evolution operator, we obtain:

$$|\vec{\chi}(t,t)\rangle \;=\; -i \int_0^\infty ds\, F(s)\, \vec{h}(t-s)\, |\Psi_I(t)\rangle \;, \tag{C.19}$$

where $s = t - t_2$. In order to simlify further, we have to substitute in the above integral the explicit form of $\vec{h}(t)$, which is in the interaction representation (C.5). For the case considered in Chapter 2 it is:

$$\begin{aligned} \vec{h}(t) \;=& \left(e^{iH't} \sum_{a=1}^{9} X_a \lambda_a\, e^{-iH't} \right) \vec{d}_{01}\, e^{-i\omega_b t} \\ &+ \left(e^{iH't} \sum_{a=1}^{9} Z_a \lambda_a\, e^{-iH't} \right) \vec{d}_{12}\, e^{-i(2\omega_a - \omega_b)t} \;. \end{aligned} \tag{C.20}$$

In this expression for $\vec{h}(t-s)$ all functions are expanded around $s = 0$ and only lowest order terms are kept, except for the exponents $e^{-i\omega_b t}$ and $e^{-i(2\omega_a - \omega_b)t}$. Their variation is rapid and they are left as they are. We recall that the first term in eq. (C.20) gives the interaction with radiation photons

resulting from the transition $2P \rightarrow 1S$, while the second term - from the transition $3D \rightarrow 2P$. It is convenient to obtain the constraint for each of these cases separately. This is possible, because the total radiation field is the sum of each of them.[2] Using the identity [50]:

$$\delta_{\pm}(\nu) \;=\; \frac{1}{2\pi}\int_0^{\infty} e^{\pm i\nu s-\eta s}\,ds \;=\; \frac{1}{2}\,\delta(\nu) \;\pm\; \frac{i}{2\pi}\,\mathcal{P}\frac{1}{\nu}\,, \tag{C.21}$$

where $\mathcal{P}$ denotes the principal value and $\eta \rightarrow 0^{+}$, leads to the following result:

$$\vec{E}_R^{10(-)}(t)\,|\Psi_I(t)\rangle \;= \tag{C.22}$$
$$-\frac{2i}{3}\,\omega_b^3\,\vec{d}_{01}\,e^{-i\omega_b t}\left\{1 \;-\; \frac{i}{\omega_b^3\pi}\int_0^{\infty} d\nu\,\nu^3 C(\nu)^2\,\mathcal{P}\frac{1}{\omega_b-\nu}\right\}|\Psi_I(t)\rangle\,.$$

The first term gives the width of the dressed state due to fluorescent emission. The second term gives the energy shift of this state due to coupling to the radiation field. The latter is related to the Lamb shift. It is not possible to calculate it with this technique, but it is known to be small and it is dropped. Going back to the original representation by the inverse transformation of (C.4) and (C.5) and replacing ω_b by W_{10}, we obtain the desired constraint

[2] See eq. (2.42).

for the $2P \rightarrow 1S$ transition:

$$\vec{E}_R^{10(-)}(t)\,|\Psi(t)\rangle \;=\; -\frac{2iW_{10}^3}{3c^3\hbar^3}\,\vec{d}_{01}\,e^{-i\omega_b t}\,(\sum_{a=1}^{9} X_a\lambda_a)\,|\Psi(t)\rangle\,. \qquad \text{(C.23)}$$

We have also inserted in the above expression the missing constants c and The constraint for the $3D \rightarrow 2P$ transition is derived in the same way from (C.20) and (C.21). It is:

$$\vec{E}_R^{21(-)}(t)\,|\Psi(t)\rangle \;=\; -\frac{2iW_{21}^3}{3c^3\hbar^3}\,\vec{d}_{12}\,e^{-i(2\omega_a-\omega_b)t}\,(\sum_{a=1}^{9} Z_a\lambda_a)\,|\Psi(t)\rangle\,. \qquad \text{(C.24)}$$

The constraint is an expansion in the coefficient $\hbar\gamma/W_{10}$, so that subsequent results are accurate only to lowest order in this parameter.

This form of the constraint is used in Chapter 2, section 2 for the three state Ps atom. In Chapter 5 and 6 the problem is considered in such a way as to allow for annihilation from the ground state and ionization by the laser(s) from the excited states. This leads to a nonhermitian Hamiltonian, because of a complex potential included in H'. This term reflects the loss of Ps due to the transitions into continuum states (a free electron-positron pair or high energy gamma quanta). The constraint can be derived for this case as well following the steps described above without modifications. The only difference is that the evolution operator is not unitary any more. But it is well defined and its other properties are preserved since we are working in a finite

dimensional space. The constraint remains unchanged and the respective expression for $\vec{h}(t)$ has to be substituted in (C.19) in order to obtain its final form. The result for a two-state atom is given by the expression in eq. (5.16), while for the three state problem considered in the representation of the bare states - by eq. (6.19) and (6.20).

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