Interaction of Atoms with Frequency-Chirped Pulses and Surface Plasmons

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## Abstract

vii

## Kurzfassung

ix

## 1 Introduction and Motivation

1

1.1 Population transfer and coherent state preparation with single, frequency-chirped laser pulses .............................................. 1

1.2 Collective behaviour of an atomic ensemble due to an engineered reservoir ............................................................ 5

1.3 Outline of the thesis ....................................................... 7

## I Population transfer and coherent state preparation with chirped laser pulses

9

## II Quantum emitters coupled to surface plasmons of a nano-wire: a Green’s

v

### I Population transfer and coherent state preparation with chirped laser pulses

2 Manipulation of atomic population by frequency-chirped laser pulses

11

2.1 Population transfer in a Λ-atom by a single frequency-chirped pulse: a dressed state analysis ....................................................... 12

2.2 Propagation of frequency-chirped laser pulses among Λ-atoms

17

2.2.1 Analytic calculations in the adiabatic regime ................................. 18

2.2.2 Numerical simulation of the full problem .................................... 20

2.3 Creating a superposition of quantum states in tripod atoms

32

2.3.1 Theoretical framework ................................................... 32

2.3.2 Analysis of the atomic population dynamics ............................ 36

2.3.3 Applications of the processes ............................................ 41

## II Quantum emitters coupled to surface plasmons of a nano-wire: a Green’s
function approach 45

3 Collective behaviour of quantum emitters coupled to an engineered reservoir 47
3.1 Field quantization in presence of macroscopic objects and coupling to point-like quantum emitters 47
3.2 Dyadic Green’s functions 50
3.3 Effective couplings and level shifts: master equation for a many-body system coupled to the reservoir 51
3.3.1 Finding the diagonal basis for the master equation 53
3.4 Surface plasmon modes of a lossless nanowire 57
3.5 Interaction of a single emitter with a nano-wire 60
3.5.1 Width of the plasmon resonances 61
3.5.2 Spontaneous emission and level shift of an atom near a nanowire 63
3.5.3 Sub-wavelength nanowire 64
3.6 Plasmon-mediated interaction of two emitters: Dicke superradiance 69
3.6.1 Surface plasmon propagation length and decay length of superradiance 73
3.6.2 Optimizing the atom-atom coupling 77
3.7 Application for a phase gate 79
3.8 Collective behaviour of three emitters coupled by plasmons 84
3.8.1 Three emitters coupled by a single wire 85
3.8.2 Triangle configuration 90
3.9 Chain of atoms; realization of a spin-boson model 94
3.10 Dipole-dipole shifts 96
3.10.1 General method 96
3.10.2 A pair of emitters near a nano-wire 102

III Appendix 105

4 Appendix 107
4.1 Field quantization in the presence of absorbing, magnetic media 107
4.2 Green’s tensor 111
4.2.1 Some properties of the Green’s tensor 111
4.2.2 Integral representation of the Green’s tensor 112
4.3 Kramers-Kronig relations in general 118
4.4 Analytic approximation of the decay and dipole-dipole shift for two emitters coupled to surface plasmons of a nanowire 119
Abstract

This thesis contains the results of our investigations in the field of interacting light-matter systems. During the PhD work two, quite different regimes of interaction between atoms and light were studied. On the one hand, we looked at systems where an ensemble of atoms interact with external, classical laser fields. Here, the main object of interest was how one could prepare the atoms of extended, optically thick media in certain quantum states using these external fields. On the other hand, we investigated the behaviour of systems where quantum emitters are strongly coupled to quantized plasmon modes of metallic nano-structures. In this case, we were interested in the collective behaviour of the atoms due to the photon exchange among themselves, in particular the emergence of modified collective spontaneous decay rates and energy shifts.

Traditional schemes for coherent population transfer or generation of coherent superposition states in multilevel atoms or molecules usually utilize two or more constant-frequency laser beams with radiation bandwidth smaller than the frequency interval between the working levels. We, on the other hand, consider frequency-chirped pulses that induce adiabatic population transfer between the atomic levels in extended, optically thick media: since only a single pulse is applied, the problem of pulse synchronisation is eliminated and also, because of the sweep of the frequency chirp, the method is resistant to inhomogeneous broadening in the medium. To ascertain whether the frequency-chirped pulse maintains its characteristics as it propagates in an extended medium, we study the propagation of such pulses in optically thick media consisting of atoms with a Λ level scheme and also, for comparison, a medium of two-level atoms. The pulses induce transitions between the two lower (metastable) levels of the Λ atoms and between the ground and excited states of the two-level atoms. We show that associated with the adiabatic population transfer in Λ atoms there is a regime of enhanced transparency of the medium - the pulses are distorted much less than in the medium of two-level atoms and retain their ability to transfer the atomic population much longer during propagation. We also show the possibility of creation of coherent superposition of two ground states in a four-level atom with a tripod-like level structure using a single, short, frequency-chirped laser pulse. The bandwidth of the pulse envelope (without chirp) must be comparable to or exceeding the frequency difference between two of the three ground levels. No appreciable excitation of the atom takes place during the creation of the coherent superposition state, thus, decoherence effects due to the spontaneous decay of the excited state are significantly suppressed. The proposed method of creation of superposition states is robust against variations in the laser pulse parameters and, being insensitive to resonance conditions, it is efficient both in homogeneously and inhomogeneously broadened media.

In the second part of the thesis we deal with atoms interacting with quantized modes of plasmonic nanostructures. Plasmonic structures are suitable for creating a structured reservoir and, as a consequence, extremely strong coupling between quantum emitters and the plasmonic modes. For an appropriate geometry, i.e., a thin metallic nano-wire, one gets a quasi-1D plasmonic reservoir that ensures strong, long-distance coupling between several quantum emitters. We investigate systems
consisting of a single, as well as two and three quantum emitters strongly coupled to surface plasmon modes of one or more metallic nano-wires, using a Green’s function approach. The formalism involving Green’s functions is especially suitable for our purposes since one can easily incorporate intrinsic material losses into the reservoir by macroscopic quantities (the electric permittivity and magnetic permeability of the media) and the position, as well as the number of emitters can simply be varied. Explicit expressions are derived for the spontaneous decay rate into the plasmon modes and for the atom-plasmon coupling as well as a plasmon-mediated atom-atom coupling. Phenomena due to the presence of intrinsic losses in the metal are discussed. In case of two atoms, we observe a Dicke subradiance and superradiance - even for separations of the atoms that exceed the plasmon wavelength by an order of magnitude - resulting from their plasmon-mediated interaction. Based on this phenomenon, we propose a scheme for a deterministic two-qubit quantum gate. For three emitters, coupled by a single or three wires, we find two- or three-qubit subradiant entangled states. The latter could possibly be a prototype for lattice coupling with frustration effects. We discuss the possible ways of driving the atomic populations into those states by the aid of external fields. Also, we propose a possible realization of interesting many-body Hamiltonians, such as the 1D spin-boson model, using strong emitter-plasmon coupling. Besides modified collective decay rates, the strong plasmon-mediated interaction also induces Lamb and dipole-dipole level shifts, the calculation of which can be quite cumbersome if done in a direct, brute-force way. We present a general method which allows for a much simpler calculation of these shifts than a direct approach. We subsequently apply the method to the case of a pair of atoms coupled to the guided modes of a nano-wire.
Kurzfassung


Im zweiten Teil der Arbeit betrachte ich die Wechselwirkung von Atomen mit quantisierten Moden von plasmonischen Nanostrukturen. Plasmonische Strukturen sind geeignet um ein strukturierten Reservoir zu gestalten, und, als eine Folge, eine extrem starken Kopplung zu erzeugen..
CHAPTER 1

INTRODUCTION AND MOTIVATION

1.1 Population transfer and coherent state preparation with single, frequency-chirped laser pulses

Different schemes of coherent population transfer and coherent creation of superposition states have been investigated extensively in recent years, [1] - [9], with a number of new applications in the field of quantum optics [10], quantum information [11, 12], generation of high harmonics and improving efficiency of nonlinear processes in resonant gases [13] - [18], control of chemical reactions [19], electromagnetically induced transparency [20] - [28] and others.

In atomic physics, there are numerous techniques that use laser pulses to coherently manipulate the quantum state of atoms. Some of the best known ones include the application of resonant $\pi$ pulses or rapid adiabatic passage (RAP) [29] to achieve population transfer in two-level systems. For atoms with a $\Lambda$ level scheme (also known as $\Lambda$-atoms), two laser pulses that fulfill the two-photon resonance condition can be used to transfer the atomic population from one of the lower (metastable) states to the other one in the adiabatic interaction regime. The pulses have to partially overlap and must be applied in a counter-intuitive order: this is the well-known stimulated Raman adiabatic passage (STIRAP) [30] - [32]. An important property of this interaction is that the population of the excited state during the process is negligibly small.

However, the previously suggested methods for transferring atomic population and creating superpositions in multilevel atomic systems require at least two laser pulses with constant carrier frequencies. Thus, the problem of synchronization of multiple pulses in time and space has to be addressed in all these schemes. This is especially relevant in optically thick media where the synchronization will inevitably be disturbed during the propagation of the pulses. Another unwanted feature is the transfer of photons from one (pump) pulse into the other (Stokes) pulse which also limits the transfer process.

Therefore, a question arises: can a single laser pulse prepare a coherent superposition of ground
states in a multilevel atom, without exciting the system? This, if true, apart from offering a simpler mechanism for creating a quantum state superposition, could prove very useful from the point of view of practical applications in optically thick media. Naturally, one must consider the propagation effects too when such a pulse is used to coherently prepare extended, optically thick media. A main point of our investigation is if, and for how long the laser pulse can induce adiabatic population transfer between the atomic levels as it propagates in the medium.

The propagation of laser pulses in quasi-resonant, optically thick media is an issue frequently investigated. The propagation problem is an interesting question in its own right, especially in a medium of coherently prepared atoms that may have peculiar and useful nonlinear optical properties \[14, 16, 17\]. Such a medium can be prepared, for example, using laser pulses that perform some coherent manipulation of the atomic quantum states, like a complete transfer of the atomic population from one atomic state to another, or the creation of a coherent superposition of quantum states. For instance, when laser pulses are used to prepare such an atomic medium it is important to know how well the pulses that perform a certain type of coherent quantum state manipulation retain their properties while propagating among the atoms of the optically thick sample.

Some of the coherent processes induced in atoms by laser pulses can give rise to startling propagation phenomena. For example, the coherent excitation and return to the ground state experienced by two-level atoms that are exposed to precisely resonant laser pulses with an envelope area of \(2\pi\) gives rise to the phenomenon called self-induced transparency (SIT): a stable propagation of "slow", soliton-like light pulses without considerable attenuation \[33\]. Soliton-like pulse pair solutions known as simultons have also been discovered \[34\].

There are other notable propagation phenomena which are (in contrast to solitons) linear effects associated with the adiabatic evolution of the atoms in a dark state under the influence of the laser pulse \[35, 1\]. For instance, in the case of a medium of \(\Lambda\)-atoms, an intense field close to resonance with one transition can have notable effects on the propagation of a weak probe pulse that is close to resonance with the other transition. The resulting phenomenon, well known as electromagnetically-induced transparency (EIT), gives rise to anomalously long propagation lengths, the slowing down, and even the storage of the weak light pulse \[36, 37\]. In case of two laser pulses with similar amplitudes traveling in a medium composed of \(\Lambda\)-atoms, stable propagation of pulse pairs is possible under various conditions. Matched pulses may propagate in a coherently prepared medium or the medium itself may generate them as response to a single input pulse under appropriate conditions \[38, 22\].

For frequency-chirped laser pulses that excite two-level atoms via RAP, there is no transparency phenomenon similar to SIT while propagating through a medium of two-level atoms. One of the several reasons is that these pulses always transfer the atoms to the excited state so they continue to lose energy during propagation. This is contrary to how resonant SIT pulses work since they return the atoms to their ground state at the end of the interaction and can therefore propagate without loss (provided that relaxation due to spontaneous emission during interaction can be neglected).

Recently, it was discovered that robust adiabatic population transfer between the two lower states
of a Λ-atom can also be induced by a single frequency-chirped laser pulse \([39, 40]\) coupling simultaneously both lower states to the upper one. In these works, the population dynamics of Λ-atoms under influence of chirped pulses has been investigated, without considering the problem of pulse propagation. It has been shown that under certain conditions, the population transfer between the two lower states is quite similar to STIRAP. The atomic population may be transferred completely from one stable state to the other, and the excited state of the atom is only weakly populated during the interaction. Thus, in a medium of Λ-atoms one may anticipate a transparency phenomenon to exist for chirped laser pulses that fulfill the conditions for such an adiabatic population transfer. However, there is one major difference between the two processes: in case of the chirped pulse, since the frequency-modulated field interacts with both transitions, the photons are absorbed from and emitted into the same field by the atoms, in contrast to the STIRAP where two fields with constant carrier frequencies are present and each of them interacts with only one atomic transition.

Here, we examine the propagation of frequency-chirped laser pulses through a medium of Λ-atoms, pulses that are capable of producing adiabatic population transfer between two of the lower (metastable) states of the atomic level structure - this question is especially important when considering chirped pulses as a tool for manipulating atoms in a spatially extended region.

We compare the propagation of these pulses to that in a medium of two-level atoms, and show that the distortion of the pulses is greatly reduced in the three-level case. We show that the pulse traveling in a medium of Λ-atoms maintains its capability to produce adiabatic population transfer for a much larger distance than the same pulse traveling in a medium of two-level atoms. In fact, there exists a transparency phenomenon for frequency-chirped laser pulses propagating in a medium of Λ-atoms that is somewhat similar to electromagnetically induced transparency. The most important difference is, however, that in our case it is the chirped pulse that renders the medium transparent for itself. We calculate the propagation through both an inhomogeneously broadened medium and a medium having only homogeneous broadening and discuss the similarities and differences of the present process with EIT and the propagation of matched pulses.

Another interesting and relevant problem is when, instead of completely transferring atomic population from one state to the other, we want to prepare a quantum state superpositions in an atomic ensemble. A well-known technique for coherent generation of such superpositions is the application of pulses with envelope areas equal to \(\pi/2\). This method, similarly to the \(\pi\) pulse technique for population transfer between quantum states is sensitive to the laser pulse area, the resonance conditions and the transverse distribution of the laser beam intensity \([41]\). A technique for creation and measurement of a coherent superposition of two ground (metastable) states based on tripod-STIRAP and on adiabatic evolution of the superposition states was developed in \([2-4]\). The drawback of the latter approach is the requirement of two-photon Raman resonance that is necessary for the STIRAP scheme to be efficient. While this condition may be achieved in atomic beams with narrow velocity distribution of the atoms (small Doppler shift of the resonance lines), it cannot be fulfilled in inhomogeneously broadened quantum systems such as an ensemble of quantum dots or rare-earth-metal-ion doped crystals \([42]\) or when a single laser pulse must be used for population
transfer and creation of coherent superposition of quantum states in atoms having a Λ level linkage \[9, 10\]. The same is true when a single laser pulse is applied for coherent population transfer between Rydberg states in e.g. potassium atoms \[43\]. Note that an extension of the STIRAP scheme that is applicable to degenerate levels allows creation of a predefined superposition of multiple magnetic sublevels \[44, 45\].

The laser pulses are assumed to be narrowband in the majority of the schemes dealing with the creation of coherent atomic or molecular quantum state superpositions, or in the schemes of coherent population control of the quantum states, i.e., the bandwidth of the laser radiation is assumed to be narrower than the frequency separation between the members of any two atomic (molecular) levels involved in the interaction. Usually it is also assumed that each laser pulse involved in the process interacts with a single quasi-resonant transition.

There are papers, though, which investigate with cases of intense narrowband laser pulses where, due to the Rabi frequency exceeding the frequency spacing between the atomic levels, the pulse interacts with multiple transitions in the atom, see for example \[46, 47\] and \[4\]. For example, in the case of short, frequency-chirped laser pumping acting on Λ-atoms, analyzed in \[40\], the bandwidth of the laser pulse may be larger than the spacing between some quantum levels involved in the interaction.

In such cases, i.e., when the bandwidth of the short-pulsed laser radiation exceeds the energy separation between two atomic levels involved in the interaction (from now on, referred to as "close" levels), one can apply a basis transformation so that instead of the two "close" levels we get their "dark" and "bright" superpositions. From these two new states, the dark one is effectively decoupled from the laser field, whereas the bright state and the excited state are fully involved in the interaction process. Thus, instead of two unresolved states we have to regard only a single state coupled to the excited state.

Accordingly, if a Λ-linkage atom interacts with a frequency-chirped short laser pulse in the adiabatic regime, the bright and dark superpositions of the ground states are separated from each other: the population of the bright component is transferred to the excited state of the Λ-atom and the dark component is left intact (\[7, 8, 9\]). This is a robust method for creating coherent superposition of metastable states by a single frequency-chirped laser pulse. The drawback of this method is, however, that the atoms get excited which leads to decoherence processes due to spontaneous decay from the excited state.

In the thesis it is shown that it is possible to create a coherent superposition of ground states in a four-level atom with a tripod level structure (i.e., three ground states and a single excited state), using a single, short, frequency-chirped laser pulse. An important additional requirement is that two of the ground states have a frequency separation smaller than the bandwidth of the laser pulse envelope (thus, they are termed "close" states) and the frequency distance of the third, "far" ground state from the two "close" ones exceeds the envelope bandwidth. We show that for a sufficiently strong laser pulse, i.e., having a peak Rabi frequency exceeding the largest frequency separation between the ground states, the frequency-chirped laser pulse may efficiently transfer the bright
superposition of the two "close" ground states into the third, "far" ground state without appreciable excitation of the atom, leaving the "dark" superposition untouched. This means that in the end of the interaction a dark superposition of the two "close" ground states is created. The process can also be used to map the "far" state on the bright superposition of the "close" states if one changes the sign of the chirp and the initial population is in the "far" ground state. Besides being an efficient method for creating a coherent superposition of the ground states while suppressing atomic excitation, the scheme is robust against variations of the pulse parameters such as maximum intensity, duration and chirp rate. Because there are no restrictions concerning resonance conditions, due to the frequency chirp of the pulse, this method is efficient both in homogeneously and inhomogeneously broadened quantum systems.

1.2 Collective behaviour of an atomic ensemble due to an engineered reservoir

In the second part of the thesis we examine the behaviour of systems where multiple atoms are interacting with the quantized modes of a tailored reservoir. Due to this coupling, effective interactions arise between the atoms. As a result, a collective atomic behaviour emerges where atoms trade excitations among each other with the assistance of the reservoir and it is no longer possible to distinguish between the individual atoms. Instead, we get collective atomic states involving the whole ensemble, having modified decay rates and energy shifts due to the atom-reservoir and the effective atom-atom interactions. As one might expect, the magnitude of these shifts and decays heavily depends on the strength of the atom-reservoir coupling and the properties of the reservoir.

In order for the effective atom-atom interactions to be strong, it is important for the atoms to be coupled to a few modes of the reservoir much more strongly than to the others. In this way the probability that a photon emitted by one of the atoms is going to be picked up by another is greatly increased and so is the strength of the effective interaction between those two atoms. The geometry of the reservoir is a crucial property for determining which modes are coupled stronger and which weaker to the atoms. The reason for this is that the coupling strength depends on the local density of states which is greatly influenced by the geometry of the system. In particular, dimensionality plays an important role. In free space the radiative atom-atom coupling quickly disappears as soon as the average distance between the emitters exceeds the resonant radiation wavelength as the Green's function of the electromagnetic field in 3D decays as $1/r^2$. However, the situation changes dramatically if the most dominant reservoir modes are reduced to one or zero spatial dimensions as in the case of a nano-wire or a single-mode resonator. Here interactions over large distances can emerge [48].

Such radiative coupling between selected modes of the reservoir and the atomic ensemble is extremely important in the field of quantum optics and quantum information. A strong coupling between individual quantum emitters and photons as well as the emerging long-lived (subradiant) collective entangled atomic states are key ingredients for photon-based quantum information pro-
processing. They are needed to achieve a reliable transfer of excitation between stationary and flying qubits and to realize quantum logic operations.

This kind of coupling can be achieved e.g. when quantum emitters are coupled to surface plasmons of conducting nanowires as proposed in [49, 50]. Besides its simple structure, the atom-nanowire scheme guarantees an exceptionally strong field-emitter coupling. The original setup involves a single atom placed close to the surface of a metallic wire having sub-wavelength radius. In the present paper we analyze this scheme in detail by means of a Green function approach taking into account metal losses and extend it to a plasmon-mediated coupling between different atoms.

The unique properties of the propagating electromagnetic modes of the wire, the surface plasmons, have opened up new possibilities in many fields such as waveguiding below the diffraction limit [51], sub-wavelength imaging [52, 53] or enhanced fluorescence [54 - 58]. The small transverse-mode area also enables one to strongly couple an emitter to the plasmon, giving rise to a substantial Purcell effect with a Purcell factor of $10^2$ or larger. The effect of strong coupling to the plasmon modes of a waveguide has been studied for various specific scenarios ([57, 59]). The system has been proposed as an efficient single-photon generator [49, 50] as well as a single-photon transistor [60], strong emitter-plasmon coupling has been experimentally shown using quantum dots [61], single plasmons along the wire have been detected, using N-V centers as emitters [62] and the wave-particle duality of surface plasmons has been verified in experiment [63].

In our approach, the structure and all other properties of the reservoir are contained by the dyadic Green’s function which is thus also involved in the quantization of the reservoir modes. This is a convenient way to treat systems where multiple atoms are coupled to a common reservoir since the same Green’s tensor can be used for treating cases of various numbers and configurations of atoms. Also, it is very simple to incorporate intrinsic losses into the reservoir using this kind of formalism. Thus, both the collective decay rates as well as the effective interaction Hamiltonian are fully determined by the dyadic Green’s function of the electromagnetic field characterizing the response of the tailored reservoir.

In the thesis, we describe a system where a single, as well as two emitters interact with the surface plasmons of the wire in a fully quantum mechanical approach (see also in [64]). As mentioned above, due to the strong coupling, a substantial Purcell effect is observed if the emitter is close enough to the wire that results in a single-atom decay rate enhancement on the order of magnitude $10^2$ relative to the decay rate in vacuum. We also discuss the scaling of plasmon propagation losses with respect to the geometrical parameters of the wire. Subsequently, we derive an expression for calculating the decay rate directed only into the plasmon modes and relate this to the atom-plasmon coupling strength. Analysing the expressions, we see that there is an optimum wire-atom distance where the decay rate into the plasmons is maximal.

The plasmons mediate an intense long-range interaction between different emitters. As a result, we observe sub- and superradiance in a two-atom system which can be efficiently controlled by adjusting the distance between the atoms. Even for a distance of several atomic transition wavelengths we get a good superradiance-subradiance contrast. This yields a system where subradiant entangled
1.3 Outline of the Thesis

The thesis is comprised as follows. Chapter 2 contains the results of our investigations concerning the interaction of atomic ensembles with strong, classical frequency-chirped laser fields. First, in Sec. 2.1 we present the dressed state analysis of the population transfer for a single atom, then, subsequently, in Sec. 2.2 we discuss the propagation of a single, frequency-chirped laser pulse that transfers the population between the ground states of a $\Lambda$-linkage atom, in a medium of such atoms. Within, in Sec. 2.2.1 an analytic estimation is given for the behaviour of the laser pulse upon propagation which is followed by the results of the numerical simulation of the propagation problem in Sec. 2.2.2. Also, the effects of homogeneous and inhomogenous broadening of the medium on the
propagating pulse are discussed. Subsequently we examine the dynamics of the coherent creation of a quantum state superposition of ground states in a four-level tripod-linkage atom using a single, frequency-chirped pulse, in Sec. 2.3. The envelope bandwidth of the pulse is larger than the frequency separation between two of the ground states but smaller than the frequency separation between the mentioned "close" ground states and the third ground state.

Chapter 3 deals with the interaction of atoms coupled to the quantized modes of a common tailored reservoir, with special respect to atoms strongly coupled to surface plasmon eigenmodes of metallic nanowires - systems where strong effective atom-atom coupling is realized.

We first present the tools used for our investigations. Sec. 3.1 contains the quantization scheme involving the Green’s tensor of the reservoir. After that, in Sec. 3.2 we briefly introduce the dyadic Green’s functions. Following this, we derive the master equation for the atomic system in Sec. 3.3 where we can witness the emergence of the modified single-atom decay rates, effective atom-atom couplings and dipole-dipole shifts as the manifestation of the effective interaction between the atoms. We discuss the significance of the diagonal basis for the master equation (a special basis where the collective atomic decay rates acquire a physical meaning) and present a method to calculate it. In Sec. 3.4 we describe the mode structure of the surface plasmons of a metallic, infinitely long, cylindrical wire.

The necessary tools thus introduced, the results of our investigations about atoms coupled to surface plasmon modes of a cylindrical nanowire are presented. We first take a look at the behaviour of a single emitter coupled to the surface plasmons of a wire (Sec. 3.5), then, in Sec. 3.6 we examine the long-range interaction of two atoms, both of which are strongly interacting with the basic plasmonic mode of the wire and find a Dicke superradiance effect that persists over inter-atomic distances of several optical wavelengths. We also establish a relation between the propagation length of plasmons in a lossy wire and the range of the superradiance effect. Subsequently, in Sec. 3.7 we propose a scheme for a deterministic quantum phase gate based on this phenomenon. Sec. 3.8 deals with the collective states and decay rates of three emitters coupled to the modes of a single as well as three metallic wires. We also propose schemes for transferring the atomic population into the entangled, subradiant collective states by means of external, resonant laser pulses. As another application, we propose the use of the atom-wire system for the realization of a 1D spin-boson model, in Sec. 3.9 since in such a system the atoms are coupled to a one-dimensional continuum of bosonic modes.

As the last topic of the thesis, in Sec. 3.10 we look at the modification of Lamb and dipole-dipole shifts resulting from the coupling to the tailored reservoir and the effective atom-atom interactions. We introduce a general method that greatly reduces the difficulties associated with the calculation of these energy shifts and, subsequently we apply it for the case of a pair of emitters interacting with the basic plasmonic mode of a metallic nanowire.

The Appendices contain the information that is important from the point of view of understanding the technicalities but have been left out from the main body of the thesis in order to enhance transparency.
Part I

Population transfer and coherent state preparation with chirped laser pulses
As already mentioned in the Introduction, if we want to prepare a single atom in a desired quantum state by aid of quasi-resonant laser fields, there are several methods at our disposal to choose from. However, if our aim is to do so with an extended, optically thick medium, we have to carefully choose the scheme that is most efficient for the purpose, i.e., the process should be robust against pulse parameter variations and the pulse, while propagating in the medium, should maintain its population-transferring properties as long as possible.

For a two-level system, one can use single pulses with an envelope area of \((2n + 1)\pi\), transferring the atomic population from ground state A into excited state B through a dipole-allowed transition. However, in case of an extended medium, we face several problems. For instance, the pulse area must be very precisely adjusted in order to achieve complete population transfer. Furthermore, the atomic back-action on the pulse is going to be substantial because the transition has a non-zero dipole moment and there is a continuous energy transfer from the pulse to the atoms.

The problem of pulse area fine-tuning is eliminated if, instead of a \(\pi\) pulse, one applies a frequency chirped pulse, transferring the population by means of RAP process. Unfortunately, the problem of energy loss and strong pulse distortion is still present when the pulse propagates in the medium.

Choosing a medium where the transition between A and B is dipole-forbidden, for example a three level \(\Lambda\)-atom, can help to eliminate the pulse distortion due to the vanishing dipole moment of the atomic transition. If one uses a STIRAP process to transfer the population from ground state A to ground state B, the envelopes and the overlap of the pulses do not need a rigorous fine-tuning. Furthermore, upon propagation instead of losses, the energy is gradually transferred from the pump pulse into the Stokes pulse, resulting in a greatly enhanced propagation length without substantial change in population transfer efficiency compared to the preceding schemes. However, one still has to synchronize the pump and Stokes pulses with each other, and, as an additional problem, the process is quite vulnerable to inhomogeneous frequency broadening of the medium since it is crucial
for the transfer efficiency that the pulses be resonant with the atomic transitions.

In the following, we discuss the application of a method, involving a frequency-chirped laser pulse, that takes care of all the problems mentioned above. Besides transferring the population from one ground state of a Λ-atom to the other, it is robust against the variation of the pulse parameters (i.e., the amplitude, the chirp rate and the central frequency) and, interacting with both transitions of the atoms, the photons are absorbed from and emitted into the same field. Additionally, because the frequency is chirped, there are no strict resonance conditions required, the effect of inhomogeneous broadening upon the process is strongly reduced. We investigate the propagation of such a pulse in a medium of Λ-atoms and present a method which, instead of transferring the whole population from one state to the other, produces a coherent superposition of ground states in a four-level tripod atom by use of a single, frequency-chirped laser pulse.

2.1 Population transfer in a Λ-atom by a single frequency-chirped pulse: a dressed state analysis

To describe the population transfer process in a three-level Λ-atom due to the interaction with a single, frequency-chirped laser pulse, we reexamine the corresponding part of the discussion found in [40].

![Level structure of the Λ-atom](image)

Figure 2.1: Level structure of the Λ-atom. Two stable or metastable states (|1⟩, |3⟩) are connected to a single excited state (|2⟩), the transition frequencies being ω_{12} and ω_{32}.

The three-level Λ-linkage atom, depicted in Fig. 2.1, has two stable (or metastable) states which we refer to as ground states and a single excited state. Dipole transitions are only allowed between a ground state and the excited state, the transition between the two ground states is dipole-forbidden. We assume that the total atomic population is in ground state |1⟩ prior to the interaction with the pulse. Furthermore, we require that the envelope is sufficiently long so that the transform-limited bandwidth of the pulse (i.e., the bandwidth without chirp) is smaller than the frequency difference ω_{13} ≡ ω_R between the two ground states of the atom. In this way, the pulse interacts separately
with the two atomic transitions. For the sake of simplicity, we also assume that the pulse duration is shorter than the decay time of the excited state $|2\rangle$, i.e., we neglect spontaneous decay from $|2\rangle$ for the moment. We thus can describe the dynamics of the system using the Schrödinger equation. Later on we shall see that the excited state population can be made very small, thus allowing us to extend the results also to longer pulses.

For convenience, we represent the atomic state vector as

$$a = a_1 e^{-i\omega_1 t} \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} + a_2 e^{-i\varepsilon_{21}(t)t} e^{-i\omega_2 t} \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} + a_3 e^{-i(\varepsilon_{23}(t)-\varepsilon_{21}(t))t} e^{-i\omega_3 t} \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix} \tag{2.1}$$

where

$$\begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} = |1\rangle, \quad \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} = |2\rangle, \quad \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix} = |3\rangle, \tag{2.2}$$

$\omega_{1,2,3}$ are the angular frequencies associated with the respective states of the atom and $a_{1,2,3}$ are the corresponding probability amplitudes.

$$\varepsilon_{21} = \omega_L(t) - \omega_{21}$$

$$\varepsilon_{23} = \omega_L(t) - \omega_{23} \tag{2.3}$$

are detunings from the one-photon resonance where $\omega_L(t)$ is the time-dependent carrier frequency of the laser pulse and $\omega_{21}, \omega_{23}$ are the resonant transition frequencies between the corresponding states (see Fig. 2.1).

The Schrödinger equation then assumes the form

$$i \frac{d}{dt} a = \bar{H} a \tag{2.4}$$

where the rotating-wave approximated Hamiltonian in the bare-state basis is

$$\bar{H} = -\begin{pmatrix} 0 & \Omega_{12} & 0 \\ \Omega_{21} & \varepsilon_{21} + \frac{d}{dt}\varepsilon_{21} & \Omega_{23} \\ 0 & \Omega_{32} & \omega_R \end{pmatrix}. \tag{2.5}$$

The matrix contains the $\Omega_{ij} = \Omega_{ij}^* = 1/(2\hbar)d_{ij}A(t)$ Rabi frequencies with $(i,j = 1,2,3)$, $d_{ij}$ being the dipole moment matrix element for transitions $|j\rangle \rightarrow |i\rangle$ and $A(t)$ is the (real) envelope of the laser pulse. From here on, we assume the laser pulse to have a carrier frequency

$$\omega_L(t) = \omega_{L0} + \beta t, \tag{2.6}$$

where $\omega_{L0}$ is the central frequency and $\beta$ is the chirp rate. Note that the Raman frequency $\omega_R = \omega_{L0} + \beta t$ is a constant dependent on the chirp rate.
\( \varepsilon_{21}(t) - \varepsilon_{23}(t) = \omega_{13} \) is a constant quantity.

To understand the population dynamics we proceed with a dressed state analysis of the interaction. The atomic state vector can be expanded in the eigenbasis of the Hamiltonian (2.5), i.e., the dressed eigenstates, as

\[
a(t) = \sum_{k=1}^{3} C_k(t)b^{(k)}(t),
\]

where \( C_k(t) \) are time-dependent expansion coefficients and

\[
\tilde{H}b^{(k)}(t) = w_k(t)b^{(k)}(t)
\]

with \( w_k(t) \) being the quasienergies, i.e., the eigenvalues belonging to the corresponding dressed eigenstate \( b^{(k)}(t). \) Writing up the Schrödinger equation using the expansion (2.7), we obtain

\[
i \sum_k \left( \dot{C}_k b^{(k)}(t) + C_k(t) \dot{b}^{(k)}(t) \right) = \sum_k C_k(t)w_k(t)b^{(k)}(t).
\]

Supposing that the rate of change in probability amplitudes \( C_k(t) \) is much faster than that in the dressed state eigenvectors \( b^{(k)}(t), \) i.e.,

\[
|\dot{C}_k b^{(k)}(t)| \gg |C_k(t) \dot{b}^{(k)}|,
\]

we perform an adiabatic approximation, neglecting the second term on the LHS of (2.9). With this, we assume that the interaction is slow enough so that the system is able to adiabatically follow the dressed states of the Hamiltonian (see also \[30\]). Note that an equivalent, general condition for adiabaticity is (\[69\])

\[
|\langle \dot{b}^{(i)}(t)|b^{(j)}(t)\rangle| \ll |w_i(t) - w_j(t)|, \quad (i \neq j)
\]

setting a lower limit to the difference between quasienergies during the interaction.

Solving the Schrödinger equation in adiabatic approximation and expressing the bare state probability amplitudes with the solution yields

\[
a(t) = \sum_k R_k b^{(k)}(t) \exp \left[ -i \int_{-\infty}^{t} dt' w_k(t') \right],
\]

with the initial condition as \( t \to -\infty \)

\[
a(-\infty) = \sum_k R_k b^{(k)}(-\infty)
\]

where the \( R_k \) are constant coefficients. Solving the eigenvalue problem (2.8), we obtain the following characteristic polynomial for the quasienergies:

\[
w^3 - w^2(\omega_{13} + \varepsilon_{21}) + w \left[ \varepsilon_{21}\omega_{13} - (|\Omega_{12}|^2 + |\Omega_{32}|^2) \right] + |\Omega_{12}|^2\omega_{13} = 0
\]

(2.14)
and the components of the dressed state vectors, corresponding to the root \( w_k \) of (2.14), assume the form

\[
\begin{align*}
    b_1^{(k)} &= \frac{\Omega_{12}(w_k - \omega_{13})}{\sqrt{N}}, \\
    b_2^{(k)} &= \frac{w_k(w_k - \omega_{13})}{\sqrt{N}}, \\
    b_3^{(k)} &= \frac{\Omega_{32}w_k}{\sqrt{N}}, \\
\end{align*}
\]  

(2.15)

where the normalization factor

\[
N = \omega_{13}^2(w_k - \omega_{13})^2 + w_k^2(w_k - \omega_{13})^2 + \Omega_{32}^2w_k^2.
\]

Note that because the laser pulse spectrally resolves the two transitions of the \( \Lambda \)-atom, the Raman detuning \( \omega_R = \omega_{13} \) is non-zero and, as a consequence, the characteristic polynomial does not have a root \( w = 0 \).

\[\text{Figure 2.2: Time evolution of the quasienergies during the action of a single, frequency-chirped laser pulse on a \( \Lambda \)-atom. One observes that the value of } w_1\text{ is restricted between 0 and } \omega_R\text{ (the frequency difference between the ground states). Dashed lines are the diabatic lines, i.e., quasienergies in presence of zero Rabis frequencies. The quasienergies are plotted in units of the frequency distance } \omega_R\text{ between the ground states and the time is normalized by the pulse duration } \tau_p.\]

The dynamics of the quasienergies for a laser pulse with a Gaussian envelope \( A(t) \) and a positive, linear frequency chirp (\( \beta > 0 \)) are plotted in Fig. 2.2. The value of the quasienergy denoted by \( w_1 \) is restricted between zero and \( \omega_R \), the frequency distance between the ground states, independently from the value of the Rabi frequency of the laser pulse:

\[0 \leq w_1 \leq \omega_{13} = \omega_R.\]  

(2.16)

Furthermore, the asymptotic values of \( w_1 \) are \( w_1(-\infty) = 0 \) and \( w_1(\infty) = \omega_{13} \). This, in turn means, considering (2.15), that for the dressed state \( b^{(k)} \) we obtain

\[
\begin{align*}
    b^{(1)}(-\infty) &= \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}, &
    b^{(1)}(\infty) &= \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}
\end{align*}
\]  

(2.17)
which means that if the atomic system is initially in state $|1\rangle$, adiabatically following $b^{(1)}(t)$, it ends up in $|3\rangle$ so that the complete atomic population is transferred from one ground state to the other.

One also observes, however, that $b^{(1)}$ has a non-zero projection on the excited state $|2\rangle$ during the interaction. If the amount of population temporarily transferred into $|2\rangle$ is substantial, the efficiency of the transfer process may be considerably reduced due to the presence of spontaneous emission in a realistic system.

Using (2.15), we have the following relation between the dressed state vector components

$$\frac{b_2^{(1)}}{b_1^{(1)}} = \frac{w_1}{\Omega_{12}}$$

(2.18)

which, combined with (2.16), leads to

$$\left| \frac{b_2^{(1)}}{b_1^{(1)}} \right| \leq \frac{\omega_R}{\Omega_{12}}$$

(2.19)

Thus, the contribution of $|2\rangle$ decreases as the Rabi frequency increases and becomes negligibly small if $\Omega_{12} \gg \omega_R$, i.e., when the Rabi frequency is much larger than the frequency gap between the two ground states of the $\Lambda$-atom.

Figure 2.3: Atomic state population dynamics of a $\Lambda$-atom under the action of a single, frequency-chirped laser pulse in the adiabatic approximation. The population, initially in ground state $|1\rangle$, is transferred to ground state $|3\rangle$ with negligible excitation during the interaction. The dashed line is the normalized envelope of the pulse. The parameters applied are the pulse duration $\tau_p = 1ns$, frequency distance between the ground states $\omega_R = 1GHz$, frequency chirp rate $\beta = 2GHz$ and the peak Rabi frequency $\Omega_0 = 10GHz$. Time is normalized by $\tau_p$.

Fig. 2.3 shows the adiabatic population dynamics for a pulse where $\Omega_{12} = 10\omega_R$. Juxtaposing it with Fig. 2.2 one observes that during the interaction, the change of the quasienergy $w_1$ is
indeed linked with the change of atomic populations: at the start of the pulse, when the chirp sweeps through transition \(|1⟩ - |2⟩\), as well as towards the end, when the other transition is swept through, \(w_1\) changes more rapidly - and the population change of the ground states also has a steeper slope. Between these two regions, where \(w_1\) remains almost constant, the populations do not change considerably, either. By the end of the interaction, the population is completely transferred from state \(|1⟩\) to state \(|3⟩\) while the population of the excited state is small throughout the process: where the pulse is strong, excitation is efficiently suppressed and even in the wings, where \(\Omega_{12}\) is small, there is only a slight population of the excited state.

Throughout the preceding analysis we neglected spontaneous emission from the excited state \(|2⟩\). However, using the obtained results, one can estimate the losses due to relaxation. The population lost during the interaction with the pulse can be written as

\[
L = \int_{-\infty}^{\infty} dt \Gamma |a_2(t)|^2
\]  
(2.20)

where \(\Gamma\) is the spontaneous emission rate. Since in adiabatic approximation the system follows the dressed state \(b^{(1)}(t)\) thus, according to 2.15 one can express \(a_2(t)\) as

\[
a_2(t) = b_2^{(1)}(t) = \frac{w_1(t)(w_1(t) - \omega_{12})}{\sqrt{N}}.
\]  
(2.21)

Examining the behaviour of \(w_1(t)\) in Fig. 2.2 and recalling (2.81), one has the relation

\[
|w_1(t)(w_1(t) - \omega_{13})| \leq |\omega_{13}^2\left(\frac{\omega_{13}}{2} - \omega_{13}\right)| = \frac{\omega_R^2}{4}.
\]  
(2.22)

Thus, the duration of the pulse being \(\tau_p\), we approximate \(w_1\) as

\[
w_1(t) \approx \frac{\omega_{13}}{2} \left(\Theta(-\frac{\tau_p}{2}) + \Theta(\frac{\tau_p}{2})\right)
\]  
(2.23)

and substituting this into (2.21), for the estimated population loss during interaction we obtain

\[
L = \int_{-\infty}^{\infty} dt \Gamma |a_2(t)|^2 \lesssim \frac{\tau_p \Gamma}{5 + \frac{4\Omega_{12}^2}{\omega_{13}^2}}.
\]  
(2.24)

According to this result, in case \(\Omega_{32} \gg \omega_R\), one can efficiently suppress losses originating from spontaneous emission even if \(\tau_p \Gamma \gg 1\).

### 2.2 Propagation of frequency-chirped laser pulses among Λ-atoms

In Sec. 2.1 we saw that using a single chirped laser pulse could be an efficient way to transfer the population of an ensemble of Λ-atoms from one ground state into the other. In the current Section we discuss the propagation of such a pulse in an extended, optically thick medium of Λ-atoms.
First, we present analytic estimations in the adiabatic approximation, and, subsequently, the numerical simulation of the full problem is performed in the presence of homogeneous, as well as inhomogeneous frequency broadening of the medium. We compare the propagation of the same, frequency-chirped laser pulse in a medium of three-level $\Lambda$-atoms and in that of two-level atoms.

2.2.1 Analytic calculations in the adiabatic regime

Since the laser pulse, as we have seen it in Sec. 2.1 has a large Rabi frequency in order to suppress atomic excitation while performing the population transfer, we consider the propagating field to be classical. For that reason, the propagation is discussed by means of Maxwell’s equations. For simplicity, we only investigate the propagation in one spatial dimension. Thus, the wave equation for the electric field strength vector assumes the form

$$\partial_x^2 E(x,t) - \frac{1}{c^2} \partial_t^2 E = \mu_0 \partial_t^2 P(x,t),$$

(E being a transverse field in the vacuum between the atoms ($\nabla \cdot E = 0$). $c$ is the vacuum speed of light, $\mu_0$ is the magnetic permeability of vacuum and $P$ is the macroscopic polarization created by the atomic ensemble.

Since only the electric field component parallel to the atomic polarization vector is affected upon propagation, we can omit the vector notation and use scalar fields instead. We write the electric field in the convenient form of

$$E(x,t) = A(x,t)(e^{i(kx - \omega L_0 t + \phi(x,t))} + c.c.)$$

where $A(x,t)$ is the real-valued amplitude, $k$ is the wave number and $\omega L_0$ is the central carrier frequency of the field. $\phi(x,t)$ is the part of the phase of the field that contains the frequency modulation and at the medium entrance ($x = 0$) we set it to be a linear chirp:

$$\phi(0,t) = \frac{1}{2} \beta t^2,$$

$\beta$ being the chirp rate. Similarly, we write the polarization density as

$$P(x,t) = \eta(u(x,t) + iv(x,t))e^{i(kx - \omega L_0 t + \phi(x,t))} + c.c.$$  

where $\eta$ is the density of the atoms, $u(x,t)$ and $v(x,t)$ are real-valued functions related to single-atom polarization. Substituting (2.26) and (2.28) into (2.25) and assuming slowly varying amplitudes and phase in time and space, i.e.,

$$|\partial_t A| \ll |\omega A|, \quad |\partial_x A| \ll |k A|, \quad |\partial_t u| \ll |\omega u|, \quad |\partial_t v| \ll |\omega v|,$$

$$|\partial_t^2 A| \ll |\omega^2 A|, \quad |\partial_x^2 A| \ll |k^2 A|, \quad |\partial_t^2 u| \ll |\omega^2 u|, \quad |\partial_t^2 v| \ll |\omega^2 v|,$$

$$|\partial_t \phi| \ll \omega, \quad |\partial_x \phi| \approx 0, \quad |\partial_t \phi| \ll |k|, \quad |\partial_x^2 \phi| \approx 0$$  

(2.29)
one can separate real and imaginary parts of the equation and obtain
\[ \left[ \frac{\partial}{\partial x} + \frac{1}{c} \frac{\partial}{\partial t} \right] A(x,t) = -\frac{\mu_0 \eta \omega L_0 c}{2} v(x,t) \]
\[ A(x,t) \left[ \frac{\partial}{\partial x} + \frac{1}{c} \frac{\partial}{\partial t} \right] \phi(x,t) = \frac{\mu_0 \eta \omega L_0 c}{2} u(x,t), \] (2.30)

the absorption and the dispersion of the field being governed by the imaginary and the real part of the polarization, respectively. The equations can be further simplified by choosing a frame of reference moving together with the light pulse. Choosing new space and time variables
\[ \xi = x, \quad \tau = t - \frac{x}{c} \] (2.31)
equations (2.30) transform into
\[ \partial_\xi A(\xi,\tau) = -\frac{\mu_0 \eta \omega L_0 c}{2} v(\xi,\tau) \]
\[ A(\xi,\tau) \partial_\xi \phi(\xi,\tau) = \frac{\mu_0 \eta \omega L_0 c}{2} u(\xi,\tau). \] (2.32)

As in Sec. 2.1 for simplicity we neglect spontaneous emission from the excited state for the moment and assume that the atomic system can be represented with a single wave vector
\[ |\psi\rangle = a_1 e^{-i\omega_1 t}|1\rangle + a_2 e^{-i\varepsilon_{21}(t)} e^{-i\omega_2 t}|2\rangle + a_3 e^{-i\omega_R} e^{-i\omega_3 t}|3\rangle \] (2.33)
where, because of propagation, we have to generalize the definition (2.3):
\[ \varepsilon_{21}(x,t) = \omega_{L0} t - kx - \phi(x,t) - \omega_{21} t. \] (2.34)
\omega_{21} is the angular frequency difference between states |1\rangle and |2\rangle, see Fig. 2.1. The connection between the macroscopic polarization and the expectation value of the atomic dipole moment operator is
\[ P = \eta \langle d \rangle \] (2.35)
where the matrix representation of the dipole moment operator written in the bare state basis of the \( \Lambda \)-atom has the form
\[ \bar{d} = \begin{pmatrix} 0 & d_{12} & 0 \\ d_{21} & 0 & d_{23} \\ 0 & d_{32} & 0 \end{pmatrix}, \] (2.36)
the elements \( d_{ij} \) being the transition dipole matrix elements between states \( |i\rangle \) and \( |j\rangle \). Calculating the expectation value for \( \bar{d} \) in state \( |\psi\rangle \) we get
\[ \langle d \rangle = (d_{12} a_1^* a_2 + d_{32} a_3^* a_2) \text{e}^{i(kx - \omega_{L0} t + \phi(x,t))} + \text{c.c.}, \] (2.37)
from which it follows that
\[ u = \text{Re}(d_{12}a_1^*a_2 + d_{32}a_3^*a_2) \]
\[ v = \text{Im}(d_{12}a_1^*a_2 + d_{32}a_3^*a_2). \] (2.38)

Substituting (2.38) into (2.32), we are left with a system of coupled Maxwell-Schrödinger equations to solve. However, assuming that the properties of the laser pulse allow for the adiabatic approximation, we can make use of the results obtained in Sec. 2.1. Instead of solving the Schrödinger equations, we express the bare probability amplitudes with the dressed state vectors according to (2.12). As a result, only the Maxwell part of the problem is left to be solved, i.e., we obtain
\[ \partial_\xi A(\xi, \tau) = \frac{\mu_0 \eta \omega L_0 c}{2} \text{Im}(d_{12}b_1^*b_2 + d_{32}b_3^*b_2) \]
\[ A(\xi, \tau) \partial_\xi \phi(\xi, \tau) = \frac{\mu_0 \eta \omega L_0 c}{2} \text{Re}(d_{12}b_1^*b_2 + d_{32}b_3^*b_2) \] (2.39)

where the dressed state vector components are defined by (2.15) and, the Hamiltonian being Hermitian, real-valued. Without lack of generality, choosing the dipole matrix elements real the field equations assume the form
\[ \partial_\xi A(\xi, \tau) = 0 \]
\[ \partial_\xi \phi(\xi, \tau) = \frac{\mu_0 \eta \omega L_0 c}{2\hbar N} \left[ |d_{12}|^2 w_1(t)(w_1 - \omega_R)^2 + |d_{32}|^2 w_1^2(t)(w_1(t) - \omega_R) \right]. \] (2.40)

So, as a result we get that in adiabatic approximation the envelope of the field is not affected during the propagation process. As for the phase, the RHS contains a complicated expression so that there is not much hope of analytically solving the second equation in (2.40). However, because the values of the quasienergy \( w_1(t) \) is restricted according to (2.16), we can expect that \( \phi \) is only moderately affected upon propagation.

Thus, we anticipate that a frequency-chirped laser pulse inducing population transfer between the ground states of a \( \Lambda \)-atom has an enhanced propagation length before it loses its ability to do so. To confirm these expectations, we performed a numerical simulation of the full atom-field interaction, taking into account spontaneous emission from the excited state as well.

### 2.2.2 Numerical simulation of the full problem

In order to prove whether a frequency-chirped laser pulse, that initially transfers the atomic population from one ground state of a \( \Lambda \)-atom to the other, is indeed capable of propagating a considerable distance before losing this ability, we compared the propagation of the same pulse in a medium consisting of three-level \( \Lambda \)-atoms and among two-level atoms. The justification of this comparison is that the interaction involves single-photon transitions between a ground state and a decaying excited state in both cases, the only difference is that for the \( \Lambda \)-atoms the target state is a ground state and for the two-level atoms it is the excited state.
To treat the problem of the frequency-chirped laser pulse propagating in a medium of $A$-atoms we follow the well-established procedure of using the classical Maxwell’s equations for the electric field of the pulses and the equation of motion for the density operator for the time evolution of the atomic variables $[41]$. Unlike our analytic treatment where we regarded real-valued field amplitude and phase separately, it is now more convenient to write the electric field of the laser pulse as

$$E(x,t) = \hat{A}(x,t)e^{i(kx-\omega L_0 t)} + c.c.$$

(2.41)

where $\hat{A}(x,t)$ is the complex field amplitude varying slowly in time and space compared to the central laser frequency $\omega L_0$ and wave number $k$ (assumed to be constants). The frequency-chirp is contained in the phase of the complex amplitude $\hat{A}(x,t)$. The polarization of the medium is again written as

$$P(x,t) = \eta [u(x,t) + iv(x,t)]e^{i(kx-\omega L_0 t)} + c.c.$$

(2.42)

where $\eta$ is the atomic density and $u(x,t), v(x,t)$ are the real and imaginary parts of the slowly-varying single-atom polarization function. Using this notation, the wave equation for the propagation of the pulse in the slowly-varying envelope approximation becomes

$$\left( \frac{\partial}{\partial x} + \frac{1}{c} \frac{\partial}{\partial t} \right) \hat{A}(x,t) = i\frac{\mu_0\omega L_0 c}{2}\eta [u(x,t) + iv(x,t)].$$

(2.43)

Assuming the usual dipole interaction between a classical electric field and a quantized atom, the equation of motion for the elements of the atomic density matrix can be written as

$$\begin{align*}
\frac{\partial \rho_{11}}{\partial t} &= i(\Omega \sigma_{12}^{\ast} - \Omega^{\ast} \sigma_{12}) + \frac{\Gamma}{1 + |D|^2} \rho_{22}, \\
\frac{\partial \rho_{22}}{\partial t} &= -i(\Omega \sigma_{12}^{\ast} - \Omega^{\ast} \sigma_{12} + D\Omega \sigma_{32}^{\ast} - D^{\ast} \Omega^{\ast} \sigma_{32}) - \Gamma \rho_{22}, \\
\frac{\partial \rho_{33}}{\partial t} &= i(D\Omega \sigma_{32}^{\ast} - D^{\ast} \Omega^{\ast} \sigma_{32}) + \frac{\Gamma |D|^2}{1 + |D|^2} \rho_{22}, \\
\frac{\partial \sigma_{12}}{\partial t} &= -i\Delta \sigma_{12} + i\Omega (\rho_{22} - \rho_{11}) - iD\Omega \rho_{13} - \frac{\Gamma}{2} \sigma_{12}, \\
\frac{\partial \sigma_{32}}{\partial t} &= -i(\Delta + \omega_{31}) \sigma_{32} + iD\omega (\rho_{22} - \rho_{33}) - i\Omega \rho_{13}^{\ast} - \frac{\Gamma}{2} \sigma_{32}, \\
\frac{\partial \rho_{13}}{\partial t} &= -i\omega_{31} \rho_{13} + i(\Omega \sigma_{32}^{\ast} - D^{\ast} \Omega^{\ast} \sigma_{12}) - \frac{\Gamma |D|^2}{1 + |D|^2} \rho_{13}.
\end{align*}$$

(2.44)

where we have defined $\Omega = \hat{A}^{\ast}d_{12}/\hbar$ the complex Rabi frequency with $d_{12}$ being the dipole matrix element between states $|1\rangle$ and $|2\rangle$. $\Delta = \omega_{L0} - \omega_{12}$ is the detuning of the laser from the $|1\rangle \rightarrow |2\rangle$ transition of the atom, $\omega_{31} = \omega_{31} - \omega_{12}$ the Raman frequency, $D = d_{32}/d_{12}$ the ratio of the dipole matrix elements, and $\Gamma = 1/\tau_{sp}$ the inverse lifetime of the excited state. We have also defined $\sigma_{12} = \rho_{12}e^{i(kx-\omega L_0 t)}$ and $\sigma_{32} = \rho_{32}e^{i(kx-\omega L_0 t)}$. To calculate $u(x,t), v(x,t)$ that yield the polarization
of the medium needed in the wave equation, we have to average over the inhomogeneous line shape function \( g(\Delta) \):

\[
\begin{align*}
\psi(x,t) &= \int \Re [\sigma_{21}(\Delta)d_{12} + \sigma_{23}(\Delta)d_{32}] g(\Delta)d\Delta \\
\chi(x,t) &= \int \Im [\sigma_{21}(\Delta)d_{12} + \sigma_{23}(\Delta)d_{32}] g(\Delta)d\Delta.
\end{align*}
\] (2.45)

The inhomogeneous line shape function is assumed to be Gaussian, \( g(\Delta) = \exp \left[ -\left( \Delta + \Delta_0 \right)^2 / 2\sigma_\Delta^2 \right] / \sqrt{2\pi\sigma_\Delta^2}, \)

with a width \( \sigma_\Delta \) and a possible offset between the central frequency and the laser \( \Delta_0 \), taken hereafter to be zero. In the limit where \( \Gamma \gg \sigma_\Delta \) (the case when homogeneous broadening dominates) \( \psi(x,t), \chi(x,t) \) are given simply by

\[
\psi(x,t) + i\chi(x,t) = \sigma_{21}(0)d_{12} + \sigma_{23}(0)d_{32}.
\] (2.47)

As in Sec. 2.2.1, we then introduce the retarded time \( t' = t - x/c \) and assume Gaussian pulses with a linear frequency chirp \( \Omega(t') = \Omega_0 \exp(-i\beta t'^2) \exp(-t'^2/(2\tau_p^2)). \) Note that the frequency sweep rate is thus \( 2\beta \). It is convenient to introduce dimensionless space and time coordinates with \( \tau = t'/\tau_p \) and \( \xi = x/\xi_0 \)

where

\[
\xi_0 = \frac{\epsilon_0 hc}{\eta \omega L_0 |d_{12}|^2 T^*}
\] (2.48)

is the so-called absorption length which contains the dephasing time \( T^* \) of the atoms and their spatial density \( \eta \). In the limit where the inhomogeneous broadening of the transition is dominant \( T^* \) is connected with the width of the inhomogeneous lineshape function as \( T^* = T_{ih}^* = \pi g(0) = \sqrt{\pi/2\sigma_\Delta^2}. \) On the other hand, when inhomogeneous broadening can be neglected, \( T^* = T_h^* = \tau_{spont} = 1/\Gamma. \)

Using these units, the wave equation (2.43) becomes

\[
\frac{\partial \Omega}{\partial \xi} = -i \frac{1}{2\tau_{ih}^*} \int [\sigma_{12}(\Delta) + D^*\sigma_{32}(\Delta)] g(\Delta)d\Delta
\] (2.49)

if the inhomogeneous broadening dominates or

\[
\frac{\partial \Omega}{\partial \xi} = -i \frac{1}{2\tau_h^*} (\sigma_{12} + D^*\sigma_{32})
\] (2.50)

if the homogeneous broadening is dominant. Here, \( \tau_{ih}^* = T_{ih}^*/\tau_p \) is the dimensionless relaxation time in the homogeneous and inhomogeneous case, respectively.

Equation (2.49) or (2.50) for the propagation of the light pulse together with (2.44) constitute the starting point for our investigations. Note that the equations of (2.44) are unchanged by introducing a retarded time and transforming to dimensionless quantities, only \( t \) has to be replaced by \( \tau \). The equations can be solved numerically to explore the behaviour of the interaction between the propagating pulses and the atomic medium.

To treat the two-level atom case, they can easily be reduced to the usual Maxwell-Bloch equations
by substituting $D = 0$ (i.e., setting $d_{32} = 0$) and discarding the superfluous equations for $\rho_{33}$, $\sigma_{32}$ and $\rho_{13}$ from (2.44).

It is clear from (2.44) and (2.49) or (2.50) that in general the medium will affect the pulses propagating through it due to the induced polarization. The most obvious process is the absorption of pulse energy by the atoms, which, in turn, is lost due to spontaneous emission; but even if the pulses are much shorter than the natural lifetime of the excited state, i.e., $\tau_p \ll \tau_{\text{spont}}$, the induced polarization may considerably distort the pulses during propagation. In two-level media, the propagation of resonant pulses with constant carrier frequency, having a sech envelope, i.e.

$$A(t) = A_0 \frac{2}{e^{t/\tau_p} + e^{-t/\tau_p}}$$

Eq. (2.51)

where $A_0$ and $\tau_p$ are adjustable parameters so that the envelope area of the Rabi frequency is $2\pi$, is a notable exception to this provided the pulses are also short enough so that effects of spontaneous emission are negligible. In this case, even though the atoms are excited during the interaction and the medium thus has strong polarization, the pulse travels undistorted. The reason for this is that by the end of the interaction the atomic population is returned to the ground state and the energy absorbed by the atoms from the front of the pulse is coherently re-emitted into the tail, so that the shape and amplitude of the pulses do not change. The propagation will, however, be slowed down drastically. This effect is the well-known self-induced transparency (SIT, see [33]).

Clearly, such a mechanism cannot work for a frequency-chirped laser pulse that excites two-level atoms via RAP (rapid adiabatic passage, see [29]), since the atoms are transferred to the excited state at the end of the process. In this way, photons are continuously absorbed from the pulse and, correspondingly, its ability to transfer the atomic population to the excited state is expected to decrease gradually. Furthermore, the pulses that are capable of inducing RAP in two-level media always have a fairly large intensity. Even though a pulse area analogous to the one defined for resonant, transform-limited pulses is not really meaningful for a frequency-chirped pulse, the integral of its envelope function corresponds to a resonant pulse area of many times $2\pi$. It is well known that resonant pulses with a large area will, in general, reshape and break up into smaller pulses having areas of $2\pi$ each [71].

### Population transfer by a single frequency-chirped pulse

In Sec. 2.1, we saw that a single frequency-chirped laser pulse can, under certain conditions, transfer the atomic population of a $\Lambda$-atom from one stable state to the other via adiabatic population transfer. According to [39, 40], to accomplish a population transfer in a two-level (from ground state to excited state) or three-level (from one ground state to the other) atom using a frequency-chirped laser pulse, one obtains the requirements discussed in the following.

First, the time integral of the pulse envelope (the Rabi frequency) should be at least $(40 - 50)\pi$ and, second, the range of the frequency chirp should be on the same order of magnitude as the largest Rabi frequency, $\beta \times 5\tau_p \approx \Omega_0$. In addition to this, for a three-level $\Lambda$-atom the energy separation $\hbar \omega_{13}$
between the two stable states must be large compared to the transform-limited bandwidth of the pulse but smaller than the overall bandwidth covered by the frequency chirp. Furthermore, the chirp direction must be such that the laser pulse becomes resonant with the initially populated transition first. Note that this latter requirement, corresponding to an "intuitive" order of resonances, is contrary to the STIRAP scheme where a counter-intuitive pulse sequence is required.

In our case $\omega_{12} > \omega_{32}$ so, if the atom is initially in state $|1\rangle$, the sign of the chirp must be positive so that the population transfer takes place (see Fig. 2.1). Note that the first condition that follows from the adiabatic theorem is in fact a requirement for the pulse area rather than directly for the intensity. As the first and second requirements are pairwise the same for the case of two- and three level atoms, it is clear that, under appropriate conditions, the very same pulse that transfers the atomic population to the excited state $|2\rangle$ in the two-level atom, will bring it to $|3\rangle$ in a $\Lambda$-atom.

Before discussing the propagation itself, we first looked at the population transfer the pulse produces at the entrance of the medium ($x = 0$) in the two-level as well as in the three-level case. Fig. 2.4 shows the time evolution of the atomic populations $\rho_{11}$, $\rho_{22}$, $\rho_{33}$ for a three-level $\Lambda$-atom and $\rho_{11}$, $\rho_{22}$ for a two-level atom interacting with a frequency-chirped laser pulse. The data was obtained from the numerical solution of (2.44) using the following parameters: we set the amplitude of the pulse $\Omega_0 = 50/\tau_p$, the chirp parameter $\beta = 10/\tau_p^2$, the separation of the two lower states of the $\Lambda$-atom $\omega_{13} = 7/\tau_p$ and $\Gamma = 0.025/\tau_p$ which corresponds to $T_{h}^* = 40\tau_p$ so the effect of spontaneous emission is relatively small during the interaction. These pulse parameters fulfill the conditions for adiabatic population transfer mentioned in the previous paragraph and the figures show that the same pulse indeed induces adiabatic population transfer in both cases. Note that the transfer is not perfect (around 95%) due to the finite lifetime of the excited state and the finite pulse length.

Comparing Fig. 2.4 with Fig. 2.3 one sees an apparent correspondence in the population dynamics. However, as a difference, we observe quick, low-amplitude oscillations between the populations of the ground states during the interaction in contrast to the case of the adiabatic approximation where (due to the smooth behaviour of the quasienergy $w_1$) the population was transferred from state $|1\rangle$ to state $|3\rangle$ without any oscillations. This additional feature is due to the presence of the non-adiabatic term on the LHS of (2.9), proportional to the time derivative of the eigenvectors of the Hamiltonian. In the dressed state analysis we made the adiabatic approximation by neglecting this term.

From the point of view of transfer efficiency, it is not crucial that the pulse amplitude and chirp be precisely the numerical values represented here. As adiabatic processes are "robust", a slight variation in the pulse parameters does not compromise the efficiency. In fact, for the present value of the amplitude, taking a chirp rate parameter as low as $\beta = 5/\tau_p^2$ or as high as $\beta = 50/\tau_p^2$ still yields a pulse that adiabatically transfers the atomic population. It is also apparent from Fig. 2.4(a) that during the population transfer induced in the $\Lambda$-atoms the excited state is only slightly populated. This property is similar to what one has for the population transfer induced by a pair of laser pulses in the STIRAP scheme. However, there is one major difference: while for STIRAP, the dressed state that must be adiabatically followed in order to transfer the population is composed solely of
2.2. PROPAGATION OF FREQUENCY-CHIRPED LASER PULLSES AMONG Λ-ATOMS

|1⟩ and |3⟩ and is thus "perfectly dark", this is not the case for the chirped-pulse process. In Sec. 2.1 we have seen that in this case the dressed state responsible for the population transfer also contains the excited state |2⟩ with a certain small amplitude. Even though this amplitude can be efficiently suppressed by increasing the pulse intensity, it can never be completely eliminated. So, the population transfer by a chirped pulse might be slightly degraded by incoherent effects involving the excited state even in the perfectly adiabatic limit.

Figure 2.4: (a) Time evolution of the atomic population of a Λ-atom during the interaction with the frequency-chirped laser pulse entering the medium. The population is transferred from state |1⟩ to |3⟩. The dimensionless parameters used for the solution of (2.44) are Ω₀ = 50, β = 10, ω₁₃ = 7 and Γ = 0.025. (b) Time evolution of the population of a two-level atom interacting with the same pulse. The atom is excited via RAP.
Pulse propagation in homogeneously broadened media

The major difference in the effect of the chirped laser pulse on time evolution of the atomic populations in the two- and three-level cases is clearly expected to also show in the propagation of the laser pulse. In the $\Lambda$-atom case the atom is no longer left in the excited state after the interaction but it is transferred to another stable state whose energy separation from the initial state $|1\rangle$ is much smaller than that of the excited state (see Fig. 2.1). Furthermore, since polarization of the medium is associated with populating the excited state (there is no dipole transition between states $|1\rangle$ and $|3\rangle$) the back-action of the $\Lambda$-atoms on the pulse will be much weaker during propagation than that of the two-level atoms.

![Figure 2.5](image)

Figure 2.5: Evolution of the magnitude of the frequency-chirped laser pulse envelope $|\tilde{A}(\xi, \tau)|$ in space and time while propagating through a medium of $\Lambda$-atoms. The dimensionless distance $\xi$ is measured in units of the absorption length, the dimensionless time $\tau$ is measured in units of the pulse width. The parameters used for the numerics are $\Omega_0 = 50$, $\beta = 10$, $\omega_{13} = 7$ and $\Gamma = 0.025 \rightarrow \tau_h^* = 40$.

Fig. 2.5 shows the evolution of the magnitude of the laser pulse envelope $|\tilde{A}(\xi, \tau)|$ as the frequency-chirped laser pulse propagates through a medium of three-level atoms with a $\Lambda$ level scheme. The data for the plots was obtained by solving (2.44) and (2.50) numerically with initial pulse parameters identical to those used to produce Fig. 2.4. Apparently, the modulations of the pulse envelope induced by the atoms are not substantial. It is also clear from the figure that the most important effect on the pulse is the modulation, the distortion of the pulse envelope as it propagates inside the medium, and not the attenuation of the pulse due to absorption. This is in accordance with the analytic results in Sec. 2.2.1 namely, that - in adiabatic approximation - the pulse amplitude does not suffer any changes while propagating in the medium.
More important from a practical point of view is the ability of the pulse to produce adiabatic population transfer between the atomic levels. Fig. 2.6 shows the final level populations of (a) a Λ-atom and (b) a two-level atom after the passing of the laser pulse, as a function of the dimensionless penetration depth $\xi$. In the case of two-level atoms, final population of the excited state should be close to 1 if the population transfer is adiabatic and, correspondingly, a final population of $|3\rangle$ close to 1 has the same meaning for the Λ-atom. Note that, at the point of entering the media, the pulses
do perform adiabatic population transfer to a very good measure, as seen in Fig. 2.4. It is evident from Fig. 2.6 (a) that the population transfer induced by the pulse in the medium of \( \Lambda \)-atoms is unchanged until about \( \xi = 8000 \), i.e., 8000 times the absorption length defined by the parameters of the medium. After this, the capability of the pulse to perform adiabatic population transfer starts to gradually degrade and is lost relatively quickly afterwards. The reason for this behaviour can be deduced from Fig. 2.7 where the time evolution of the atomic populations of a \( \Lambda \)-atom has been plotted at \( \xi = 10000 \). While population transfer between the two lower states still works quite well, the peak population of the excited state during the interaction has increased several times over the initial peak value seen in Fig. 2.4. As the pulse propagates, the maximal excited state population during interaction increases gradually.

![Figure 2.7: Time evolution of the level populations of a \( \Lambda \)-atom during the interaction with the laser pulse at \( \xi = 10000 \) in the medium.](image)

As this happens, polarization of the medium that distorts the pulse also grows. Since the back-action on the pulse increases, the degradation of the pulse’s capability to induce adiabatic population transfer speeds up.

In some sense, the enhanced transparency of the medium due to the coupling to a third atomic state is reminiscent of EIT where the coupling to a third state renders the medium transparent to the probe beam. Of course, one major difference is that in our case there is no additional field to realize the coupling: the frequency-chirped laser pulse itself couples the excited state with the third state, increasing the transparency of the medium for itself.

However, it is obvious that there are other important differences too compared to EIT. As seen in Sec. 2.2.1 separating the complex pulse envelope into real amplitude and phase functions as \( \tilde{A}(\xi, \tau) = A(\xi, \tau) \exp[i\phi(\xi, \tau)] \) it is possible to show that the real amplitude is decoupled from the
medium when the population transfer is perfectly adiabatic. This is despite the fact that the excited state of the atom is populated during the population transfer even in this limit. On the other hand, the phase function that describes the chirp is distorted due to the back-action of the medium through polarization, and the amplitude is then compromised because the population transfer ceases to be perfectly adiabatic.

Therefore, it is clear that although its propagation length in a medium of Λ-atoms is greatly enhanced, there is no complete transparency for the frequency-chirped laser pulse even in the adiabatic approximation, unlike, for instance, to the sech pulses of SIT or the probe pulse of EIT or matched pulse pairs [38, 34]. Another difference compared to SIT and EIT, also visible in Fig. 2.5, is the fact that the peak of the pulse remains at τ = 0 in the retarded time coordinate, i.e., there is no reduction of the group velocity as in SIT or EIT.

It is interesting to note that in practice, pulse propagation in the sharp-line limit is often explored using picosecond pulses [72]. However, in that case the two-level approximation fails and the true multilevel nature of the atoms must be taken into account. Since the coupling constants for the various transitions are different, their contributions to the polarization will soon get out of phase and thus pulse distortion and pulse breakup will be much smaller than that calculated from a simple two-level model. Such a mechanism is no doubt at work for frequency-chirped laser pulses when traveling in a medium of multilevel atoms.

Pulse propagation in inhomogeneously broadened media

One of the primary reasons why frequency-chirped laser pulses are convenient for the manipulation of atomic quantum states is the ability to transfer the atomic populations even when there is a fairly large inhomogeneous broadening of the transition line. In the case of population transfer between the lower states of a Λ-atom, this broadening can be tolerated as long as it is considerably smaller than the total bandwidth of the laser pulse. Atoms with a transition frequency farther away from the center of the inhomogeneously broadened spectrum than that experience an interaction that does not fulfill the criteria for an adiabatic process, and therefore the population transfer will be imperfect. In case the broadening is very large, the pulse simply does not interact with a certain part of the atoms: they remain in state |1⟩.

To investigate the effect of inhomogeneous broadening of the atomic transition on the propagation of a frequency-chirped laser pulse, we solved (2.44) together with (2.49). For the width of the inhomogeneous broadening, we used σΔ = 10/τp which corresponds to a dephasing time τih* = 0.1253. The pulse parameters were the same as in the previous section. With these parameters, we are in a regime where the pulse is capable of inducing adiabatic population transfer over the whole atomic ensemble. To separate the effects of spontaneous emission from the dephasing due to inhomogeneous broadening, we assume Γ = 0 for now.

The evolution of the pulse amplitude |A(ξ, τ)| is shown in Fig. 2.8 as the pulse propagates in the medium of Λ-atoms. Again, the amplitude is plotted as a function of the dimensionless distance ξ. However, since the dephasing time has decreased a great deal (from $T_h^* = 40τ_p$ in the homogeneous
CHAPTER 2. MANIPULATION OF ATOMIC POPULATION BY FREQUENCY-CHIRPED LASER PULSES

case to $T_{th}^* = 0.1253\tau_p$ in the present case), the unit of distance has also changed compared to that obtained in the previous subsection where we dealt with propagation in homogeneously broadened media. Using the definition (2.48) for the absorption length, we have

$$\xi^h T_{th}^* = \xi^{ih} T_{ih}^*$$

(2.52)

where the superscript in $\xi^{h,ih}$ refers to substituting $T_{h,ih}^*$ into (2.48) as dephasing time. The real physical distance can be expressed with the dimensionless distance $\xi^{h,ih}$ for the homogeneous and inhomogeneous case, respectively, as

$$x = \xi^h \xi_0 = \xi^{ih} \xi_0$$

(2.53)

Thus, to compare Figs. 2.5 and 2.8 one must note that dimensionless distance $\xi^h = 10000$ in the homogeneously broadened medium corresponds to the same physical distance $x$ as $\xi^{ih} = 31$ dimensionless distance in the inhomogeneously broadened case (provided all the other parameters characterizing the atomic ensemble are the same). Examining Figs. 2.5 and 2.8 one can see that the distortion of the pulse amplitude is somewhat less at roughly double the distance in the inhomogeneous case compared to the homogeneous one.

Figure 2.8: The evolution of the magnitude of the frequency-chirped laser pulse envelope $|\tilde{A}(\xi, \tau)|$ in space and time while propagating through a medium of $\Lambda$-atoms with inhomogeneous broadening. The dimensionless distance $\xi$ is measured in units of absorption length, the dimensionless time $\tau$ is measured in units of the pulse width. The parameters used for the numerical calculations are $\Omega_0 = 50$, $\beta = 10$, $\omega_{13} = 7$ and $\sigma_\Delta = 10 \rightarrow \tau_{ih}^* = 0.1253$. 
This is also evident if one compares Figs. 2.6 and 2.9 showing the final populations of the atomic levels in the two cases as a function of penetration depth. During pulse propagation, inhomogeneous broadening has an important consequence: it provides a mechanism for the decay of the polarization of the medium through the dephasing of the individual atomic dipoles and thus decreases the back-action of the medium on the pulse. Therefore, a limited amount of inhomogeneous broadening tends to improve the propagation properties of the pulses. Thus, the mechanism at work functions well also in the inhomogeneously broadened three-level $\Lambda$-medium.

Figure 2.9: Final populations of the atomic levels after the passing of the pulse as a function of the penetration depth for (a) the medium of three-level $\Lambda$-atoms and (b) the medium of two-level atoms.
It is also interesting to note that expecting a strong degradation of this transparency is quite logical in case the inhomogeneous broadening of the medium is greater than the full bandwidth of the frequency-chirped laser pulse. This is because then a portion of the atoms will not experience adiabatic population transfer but will partly end up in the excited state which means that the medium will get polarized. However, numerical solution of the equations shows that this degradation of the transparency of a medium of $\Lambda$-atoms is much smaller than expected: only a small fraction of the atoms have a resonance frequency that is far enough from the center of the inhomogeneous broadening to experience an imperfect population transfer but close enough to be still considerably affected by the pulse. Therefore, the frequency-chirped pulse will travel undistorted almost to the same extent as before, only the induced overall population transfer will be imperfect due to the large inhomogeneous broadening.

2.3 Creating a superposition of quantum states in tripod atoms

If, instead of aiming for a complete population transfer, one wants to create a coherent superposition of states, a single, frequency-chirped laser pulse is appropriate for the purpose if (in contrast with the case when transferring the whole population) the transform-limited bandwidth of the pulse covers the frequency interval between the two target states for the superposition. One may use a $\Lambda$-atom to create a superposition of the two ground states. However, this is accompanied with the substantial excitation of the atom ([7, 8, 9]).

However, this problem can be eliminated if, instead of a three-level $\Lambda$-atom, one applies the chirped pulse to a four-level atom with a tripod level structure. Again, a major advantage of using frequency-chirped pulses for creating superpositions of magnetic sublevels in a tripod atomic level structure is that, in contrast with other methods involving non-chirped lasers where more than one field is necessary to perform the required process, here only a single field is required. Using frequency-chirped pulses, the problem of pulse synchronization, which in an extended medium might be quite an issue, is eliminated. Also, because of the varying carrier frequency, the detrimental effects of inhomogeneous broadening can be overcome with this method.

2.3.1 Theoretical framework

Throughout the following discussion ([73]), we will assume that the duration of the laser pulse interacting with the atom is much shorter than any relaxation time of the atomic system, which allows us to deal with the Schrödinger equation for the probability amplitudes of the quantum states of the atom instead of using a density matrix approach.

The interaction Hamiltonian of a classical electromagnetic field with a tripod linkage level system (see Fig. 2.10) in dipole and rotating-wave approximation assumes the form

$$\hat{H}_I = -E(t, z) (d_{10} |0\rangle\langle1| + d_{20} |0\rangle\langle2| + d_{30} |0\rangle\langle3|) + \text{H.c.}$$  \hspace{1cm} (2.54)
2.3. Creating a Superposition of Quantum States in Tripod Atoms

Figure 2.10: Levels of the four-level tripod-linkage atom. \( |1\rangle \) and \( |3\rangle \) are the "close" ground states (i.e., \( \Delta \omega_{31} \) is smaller than the transform-limited bandwidth of the laser pulse), \( |2\rangle \) is the "far" ground state and \( |0\rangle \) is the excited state. Dipole transitions are allowed only between ground and excited states, not between two ground states.

The classical electric field \( E(t, z) \) interacts with all transitions, where \( d_{ij} \) are the transition dipole matrix elements between states \( |i\rangle \) and \( |j\rangle \), and it can be written as

\[
E(t, z) = \frac{1}{2} A(t)e^{i\Phi(t) + i(\omega_{L0}t - kz)}. \tag{2.55}
\]

Here, \( A(t) \) is the (real-valued) envelope of the laser pulse and the carrier frequency is equal to \( \omega_L = \omega_{L0} + \dot{\Phi} \). In the following, we assume a linear modulation in time for the laser carrier frequency: \( \Phi(t) = \beta t^2 / 2 \) and so

\[
\omega_L(t) = \omega_{L0} + \beta t \tag{2.56}
\]

with \( \omega_{L0} \) being the central frequency and \( \beta \) the speed of chirp. For the linear chirp, the time-dependent detuning of the laser pulse from the resonance frequency of the transition \( |1\rangle - |0\rangle \) equals

\[
\epsilon_1'(t) = \omega_L(t) - \omega_{10} = \omega_{L0} - \omega_{10} + \beta t. \tag{2.57}
\]

Expressing the (bare) states of the system with column vectors as

\[
\begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \end{pmatrix} = |0\rangle, \quad \begin{pmatrix} 0 \\ 1 \\ 0 \\ 0 \end{pmatrix} = |1\rangle, \quad \begin{pmatrix} 0 \\ 0 \\ 1 \\ 0 \end{pmatrix} = |2\rangle, \quad \begin{pmatrix} 0 \\ 0 \\ 0 \\ 1 \end{pmatrix} = |3\rangle, \tag{2.58}
\]
CHAPTER 2. MANIPULATION OF ATOMIC POPULATION BY FREQUENCY-CHIRPED LASER PULSES

the Schrödinger equation for the components of the atomic state vector

\[ a = \begin{pmatrix} a_0 \\ a_1 \\ a_2 \\ a_3 \end{pmatrix} \]  (2.59)

has the form

\[ \dot{a}_0 - ia_0 \epsilon_1(t) = \frac{i}{2} [a_1' \Omega_{10} + a_2' \Omega_{20} + a_3' \Omega_{30}] \]
\[ \dot{a}_1' = \frac{i}{2} a_0' \Omega_{01} \]
\[ \dot{a}_2' + i\Delta \omega_{21} a_2' = \frac{i}{2} a_0' \Omega_{02} \]
\[ \dot{a}_3' + i\Delta \omega_{31} a_3' = \frac{i}{2} a_0' \Omega_{03} \]  (2.60)

where the probability amplitudes \( a_j' \) are phase-transformed variants of the original \( \{a_j\}_{j=0,1,2,3} \) amplitudes:

\[ a_0 = a_0' e^{i \epsilon_1'(t) t} \]
\[ a_1 = a_1' \]
\[ a_2 = a_2' e^{i \Delta \omega_{21} t} \]
\[ a_3 = a_3' e^{i \Delta \omega_{31} t} \]  (2.61)

the time-dependent detuning is

\[ \epsilon_1(t) = \epsilon_1'(t) + t \frac{d \epsilon_1}{dt} = \omega_{L0} - \omega_{10} + 2 \beta t \]  (2.62)

and the real-valued Rabi frequencies are \( \Omega_{ij} = \Omega_{ij}^* = A(t)/d_{ij}, i,j=0,1,2,3. \) The Raman detunings \( \Delta \omega_{21} \) and \( \Delta \omega_{31} \) are equal to the angular frequency intervals between the corresponding ground states, as seen in 2.10.

The set of equations (2.60) have been discussed previously in a number of papers dealing with the generation of coherent superpositions of ground states [2, 3, 4], with further applications in constructing coherent beam splitters for atomic beams [74] and in adiabatic pulse propagation [75]. The condition of two-photon (Raman) resonance is an important condition to be fulfilled for these schemes to be efficient. This is one of the reasons why at least two narrowband laser pulses have been applied to generate the coherent superposition of ground states in the previous investigations.

In contrast with these approaches, for a single chirped laser pulse we assumed a simultaneous interaction of the field with all of the allowed transitions in the atom. As we have seen above, the condition of Raman resonance cannot be fulfilled in this situation: the corresponding Raman detunings are constants.
First, we analyze the population dynamics of the states of the tripod linkage atom during action of the frequency-chirped laser pulse having a Gaussian envelope function, by means of numerical solution of (2.60). The results are presented in Figs. 2.11 and 2.12 for the case when the atom is initially in one of its ground states.

![Figure 2.11: Population dynamics of the states of the tripod-linkage atom during the action of the frequency-chirped pulse, as a function of time. $n_i$ denotes the population of state $|i\rangle$ where $i = 0, 1, 2, 3$. Throughout the interaction, the excited state $|0\rangle$ is negligibly populated. Parameters applied are duration of the pulse $\tau_p = 15\, ns$, Rabi frequencies for transitions from the metastable states to the excited state (the dipole moments for the allowed transitions are assumed equal to each other) are $\Omega_{10} = \Omega_{20} = \Omega_{30} = 15\, GHz$ and the speed of the frequency chirp is $\beta = 3\, GHz/\, ns$. The frequency distance $\Delta \omega_{31}$ between the close levels is equal to $10\, MHz$ and the distance between the close and the far levels is equal to $1\, GHz$.](image1)

![Figure 2.12: Time evolution of the phase $\phi$ of the Raman coherence between ground states $|1\rangle$ and $|3\rangle$ in units of $\pi$, using the same interaction parameters as those in Fig. 2.11.](image2)
In the simulations, for simplicity, we assumed that the Rabi frequencies are the same for all allowed transitions in the atom: \( \Omega_{01} = \Omega_{02} = \Omega_{03} \). So, the Rabi frequency of the pulse is

\[
\Omega_{01}(t) = \Omega_0 e^{-\frac{t^2}{2\tau_p^2}},
\]

with \( 2\tau_p \) being the duration of the pulse. The time-dependent detuning \( \epsilon_1(t) \) contains a linear chirp as shown in (2.62). According to Fig. 2.11, part of the atomic population is transferred from the initially populated state \( |2\rangle \) and distributed among the three ground states as a result of the interaction with the frequency-chirped laser pulse. An important characteristic of the process is that only a negligibly small amount of population is temporarily transferred to the excited state \( |0\rangle \) of the atom during the interaction. Such a behaviour is possible only if the bandwidth \( \Delta \omega_L \) of the laser pulse envelope (without chirp) is larger than the frequency separation between the two "closer lying" ground states of the atom, i.e., \( |1\rangle \) and \( |3\rangle \), but smaller than the separation between the "closer lying" states and the ground state \( |2\rangle \) (see 2.10):

\[
|\Delta \omega_{13}| \ll \Delta \omega_L < |\Delta \omega_{12}|.
\]

Additionally, to suppress excitation of the system during the interaction, as discussed in Sec. 2.1, one has to set the amplitude of the Rabi frequency much larger than the frequency difference between the ground states of the \( \Lambda \)-atom. Applying this condition to the current case, we have

\[
\Omega_0 \gg |\Delta \omega_{12}|,
\]

i.e., the peak Rabi frequency \( \omega_0 \) of the laser pulse must be greater than the largest frequency distance between the ground states \( |d_b\rangle \) and \( |2\rangle \) of effective \( \Lambda \)-atom.

The sign of the chirp (\( \beta \)) also plays an important role: the pulse must get into resonance with the transition involving the initially populated ground state and the excited one and only afterwards with the other, initially empty transitions.

### 2.3.2 Analysis of the atomic population dynamics

In order to explain the population dynamics shown in Figs. 2.11 and 2.12, obtained by means of solving (2.60) numerically, in the following we show that the four-level atom with a tripod level structure may be mapped to a three-level \( \Lambda \)-atom if the condition (2.64) is fulfilled.

In this mapping, we perform a basis transformation yielding three basis states, coupled to the laser field, which can be mapped to an effective three-level \( \Lambda \) structure. The fourth (so-called dark) state of the new basis is entirely decoupled from the field. Thus, we can treat the remaining three states separately from the dark state.

After establishing the mapping to the \( \Lambda \)-system, we proceed analogously to the contents of Sec. 2.1, preforming a dressed state analysis on the system.
Since the time scale of the time evolution of the probability amplitudes is set by the pulse duration, condition (2.64), i.e. that \( \Delta \omega_{31} \) is much smaller than the transform-limited spectral width of the laser pulse, allows to neglect the term \( \Delta \omega_{31} a'_3 \) in the last equation of (2.60) compared to the term \( \dot{a}'_3 \):

\[
|\Delta \omega_{31}| \ll |\dot{a}'_3|.
\]

(2.66)

It is convenient to rewrite (2.60) in the basis of the "bright" \( |d_b\rangle \) and "dark" \( |d_d\rangle \) superposition of the ground states as

\[
|d_b\rangle = \frac{1}{\sqrt{1 + \eta^2_{31}}} (|1\rangle + \eta_{31}|3\rangle), \quad |d_d\rangle = \frac{1}{\sqrt{1 + \eta^2_{31}}} (\eta_{31}|1\rangle - |3\rangle)
\]

(2.67)

where \( \eta_{31} = \Omega_{03}/\Omega_{01} = d_{03}/d_{01} \). The probability amplitudes \( d_b \) and \( d_d \) belonging to these states can be expressed as

\[
d_b = \frac{1}{\sqrt{1 + \eta^2_{31}}} (a'_1 + \eta_{31}a'_3), \quad d_d = \frac{1}{\sqrt{1 + \eta^2_{31}}} (\eta_{31}a'_1 - a'_3).
\]

(2.68)

With the above considerations, the Schrödinger equation in this basis assumes the form

\[
\dot{d}_b = \frac{i}{2} a'_0 F_1
\]

\[
\dot{a}'_2 + i\Delta \omega_{21} a'_2 = \frac{i}{2} a'_0 F_2
\]

\[
\dot{d}_d = 0
\]

\[
\dot{a}'_0 - i a'_0 \epsilon_1(t) = \frac{i}{2} (d_b F^*_1 + a'_2 F^*_2)
\]

(2.69)

where \( F_1 = \Omega_{01} \sqrt{1 + \eta^2_{31}}, F_2 = \Omega_{02} \eta_{21} \) and \( \eta_{21} = \Omega_{02}/\Omega_{01} = d_{02}/d_{01} \). Apart from the equation containing the dark state amplitude, (2.69) are identical to the Schrödinger equations of a three-level atom with \( \Lambda \) linkage, where one of the ground states is replaced by \( |d_b\rangle \), the other one is \( |2\rangle \) and the excited state corresponds to \( |0\rangle \). As it follows from (2.69), the dark state probability amplitude stays constant throughout the interaction. Note that this behaviour was obtained when the term proportional to \( \Delta \omega_{31} \) in (2.60) was neglected. This term, if kept, would provide interaction between dark and bright components. However, the left-hand relation in (2.64) which may be rewritten as \( \Delta \omega_{31} \tau_p \ll 1 \) means that the evolution of the dark state is negligible under the relatively short duration of the interaction with the laser pulse. The result seen in Fig. 2.11 shows the behaviour in case of \( \Delta \omega_{31} \tau_p = 0.15 \) that slightly differs from the situation where \( \Delta \omega_{31} \tau_p = 0 \). For the latter case one expects the final population of the "far" ground state to be 0.5 and the populations of the "close" states to be the same and equal to 0.25 each, if we assume equal dipole transition elements.
The amplitudes $d_b, a'_0$ and $a'_2$ may be represented as components of a column state vector

$$
c = \begin{pmatrix} d_b \\ a'_0 \\ a'_2 \end{pmatrix}, \quad (2.70)
$$

in the basis

$$
\begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} = |d_b\rangle, \quad \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} = |0\rangle, \quad \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix} = |2\rangle, \quad (2.71)
$$

of the effective $\Lambda$-atom (see Fig. 2.13). As a matrix equation, (2.69) assumes the form

$$
i\hbar \dot{c} = \hat{H}_b c. \quad (2.72)
$$

Figure 2.13: Structure of the quantum states of the effective $\Lambda$-linkage atom with one of the ground states, $|d_b\rangle$, being a superposition of the "close" metastable states of the original tripod atom. Dipole transitions are only allowed between ground and excited states, i.e., $|d_b\rangle - |0\rangle$ and $|2\rangle - |0\rangle$.

In the basis (2.71), $\hat{H}_b$ is

$$
\hat{H}_b = -\hbar \begin{pmatrix} 0 & F_1/2 & 0 \\ F_1^*/2 & \epsilon_1(t) & F_2^*/2 \\ 0 & F_2 & -\Delta \omega_{21} \end{pmatrix}, \quad (2.73)
$$

So far, by transforming the initial basis into the dark-bright basis and requiring condition (2.64) be fulfilled we managed to reduce the four-level problem to that of a three-level $\Lambda$-atom, getting (2.69). We shall see in the following that it is worth to expand the solutions of (2.72) in the basis of
2.3. CREATING A SUPERPOSITION OF QUANTUM STATES IN TRIPOD ATOMS

the so-called adiabatic states, i.e., the eigenstates of \( \hat{H}_b \).

\[
c = \sum_{k=1}^{3} r_k(t) b^{(k)}(t) \exp \left[ -i \int_{-\infty}^{t} dt' w_k(t') \right]
\]  

(2.74)

where \( b^{(k)}(t) \) and \( w_k(t) \) are the \( k \)-th eigenvector and eigenvalue (quasienergy) of \( \hat{H}_b \), respectively, and the time-dependent expansion coefficients \( \{r_k(t)\}_{k=1,2,3} \) are the corresponding probability amplitudes.

\[
\hat{H}_b b^{(k)} = w_k b^{(k)}
\]  

(2.75)

According to the expansion (2.74), for initial conditions as \( t \to -\infty \) we have

\[
c(-\infty) = \sum_{k} r_k(-\infty) b^{(k)}(-\infty).
\]  

(2.76)

According to the adiabatic theorem ([69]), for adiabatic interactions the atomic system follows the adiabatic states during the whole of the interaction. In our case this means \( r_k(t) = r_k(-\infty) \).

We obtain the following equations for the quasienergies \( w_k \) and the eigenvector components \( b^{(k)} \):

\[
w^3 + w^2 (\Delta \omega_{21} - \epsilon_1) - w \left[ \epsilon_1 \Delta \omega_{21} + (|F_1|^2 + |F_2|^2) \right] - |F_1|^2 \Delta \omega_{21} = 0
\]  

(2.77)

\[
b_1^{(k)} = \frac{F_1(w_k + \Delta \omega_{21})}{\sqrt{N}}, \quad b_2^{(k)} = \frac{w_k(w_k + \Delta \omega_{21})}{\sqrt{N}}, \quad b_3^{(k)} = \frac{F_2 w_k}{\sqrt{N}}
\]  

(2.78)

with the normalization factor \( N = F_1^2 (w_k + \Delta \omega_{21})^2 + w_k^2 (w_k + \Delta \omega_{21})^2 + F_2^2 w_k^2 \) and, for the sake of simplicity, we have set the value of \( \hbar \) to 1.

The dynamics of the quasienergies \( \{w_k\}_{k=1,2,3} \), obtained from solving (2.77), is the same we got in Sec. 2.1 plotted in Fig. 2.2 for a laser pulse with a Gaussian envelope function \( A(t) \) and a linear, from red to blue chirp. The quasienergies are marked with continuous lines on the figure. As defined in Sec. 2.3.1 the chirped frequency and the corresponding detuning are

\[
\omega_L = \omega_{L0} + \beta t
\]  

(2.79)

\[
\epsilon_1(t) = \omega_{L0} - \omega_{10} + 2\beta t
\]  

(2.80)

\( \beta > 0 \).

The eigenvalue functions for zero Rabi frequencies are called diabatic lines and are marked with dashed lines in Fig. 2.2. According to the figure, the values of one of the quasienergies, referred to as \( w_1 \), are restricted between zero and the value of the angular frequency difference between the ground states of the \( \Lambda \)-atom, which in this case is \( \Delta \omega_{21} \), between ground states \( |d_b \rangle \) and \( |2 \rangle \) of the equivalent \( \Lambda \) linkage atom:

\[
0 \leq w_1 \leq |\Delta \omega_{21}|.
\]  

(2.81)
Let us assume at this point that the atom is in the ground state $|d\rangle$ at $t \to -\infty$, which means

$$c(t \to -\infty) = \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}. \quad (2.82)$$

It is apparent from (2.77) and (2.78) that the only eigenvector that equals $(1, 0, 0)^T$ as $t \to -\infty$ is $b^{(1)}(t \to -\infty)$, i.e., the one associated with $w_1$. Thus, we can identify the initial state of the system as

$$c(t \to -\infty) = b^{(1)}(t \to -\infty). \quad (2.83)$$

According to the adiabatic theorem ([69]), the state vector of the atom will follow this adiabatic state if the conditions of adiabaticity are fulfilled (see [1]). In the case under consideration, the envelope of the laser pulse must be sufficiently long so that its bandwidth is smaller than the frequency interval $\Delta \omega_{21}$ between the ground states $|d\rangle$ and $|2\rangle$, as seen in (2.64).

(2.78) yields the following relations between the components of the adiabatic state vectors:

$$\frac{b^{(k)}}{b^{(1)}} = \frac{w_k}{F_1}, \quad k = 1, 2, 3. \quad (2.84)$$

$$\frac{b^{(2)}}{b^{(1)}} = \frac{F_2 w_k}{F_1 (w_k - \Delta \omega_{21})}, \quad k = 1, 2, 3. \quad (2.85)$$

Because the system follows the adiabatic state $b^{(1)}$, the component $b^{(1)}_2$ alone accounts for the contribution of the excited state $|0\rangle$ during the interaction. Due to the limiting values for $w_1$ given in (2.81), there exists an upper limit also for the relative contribution of the excited state:

$$\left| \frac{b^{(2)}}{b^{(1)}} \right| \leq \frac{\Delta \omega_{21}}{F_1}. \quad (2.86)$$

This contribution can be made negligibly small if the Rabi frequency considerably exceeds the frequency distance between the two ground states, i.e., $F_1 \gg \Delta \omega_{21}$. The analysis of (2.77) for sufficiently large Rabi frequencies, that is, when

$$|F_1|^2 + |F_2|^2 \gg |\epsilon_1 \Delta \omega_{21}| \quad (2.87)$$

$$|F_1| \gg |\Delta \omega_{21}| \quad (2.88)$$

shows that $w_1$ does not depend on the Rabi frequency and can be approximated as

$$w_1 \approx -\frac{d_{01}^2 + d_{03}^2}{d_{01}^2 + d_{02}^2 + d_{03}^2} \Delta \omega_{21}. \quad (2.89)$$

In the region where the quasienergy $w_1$ is constant (see Fig. 2.2), we have a "trapped" super-
position of the two ground states of the Λ-atom: the bright state $|d_b\rangle$ and the ground state $|2\rangle$ of the original tripod-linkage atom. Excitation of the atom in this region is strongly suppressed. For a positive chirp ($\beta > 0$), at the end of the interaction the quasienergy $w_1$ tends to the diabatic line $w_0^0 = -\Delta \omega_{21}$. For $\beta < 0$, it would tend to the diabatic curve $w_2^0 = \epsilon_1(t)$. This latter eigenvalue is related to the excited state of the equivalent Λ-atom which is the same as the excited state $|0\rangle$ of the tripod-linkage atom.

In the adiabatic states representation it becomes apparent that it is possible to transfer the complete population of the superposition state $|d_b\rangle$ into the ground state $|2\rangle$ if the sign of the chirp is chosen appropriately. For the states of the original tripod-linkage atom this means that the population of the bright superposition of the "close" ground states $|1\rangle$ and $|3\rangle$ is transferred into the "far" ground state $|2\rangle$ without considerable excitation of the atom while the dark superposition of the "close" states remains unaffected during the process. As a result, after the interaction we are left with the dark superposition

$$|d_d\rangle = \frac{1}{\sqrt{1 + \eta_{31}^2}} (\eta_{31}|1\rangle - |3\rangle),$$

(2.90)

with $\eta_{31} = d_{03}/d_{01}$, $d_{ij}$ being the dipole matrix element of the transition $|i\rangle - |j\rangle$ in the tripod-linkage atom.

**2.3.3 Applications of the processes**

As mentioned in the Introduction, superpositions of quantum states play an important role in numerous applications in quantum and nonlinear optics as well as in other, related fields. In the scheme proposed above, a single frequency-chirped laser pulse transfers the so-called bright superposition component of the two "close" ground states (their frequency separation is less than the transform-limited bandwidth of the pulse) into the third, "far" ground state of the tripod linkage atom, leaving the "close" ground states in their dark superposition. During the transfer process, the excitation of the atom is negligibly small.

Because no strict resonance conditions are required for the laser pulse, this method may be efficiently applied also to inhomogeneously broadened media. The only limitation on the width $\Delta \omega_{inh}$ of the inhomogeneous broadening is that it must be smaller than the overall sweep of the pulse carrier frequency during the interaction, i.e., $\Delta \omega_{inh} < \beta \tau_p$.

To verify the efficiency of the method in an inhomogeneously broadened medium, we present the population dynamics for two tripod-linkage atoms moving with two different velocities, thus having different Doppler shifts of the atomic resonance line

$$\Delta \omega_D^{(1,2)} = \pm \beta \tau_p,$$

(2.91)

as seen in Figs. 2.14, 2.15 and 2.16. The Doppler shift is included in the parameter $\epsilon_0 = \omega_{L0} - \omega_{10}$ describing the detuning of the central frequency of the chirped laser pulse from the resonance.
frequency of the $|0\rangle \rightarrow |1\rangle$ transition. As it follows from Figs. 2.14, 2.15 and 2.16 the proposed scheme for creating a coherent superposition of states is valid even for these two limiting values of the Doppler shift.

Figure 2.14: Population dynamics for a tripod-linkage atom moving with a velocity causing a Doppler shift of the transition lines: $\omega_D = -2\beta\tau_p$. The rest of the interaction parameters are the same as for Fig. 2.11.

Figure 2.15: Population dynamics for a tripod-linkage atom moving with a velocity causing a Doppler shift of the transition lines: $\omega_D = 2\beta\tau_p$. The rest of the interaction parameters are the same as for Fig. 2.11.
2.3. CREATING A SUPERPOSITION OF QUANTUM STATES IN TRIPOD ATOMS

Figure 2.16: Time evolution of the phase of the Raman coherence between ground states $|1\rangle$ and $|3\rangle$ in case of the atom having a Doppler shift $\omega_D = \mp 2\beta p \tau_p$ (denoted by $\phi^\pm$), scaled by units of $\pi$. One observes that in both cases the phase tends to the same final value. In the calculation, the same interaction parameters are used as those in Fig. 2.11.

The process is robust against variations of the laser pulse peak intensity (Rabi frequency) and of the chirp rate. To verify this robustness, we introduced a harmonic modulation of the laser pulse amplitude and/or the speed of the chirp. The sole limitation on the parameters of the modulation (depth and frequency) is that the resulting spectral broadening of the pulse must be less than the frequency interval between the "close" and the "far" ground states, in order to preserve condition (2.64). Keeping this limitation in mind, our simulations showed that the proposed scheme was efficient even at a modulation of the laser pulse amplitude and the speed of the chirp with up to 50% depth.

For the propagation of a single, frequency-chirped pulse in a medium of four-level tripod-linkage atoms we have not yet made any calculations but we strongly suspect that there exists a similar transparency effect as the one we observed for a $\Lambda$-medium because we could map the population transfer in the tripod atom to that in a $\Lambda$-atom which was the subject of our investigation in Sec. 2.2.

As an appropriate quantum system for realizing the scheme for coherent superposition state creation in a tripod-linkage system, one could use for example the manifold of magnetic sublevels of a $J = 1 \leftrightarrow J' = 0$ transition in Samarium (Sm) atoms where we choose the manifolds of states having total momentum quantum numbers $J = 1$ and $J' = 0$ to be the ground and excited states, respectively (see Fig. 2.17). Note that this quantum system was used in [76] to experimentally demonstrate inversionless amplification of picosecond laser pulses due to Zeeman coherence between the magnetic sublevels of the ground state.
In order to have simultaneous interaction with all the working transitions of the tripod atom, we need both \( \pi \) and \( \sigma \) polarization components to be present in the laser radiation. It may be realized for example by applying a linearly polarized laser pulse with the electric strength vector \( \mathbf{E} \) pointing in a direction at a certain angle to the magnetic quantization axis (see Fig. 2.17). One has to apply an additional \( \pi \)-polarized non-resonant laser field to induce a Stark shift of the magnetic sublevel \( m = 0 \), thus realizing the separated "far" ground state. As for the "close" ground states, the magnetic sublevels with quantum numbers \( m = \pm 1 \) may be used. Thus, we have \( |J = 1, m = \pm 1\rangle \) for the "close" ground states, \( |J = 1, m = 0\rangle \) for the "far" ground state and \( |J = 0, m = 0\rangle \) for the excited state. While transitions from the "close" ground states to the excited state are provided by \( \sigma^{(+)\rangle} \) and \( \sigma^{(-)\rangle} \) components of the laser field, the \( \pi \) component provides transitions between the "far" ground state and the excited state, as seen in Fig. 2.17.

There is also a growing interest in studying the enhancement of nonlinear optical processes through preparation of coherent superposition states in spatially quantized systems such as quantum dots [77], including coupled quantum dots [78]. The level structure of a QD with a tripod linkage of appropriately spaced (one "far" and two "close") ground states and a single excited state may be calculated and experimentally constructed. In this case, one does not need the off-resonant laser field, introduced for the case of the Sm atoms, that shifts the \( |J = 1, m = 0\rangle \) in order for it to become the "far" ground state.
Part II

Quantum emitters coupled to surface plasmons of a nano-wire: a Green’s function approach
CHAPTER 3

COLLECTIVE BEHAVIOUR OF QUANTUM EMITTERS COUPLED TO AN ENGINEERED RESERVOIR

In the preceding chapter, our investigations involved interaction of atomic ensembles with strong, classical laser fields. In the following discussion we are going to consider the opposite limit: we examine the interaction of atoms with quantized fields, i.e., the modes of the reservoir surrounding them. It is important to note that since, in addition to the vacuum, there are macroscopic dielectric bodies present in the system, the reservoir modes are tailored according to the properties and geometry of these media.

In order to consider the behaviour of atoms coupled to such an environment, we have to go through a procedure involving several steps. As a first step, we quantize the electromagnetic field in the presence of macroscopic media. For the quantization a Green’s tensor formalism is used which is convenient for handling a varying number of quantum emitters and incorporating the action of linear media with losses in terms of macroscopic parameters such as the complex permittivity and permeability. After formulating the Hamiltonian that includes interaction of the point-like quantum emitters with the field, we then investigate their dynamics by means of a reduced master equation where tracing out the environment introduces effective interactions between them. These interactions manifest as effective couplings and dipole-dipole shifts that are key objects of our investigation since they contain the information about the collective behaviour of the quantum emitters.

3.1 Field quantization in presence of macroscopic objects and coupling to point-like quantum emitters

To describe atomic systems that interact with a reservoir modified by the presence of macroscopic linear dielectric media, we have to set the stage by introducing a quantized model of the dielectric bodies. Because of the possible presence of linear, dispersive, lossy, passive materials, we quantize the electromagnetic field according to the method described in [79, 80, 81, 82] which is well suited for
treatment these kinds of systems. Here, we summarize only the key elements of the procedure, a more detailed description of which is contained in Appendix 1. In this approach the electric field operator can be expressed in terms of the electric field and create elementary excitations of the combined vacuum-matter environment.

In case of non-magnetic media, the positive frequency components \( \omega > 0 \) of the electric field operator can be written as

\[
\hat{E}(r, \omega) = i\sqrt{\frac{\hbar}{\pi\epsilon_0 c^2}} \int d^3r' \sqrt{\epsilon''(r', \omega)} \tilde{G}(r, r', \omega) \hat{f}^e(r')
\]

and, from Maxwell’s equations,

\[
\hat{B}(r, \omega) = \frac{1}{i\omega} \nabla \times \hat{E}(r, \omega) = \frac{1}{\sqrt{\pi\epsilon_0 c^2}} \int d^3r' \sqrt{\epsilon''(r', \omega)} \left[ \nabla \times \tilde{G}(r, r', \omega) \right] \hat{f}^e(r')
\]

where \( \tilde{G}(r, r', \omega) \) is the Green tensor of the field, obeying the Maxwell-Helmholtz wave equation in the medium with a Dirac delta source at \( r = r' \):

\[
\left[ \nabla \times \kappa(r, \omega) \nabla - \frac{\omega^2}{c^2} \epsilon(r, \omega) \right] \tilde{G}(r, r', \omega) = \delta'(r - r'),
\]

\( \epsilon_0 \) is the electric permittivity of vacuum, \( \epsilon(r, \omega) \) and is the relative electric permittivity of which \( \epsilon'' = \text{Im} \left[ \epsilon(r, \omega) \right] \) is the imaginary part. \( \kappa(r, \omega) = 1/\mu(r, \omega) \) is the inverse of the relative magnetic permeability.

It should be noted that (3.1) and (3.2) do not contain a homogeneous solution. The limit of quantized vacuum can be obtained if we keep \( \epsilon'' \) at a small but finite value as \( |r| \to \infty \) and only at the end of the calculations we take \( \epsilon'' \to 0 \) (see [33, 84]).

Note that if we allow magnetic media in the reservoir we have to introduce the field operators \( \hat{f}^m(r), \hat{f}^m\dagger(r) \) in addition to those we already have. With these included, the positive-frequency electric field operator has the form

\[
\hat{E}(r, \omega) = i\sqrt{\frac{\hbar}{\epsilon_0\pi}} \int d^3r' \tilde{G}(r, r', \omega) \cdot \left\{ \frac{\omega^2}{c^2} \sqrt{\epsilon''(r', \omega)} \hat{f}^e(r') + \frac{\omega}{c} \nabla' \times \left[ \sqrt{-\kappa''(r', \omega)} \hat{f}^m(r') \right] \right\}
\]

where \( \kappa''(r, \omega) = \text{Im} \left[ \kappa(r, \omega) \right] \) is the imaginary part of the inverse of the relative magnetic permeability. \( \hat{B}(r, \omega) \) can be obtained the same way as it was calculated from (3.1).

The polaritonic operators \( \hat{f}^e_{\omega_1}(r) \) and \( \hat{f}^e_{\omega_1\dagger}(r) \) fulfill the bosonic commutation relations

\[
\begin{align*}
\left[ \hat{f}_{\omega_1}^e(r), \hat{f}_{\omega_1'}^e(r') \right] &= \delta_{ij} \delta_{\mu,\mu'} \delta(r - r') \delta(\omega - \omega') \\
\left[ \hat{f}_{\omega_1}^e(r), \hat{f}_{\omega_1'}^{e\dagger}(r') \right] &= \left[ \hat{f}_{\omega_1}^{e\dagger}(r), \hat{f}_{\omega_1'}^e(r') \right] = 0, \quad p, p' = e, m.
\end{align*}
\]

48
3.1. FIELD QUANTIZATION IN PRESENCE OF MACROSCOPIC OBJECTS AND COUPLING TO POINT-LIKE QUANTUM EMITTERS

We can write up the total electric field operator in Schrödinger picture, given by

\[ \hat{E}(r) = \int_0^{\infty} d\omega \left( \hat{E}(r, \omega) + H.c. \right). \]  

\(3.6\)

Expressing the magnetic field operator \( \hat{B}(r, \omega) \) with \( \hat{E}(r, \omega) \) by aid of Maxwell’s equations and, subsequently, constructing

\[ \hat{B}(r) = \int_0^{\infty} d\omega \left( \hat{B}(r, \omega) + H.c. \right), \]  

\(3.7\)

one can show [81] that the presented quantization scheme preserves the fundamental equal-time commutation relations for the quantized electric and magnetic fields:

\[ \left[ \hat{E}_i(r), \hat{E}_j(r') \right] = 0 = \left[ \hat{B}_i(r), \hat{B}_j(r') \right] \]  

\(3.8\)

\[ \left[ \epsilon_{ijk} \hat{E}_i(r), \hat{B}_j(r') \right] = -i\hbar \epsilon_{ijk} \partial_k^* \delta(r - r') \]  

\(3.9\)

where \( \epsilon_{ijk} \) is the \{i, j, k\} component of the antisymmetric unit tensor. The Hamiltonian for the medium-assisted field assumes the general form

\[ \hat{H}_{\text{F}} = \sum_{p=e,m} \int_0^{\infty} d\omega \int d^3r \ h \omega \hat{E}_p^+(r) \hat{E}_p(r) \]  

\(3.10\)

where, in case of non-magnetic linear dielectric media, one has to keep only the \( p = e \) term.

Having quantized the electromagnetic field, we can now proceed to write up the full Hamiltonian of the system consisting of the tailored reservoir and \( N \) quantum emitters. The free Hamiltonian for the atoms and the interaction Hamiltonian in dipole and rotating wave approximation, respectively, are

\[ \hat{H}_0^A = -\frac{1}{2} \sum_{j=1}^{N} \hbar \omega_A \hat{\sigma}_{zj} \]  

\(3.11\)

\[ \hat{H}_I = -\sum_{j=1}^{N} \hat{\sigma}_j^+ d_j \cdot \hat{E}^+(r_j) + H.c., \]

with \( \omega_A \) being the atomic transition frequency, \( \hat{\sigma}_{zj} = |g\rangle_j \langle g| - |e\rangle_j \langle e| \) and \( |g\rangle_j \) and \( |e\rangle_j \) being the ground and exited states of the \( j \)-th atom, respectively.

\[ \hat{E}^+(r) = \int_0^{\infty} d\omega \hat{E}(r, \omega) \]  

\(3.12\)

is the positive frequency part of the total medium-assisted electric field operator with \( \hat{E}(r, \omega) \) being defined in (3.1) or (3.4) and \( \hat{\sigma}_j^+ = |e\rangle_j \langle g| \) is the raising operator between the ground state \( |g\rangle \) and the excited state \( |e\rangle \) of the \( j \)-th atom. \( d_j \) is the electric dipole strength of the transition between ground and excited state in the \( j \)-th atom. The negative frequency counterpart of \( \hat{E}^+(r) \) is defined
as
\[
\hat{E}(r) = \int_0^\infty d\omega \hat{E}(r, \omega).
\]

With the above definitions, the full Hamiltonian of the system, containing the free Hamiltonians for the medium-assisted field and the field-emitter interaction Hamiltonian, assumes the form
\[
\hat{H} = \hat{H}_F + \hat{H}_A + \hat{H}_I.
\]

### 3.2 Dyadic Green’s functions

The dyadic Green’s function, or Green’s tensor, expresses the response of the environment to a single, Dirac delta excitation. Since we quantize the field in the way described in Sec. 3.1, the key element containing all the information about the reservoir is the Green’s tensor. Important advantages of using this kind of treatment are that we can incorporate intrinsic losses into the reservoir, vary the number of quantum emitters while retaining the same Green’s tensor and easily account for the effect of different emitter polarizations. Appendix 2 contains a more detailed description about the derivation and properties of the dyadic Green’s function.

The Green’s tensor obeys the classical Maxwell-Helmholtz wave equation:
\[
\left[ \nabla \times \kappa(\mathbf{r}, \omega) \nabla \times -\frac{\omega^2}{c^2} \epsilon(\mathbf{r}, \omega) \right] \bar{G}(\mathbf{r}, \mathbf{r}', \omega) = \bar{I} \delta(\mathbf{r} - \mathbf{r}'),
\]
where \(\epsilon(\mathbf{r}, \omega)\) and \(\kappa(\mathbf{r}, \omega) = 1/\mu(\mathbf{r}, \omega)\) are the relative complex electric permittivity and the inverse relative complex magnetic permeability, respectively, of the isotropic linear medium at position \(\mathbf{r}\) and frequency \(\omega\). If the environment consists of isotropic, homogeneous media with sharply defined boundaries, one has to solve the equation in every region between the boundaries. The linearity of (3.15) enables the superposition of its solutions. Thus, depending in which region the observation point \(\mathbf{r}\) is, we take the superpositions of direct, reflected and transmitted elementary solutions of the equation and then connect these superpositions to each other by imposing the boundary conditions
\[
\hat{n}_j \times \bar{G}(\mathbf{r}, \mathbf{r}', \omega)_{\mathbf{r}=\mathbf{R}_j^-} = \hat{n}_j \times \bar{G}(\mathbf{r}, \mathbf{r}', \omega)_{\mathbf{r}=\mathbf{R}_j^+}
\]
and
\[
\kappa_j(\omega) \hat{n}_j \times \nabla \times \bar{G}(\mathbf{r}, \mathbf{r}', \omega)_{\mathbf{r}=\mathbf{R}_j^-} = \kappa_{j+1} \hat{n}_j \times \nabla \times \bar{G}(\mathbf{r}, \mathbf{r}', \omega)_{\mathbf{r}=\mathbf{R}_j^+}.
\]

on them where \(\hat{n}_j\) is a unit vector orthogonal to the boundary between the \(j\)-th and \(j+1\)st layer, at the point \(\mathbf{R}_j\) located on the boundary and \(\mathbf{R}_j^\pm = \lim_{\epsilon \to 0} [\mathbf{R}_j \pm \epsilon \hat{n}_j]\). Conditions (3.16) and (3.17), respectively, are the equivalents of the boundary conditions
\[
\hat{n}_l \times \mathbf{E}(\mathbf{r}, \omega)_{\mathbf{r}=\mathbf{R}_l^-} = \hat{n}_l \times \mathbf{E}(\mathbf{r}, \omega)_{\mathbf{r}=\mathbf{R}_l^+}
\]
and
\[
\kappa_l(\omega) \hat{n}_l \times \mathbf{H}(\mathbf{r}, \omega)_{\mathbf{r}=\mathbf{R}_l^-} = \kappa_{l+1} \hat{n}_l \times \mathbf{H}(\mathbf{r}, \omega)_{\mathbf{r}=\mathbf{R}_l^+}.
\]
joining the tangential components of the electric, as well as the magnetic fields on the boundary —well known from classical electrodynamics.

Additionally, in case of lossless media, requiring that the boundary condition equations have a non-trivial solution results in the so-called mode equation that, as its name implies, defines the allowed modes in the system.

Appendix 2 contains the solutions of the Maxwell-Helmholtz equation in case of unbounded, and cylindrical as well as planar multilayered media used in our investigations.

3.3 Effective couplings and level shifts: master equation for a many-body system coupled to the reservoir

Having constructed the Hamiltonian (3.11) of the system, we can now proceed to investigate the dynamics of the coupled atoms-reservoir system. We do so starting in Schrödinger picture and arriving at a master equation for the atomic system in the end.

For the sake of simplicity, let us assume that the reservoir contains only non-magnetic materials. For magnetic media, the procedure to follow is analogous although a bit more tedious, and leads to a master equation that is formally the same as in the non-magnetic case, differing from it only in the form of the Green’s tensor.

Transforming the field and atomic operators into interaction picture spares us the inconvenience of dealing with the free Hamiltonian later on. Using the unitary operator \( \hat{U}(t) = e^{-i(\hat{H}_0^F + \hat{H}_0^A)t} \),

\[
\begin{align*}
\hat{f}_\omega^{int}(\mathbf{r}) &= \hat{U}^\dagger(t)\hat{f}_\omega(\mathbf{r})\hat{U}(t) = \hat{f}_\omega(\mathbf{r})e^{-i\omega t} \\
\hat{\sigma}^{int}_j &= \hat{U}^\dagger(t)\hat{\sigma}^\dagger\hat{U}(t) = \hat{\sigma}^\dagger e^{i\omega_A t}
\end{align*}
\]

where \( \omega_A \) is the atomic transition frequency. The interaction Hamiltonian in interaction picture assumes the form

\[
\hat{H}_I = -\int d^3r \int_0^\infty d\omega \sum_{j=1}^N i \frac{\hbar}{\pi \varepsilon_0} \frac{\omega^2}{c^2} \sqrt{\varepsilon''(\mathbf{r},\omega)} \hat{\sigma}^\dagger_j d_j^F \hat{G}(\mathbf{r},\mathbf{r},\omega) \hat{f}_\omega(\mathbf{r}) e^{-i(\omega - \omega_A)t} + \text{H.c.},
\]

This done, the equation of motion of the total system-reservoir density operator in the interaction picture can be written as

\[
\dot{\hat{\rho}}_{SR}(t) = \frac{1}{i\hbar} \left[ \hat{H}_I(t), \hat{\rho}_{SR}(t) \right] .
\]

We proceed according to the Wigner-Weisskopf theory, assuming a weak connection between the reservoir and the atoms, as well as setting them decoupled at the initial time \( t_i \). Since we are solely interested in the behaviour of the atoms, we trace out the reservoir, that is, \( \dot{\hat{\rho}}_S = \text{Tr}_R\{\hat{\rho}_{SR}\} \).

\[
\dot{\hat{\rho}}_S(t) = -\frac{i}{\hbar} \text{Tr}_R \left[ \hat{H}_I(t), \dot{\hat{\rho}}_S(t_i) \otimes \hat{\rho}_R(t_i) \right] - \frac{1}{\hbar^2} \text{Tr}_R \int_{t_i}^t dt' \left[ \hat{H}_I(t'), \left[ \hat{H}_I(t'), \dot{\hat{\rho}}_S(t') \otimes \hat{\rho}_R(t_i) \right] \right] .
\]
CHAPTER 3. COLLECTIVE BEHAVIOUR OF QUANTUM EMITTERS COUPLED TO AN ENGINEERED RESERVOIR

Assuming that the characteristic timescale of the reservoir is much faster than that of the atomic processes, or, in the language of Green’s functions, the spectral width of the Green’s tensor of the environment is much larger than the inverse of the characteristic time of the atoms (i.e., the lifetime of the excited state), we can make the Markov approximation, by replacing \( \hat{\rho}_S(t') \) by \( \hat{\rho}_S(t) \) in the second term of the RHS of (3.3). This means that the time resolution of the atomic system being much too crude to resolve the rapid reservoir processes, the atoms will always see an equilibrated environment, i.e., the system has no 'memory'. In our calculations, we will have to check if the approximation is applicable.

For tracing out the environment, we have to know what \( \hat{\rho}_R(t_i) \) looks like. We assume the reservoir \((\hat{f}_ω(r), \hat{f}_ω^†(r))\) to be in vacuum state and execute the trace accordingly. Simplifying the equation, we encounter the integral below which we replace as

\[
\int_{t_i}^{t} dt' e^{-i(\omega - \omega_A)(t-t')} \approx \int_{-\infty}^{\infty} dt' e^{-i(\omega - \omega_A)(t-t')} = \pi \delta(\omega - \omega_A) - i \frac{\mathcal{P}}{\omega - \omega_A}.
\]

(3.24)

This is a very good approximation at optical frequencies. Also, using the Green’s tensor property for nonmagnetic materials (see Appendix 2)

\[
\int d^3 r d^3 r' \frac{\omega^2}{c^2} \epsilon''(r', \omega) \bar{G}(r_1, r', \omega) \bar{G}^†(r_2, r', \omega) = \text{Im} \left[ \bar{G}(r_1, r_2, \omega) \right]
\]

(3.25)

makes the resulting expressions much more transparent. The reduced master equation in Markov approximation has the final form

\[
\dot{\hat{\rho}}_S = i \sum_{k,l=1}^{N} \delta_{kl} [\hat{\sigma}^k_\dagger \pi_l, \hat{\rho}_S] - \sum_{k,l=1}^{N} \Gamma_{kl} \left( \hat{\sigma}^k_\dagger \pi_l \hat{\rho}_S + \hat{\rho}_S \hat{\sigma}^k_\dagger \pi_l - 2 \hat{\sigma}^k_\dagger \pi_l \hat{\rho}_S \hat{\sigma}^k_\dagger \right)
\]

(3.26)

where the effects of the traced-out reservoir manifest as energy shifts, single-atom decays and unitary as well as dissipative couplings between two atoms. \( \delta_{kk} \) are the single-atom Lamb-shifts and \( \delta_{k\neq l} \) are the radiative dipole-dipole shifts. The non-Hermitian part of the equation contains \( \Gamma_{kk} \) and \( \Gamma_{k\neq l} \) which are the single-atom decay rates and dissipative atom-atom couplings, respectively. Quite intuitively, the energy shifts, decay rates and couplings of the atoms are governed by the spectral response function of the reservoir, i.e., the Green’s tensor of the electromagnetic field coupled to the dielectric media:

\[
\Gamma_{kl} = \frac{2\omega^2_A}{\hbar \epsilon_0 c^2} d_k^T \text{Im} \left[ \bar{G}(r_k, r_l, \omega_A) \right] d_l
\]

(3.27)

\[
\delta_{kl} = \frac{1}{\hbar \epsilon_0 \pi} \mathcal{P} \int_{0}^{\infty} d\omega \frac{\omega^2 d_k^T \text{Im} \left[ \bar{G}(r_k, r_l, \omega) \right] d_l}{\omega - \omega_A}.
\]

(3.28)

Here \( d_k \) and \( r_k \) are the position and the electric dipole transition strength, respectively, of the atom.
labeled with the index \( k \) and \( \omega_A \) is the atomic transition frequency. In the following, we omit the \( S \) subscript of the density operator for the atomic system since we will not deal with \( \hat{\rho}_R \) anymore.

\( \Gamma_{kl} \) and \( \delta\omega_{kl} \) play a central role in our investigations, because they contain information about the modified collective decay rates and transition frequency modifications, respectively. This, however, only becomes apparent if the master equation is written up in the proper basis. By proper basis we mean such a complete set of states in which the part of (3.26) responsible for the decay out of a given basis state is diagonal so that the decay of the population of a given state will be proportional only to that same population and it will not depend on coherences (off-diagonal elements). We will call this a diagonal basis; this is the basis in which the modified collective decay rates gain a real physical meaning.

### 3.3.1 Finding the diagonal basis for the master equation

Writing up the master equation (3.26) in an arbitrary basis of states and examining the non-unitary, or Liouvillian part (3.29) one sees that the Liouvillian contribution to the time evolution of diagonal elements (i.e. populations of the corresponding basis states) of \( \rho \) contains diagonal as well as off-diagonal density matrix elements (coherences). This makes it difficult to interpret the physics of the dissipative processes in the atomic system: it would be better - if possible - to find a basis where the Liouvillian part of the master equation is diagonal, i.e., the dissipative dynamics of populations depends only on populations, not on coherences.

Such a basis for a given system, however, can be quite an effort to find. For \( N \) atoms, we start from the original atomic basis \( \{ |g\rangle_1 |g\rangle_2 \ldots |g\rangle_N \}, \{ |e\rangle_1 |g\rangle_2 \ldots |g\rangle_N \}, \{ |g\rangle_1 |e\rangle_2 \ldots |g\rangle_N \}, \ldots, \{ |e\rangle_1 |e\rangle_2 \ldots |e\rangle_N \} \}. 

Due to the fact that generally all the density matrix elements are coupled by the system of equations arising from the Liouvillian part, one has to diagonalize the whole of it in a single step.

The dimension of a basis that spans the Hilbert space for \( N \) 2-level atoms is \( 2^N \). Thus, the density matrix has \( 2^N(2^N + 1)/2 \) independent elements. So, in order to find the desired basis one would have to diagonalize a \( [2^N(2^N + 1)/2] \times [2^N(2^N + 1)/2] \) matrix which can be an impossible task if \( N \) is large.

However, for finding a basis in which the states have physically interpretable decay rates, we do not need to diagonalize the total dissipative part of (3.26). As we will see below, this eliminates a lot of difficulties concerning the diagonalization problem.

Examining the Liouvillian (3.29) one recognizes that the first two terms of the RHS are responsible for the decay out of a given state, while the third term accounts for the population that trickles down into the state from the manifold of states having one more excitation. It is due to this third term that the manifolds of states having different numbers of excitations are coupled.

\[
\mathcal{L}\hat{\rho} = - \sum_{k,l=1}^{N} \frac{\Gamma_{kl}}{2} \left( \hat{\sigma}_k^\dagger \hat{\sigma}_l \hat{\rho} + \hat{\rho} \hat{\sigma}_l^\dagger \hat{\sigma}_k - 2 \hat{\sigma}_l \hat{\rho} \hat{\sigma}_k^\dagger \right) 
\]  

(3.29)

However, if we are interested in the collective decay rates of states in a manifold having a given
number of excitations, we can assume that higher lying states are not populated, that is, the density matrix elements associated with them are zero. In this case, the contribution of the third term will be zero, and the investigated manifold can be regarded separately: so, instead of having to diagonalize the total Liouvillian, we only have to do so in the said manifold with zero third-term contribution. The basis we thus find we shall call a *diagonal* basis for the atomic system.

The dimension of the matrices in this case depends on the number of excitations in the corresponding manifold of states. Because the 2-level atoms can contain either one or zero excitations, the number of states in a manifold with $n$ excitations is

$$M_n = \binom{N}{n}$$  \hspace{1cm} (3.30)

if $n \leq N$, otherwise $M_n = 0$. In this way, the dimension of the matrix is only $[M_n(M_n + 1)/2] \times [M_n(M_n + 1)/2]$. The states of a manifold with a fixed number of excitations $n$ and $N$ atoms can be written as

$$|n, m\rangle = \sum_{j(1), \ldots, j(n)} C^{(m)}_{j(1) \ldots j(n)} \hat{\sigma}^\dagger_{j(1)} \cdots \hat{\sigma}^\dagger_{j(n)} |0\rangle$$  \hspace{1cm} (3.31)

where $|0\rangle = |g\rangle_1 \cdots |g\rangle_N$ is the ground state of the whole ensemble and $m = 1 \ldots M_n$. Requiring $|n, m\rangle$ to be a diagonal basis element, we have

$$\langle n, m | \sum_{k,l} \frac{\Gamma_{kl}}{2} (\hat{\sigma}^\dagger_k \hat{\sigma}_l + \hat{\sigma}^\dagger_l \hat{\sigma}_k) |n, m\rangle = \gamma^{(m)} \langle n, m | \hat{\rho} | n, m\rangle.$$  \hspace{1cm} (3.32)

As we shall soon see, (3.32) leads to an eigenvalue problem, the solution of which produces the coefficients $\{C^{(m)}_{j(1) \ldots j(n)}\}_{m=1}^{M_n}$.

Below we present the calculation of the ideal basis for manifolds with one and two excitations and, as an example, apply them for the case of $N = 2$. Manifolds with a higher number of excitations can be calculated in a similar fashion.

**Single-excitation manifold**

In case of a single excitation, the number of basis states is $M_1 = N$. The states we would like to find can be expressed as

$$|1, m\rangle = \sum_{j=1}^{N} C^{(1)}_{j} \hat{\sigma}^\dagger_{j} |0\rangle.$$  \hspace{1cm} (3.33)
3.3. EFFECTIVE COUPLINGS AND LEVEL SHIFTS: MASTER EQUATION FOR A MANY-BODY SYSTEM COUPLED TO THE RESERVOIR

Applying (3.32) we have to solve

\[
- \sum_{i,j,k=1}^{N} \Gamma_{i,j} \left\{ C_{k}^{(m)*} \langle 0 | \hat{\sigma}_{k} \hat{\sigma}_{j} \hat{\rho} | 1, m \rangle + C_{k}^{(m)} \langle 1, m | \hat{\rho} \hat{\sigma}_{k} \hat{\sigma}_{j} | 0 \rangle \right\} = \frac{\gamma^{(m)}}{2} \sum_{k=1}^{N} \left\{ C_{k}^{(m)*} \langle 0 | \hat{\sigma}_{k} \hat{\rho} | 1, m \rangle + C_{k}^{(m)} \langle 1, m | \hat{\rho} \hat{\sigma}_{k} | 0 \rangle \right\} \left( = \gamma^{(m)} \langle 1, m | \hat{\rho} | 1, m \rangle \right),
\]

with \( m = 1 \ldots N \). Setting the second term on the LHS of (3.34) equal to that on the RHS, transforming the expressions by commuting the \( \hat{\sigma} \) operators and relying on the orthogonality of the original atomic basis elements yields the eigenvalue problem:

\[
\Gamma_{ij} C_{j}^{(m)} = - \frac{1}{2} \gamma^{(m)} C_{i}^{(m)},
\]

the solution of which gives the coefficients for constructing the diagonal basis in the single-excitation manifold, as well as the associated collective decay rates (note that according to the Einstein convention, we sum over the index \( j \)).

If, for example, \( N = 2 \) we can easily calculate the diagonal basis. Let us suppose that the coupling of both atoms to the reservoir is equally strong, i.e., \( \Gamma_{11} = \Gamma_{22} \). For the atom-atom couplings, \( \Gamma_{12} = \Gamma_{21} \) is always true. Thus, the decay matrix reads

\[
\bar{\Gamma} = \begin{pmatrix} \Gamma_{11} & \Gamma_{12} \\ \Gamma_{12} & \Gamma_{11} \end{pmatrix}
\]

in the basis of the single-excitation manifold states \(|eg\rangle\) and \(|ge\rangle\). Diagonalizing the matrix yields the diagonal single-excitation basis states and the collective decay rates belonging to them:

\[
|1, 1\rangle = \frac{1}{\sqrt{2}} (|eg\rangle + |ge\rangle) \rightarrow \Gamma_{11} + \Gamma_{12}
\]

\[
|1, 2\rangle = \frac{1}{\sqrt{2}} (|eg\rangle - |ge\rangle) \rightarrow \Gamma_{11} - \Gamma_{12}.
\]

This is the famous Dicke basis for a two-atom system, so, according to conventions, we call states \(|1, 1\rangle\) symmetric (|S\rangle) and \(|1, 2\rangle\) antisymmetric (|AS\rangle) states since they are constructed by symmetric and, respectively, antisymmetric superpositions of the original basis states. Regarding the collective decay rates, it is apparent that the modification compared to the single-atom decay rates lies solely in the presence of the effective interaction between the atoms that manifests in the atom-atom coupling \( \Gamma_{12} \). The structure and the decay rates of the diagonal basis for the two-atom system with \( \Gamma_{11} = \Gamma_{22} \) is shown in Fig. 3.1.
CHAPTER 3. COLLECTIVE BEHAVIOUR OF QUANTUM EMITTERS COUPLED TO AN ENGINEERED RESERVOIR

Figure 3.1: Diagonal basis for two atoms coupled to a common reservoir for the special case of identical single-atom decay rates ($\Gamma_{11} = \Gamma_{22}$), also known as the two-atom Dicke basis. The symmetric and antisymmetric basis states of the single-excitation manifold are defined as $|S\rangle = 1/\sqrt{2} (|eg\rangle + |ge\rangle)$ and $|AS\rangle = 1/\sqrt{2} (|eg\rangle - |ge\rangle)$, respectively. Due to the effective interaction between the atoms, the collective decay rates are $\Gamma_{11} \pm \Gamma_{12}$.

**Double-excitation manifold**

We proceed similarly to the case of a single excitation, with the exception that we shall have to be careful about some symmetries which the final equation has to fulfill. This is a feature that all the manifolds with numbers of excitation higher than one possess.

As ansatz for an ideal basis state we write

$$|2, m\rangle = \sum_{k,l=1}^{N} C_{kl}^{(m)} \hat{\sigma}_{k}^{\dagger} \hat{\sigma}_{l}^{\dagger} |0\rangle,$$

and $m = 1...M_{2}$. $\hat{\sigma}_{k}^{\dagger}$ and $\hat{\sigma}_{l}^{\dagger}$ commute in case of $k \neq l$ and for $k = l$ their contribution to $|2, m\rangle$ is zero, the coefficients $C_{k,l}^{(m)}$ have to be symmetric in their indices. We apply (3.32) to (3.39):

$$- \sum_{i,j,k,l=1}^{N} \Gamma_{ij} \left\{ C_{kl}^{(m)*} \langle 0| \hat{\sigma}_{i} \hat{\sigma}_{k}^{\dagger} \hat{\sigma}_{j} \hat{\rho} |2, m\rangle + C_{kl}^{(m)} \langle 2, m| \hat{\rho} \hat{\sigma}_{j}^{\dagger} \hat{\sigma}_{k} \hat{\sigma}_{l}^{\dagger} |0\rangle \right\}$$

$$= \frac{\gamma^{(m)}}{2} \sum_{p,q=1}^{N} \left\{ C_{pq}^{(m)*} \langle 0| \hat{\sigma}_{p} \hat{\sigma}_{q} \hat{\rho} |2, m\rangle + C_{pq}^{(m)} \langle 2, m| \hat{\rho} \hat{\sigma}_{q}^{\dagger} \hat{\sigma}_{p}^{\dagger} |0\rangle \right\}.$$  

Commuting the operators, exploiting the orthogonality of the original basis states and the symmetry in the indices of the coefficients $C_{k,l}^{(m)}$ leads to the equation

$$\Gamma_{ij} C_{jk}^{(m)} = -\frac{1}{4} \gamma^{(m)} C_{ik}^{(m)}.$$  

(3.40)
3.4 Surface plasmon modes of a lossless nanowire

A considerable part of our investigation involves quantum emitters interacting with an environment that contains a thin, metallic, cylindrical wire. The interaction strength in case of a quantum emitter interacting with the reservoir depends on the mode structure of the reservoir, given by its Green’s tensor. In the following, we will take a look at the properties of the guided modes of the metallic wire, i.e., the surface plasmons.

In order to do that, we have to calculate the Green’s tensor for an infinite, cylindrically symmetric system, seen in Fig. 3.2, consisting of two regions: the external region, denoted by 0, is vacuum and...
CHAPTER 3. COLLECTIVE BEHAVIOUR OF QUANTUM EMITTERS COUPLED TO AN ENGINEERED RESERVOIR

Figure 3.2: A cylindrical, metallic nanowire (region 1), embedded in vacuum (region 0). Depending on the wire radius $R$, the guided surface plasmon modes of the wire can have a strong confinement in the radial direction as they travel along the $z$ symmetry axis. The vectors $\mathbf{r}$ and $\mathbf{r}'$ are the observation and source points, respectively.

the internal one, denoted by 1, is the infinite wire itself, having a negligible magnetic response.

The wire has a radius $R$ and its symmetry axis is the $z$ axis. We solve the Maxwell-Helmholtz equation (3.15) in both regions and expand the solutions in tensorial products of cylindrical vector wave functions (see Appendix 2). As written there, if we have a single cylinder embedded in vacuum and the source point $\mathbf{r}'$ is in the vacuum region we can express the Green tensor as

$$
\bar{G}(\mathbf{r}, \mathbf{r}', \omega) = \begin{cases} 
\bar{G}_0(\mathbf{r}, \mathbf{r}', \omega) + \bar{G}_R(\mathbf{r}, \mathbf{r}', \omega) & r > R \\
\bar{G}_T(\mathbf{r}, \mathbf{r}', \omega) & r < R 
\end{cases}
$$

(3.44)

where $r$ is the distance of the point of observation from the symmetry axis $z$ of the wire. If the observation point $\mathbf{r}$ is outside the cylinder, we have the sum of a direct (or vacuum) contribution $\bar{G}_0$ and a reflected contribution $\bar{G}_R$, whereas inside the cylinder we only get a transmitted term $\bar{G}_T$.

As seen in more detail in Appendix 2 the Green’s tensor for a cylindrically symmetric system can be written as

$$
\bar{G}(\mathbf{r}, \mathbf{r}', \omega) = \sum_{n=0}^{\infty} \int_{-\infty}^{\infty} dk_z \bar{G}^{(n)}(\mathbf{r}, \mathbf{r}', \omega; k_z)
$$

(3.45)

where $\bar{G}^{(n)}(\mathbf{r}, \mathbf{r}', \omega; k_z)$ is an analytically known function, containing tensorial products of cylindrical harmonic vector wave functions (expanding the Green’s tensor in this basis makes it possible to fulfill the boundary conditions in a cylindrically symmetric geometry), $k_z$ is the wave vector component parallel to the cylindrical symmetry axis, and $n$ is the cylindrical harmonic order.

The reflection and transmission coefficients contained in the Green’s tensor are fixed by the boundary conditions (see Eqs. (3.16) and (3.17)) at the surface of the cylinder ($r = R$):

$$
\hat{r} \times \bar{G}(\mathbf{r}, \mathbf{r}')_{r=R-} = \hat{r} \times \bar{G}(\mathbf{r}, \mathbf{r}')_{r=R+} \\
\hat{r} \times \nabla \times \bar{G}(\mathbf{r}, \mathbf{r}')_{r=R-} = \hat{r} \times \nabla \times \bar{G}(\mathbf{r}, \mathbf{r}')_{r=R+}.
$$

(3.46)  (3.47)

Writing up (3.46) and (3.47), one finds a system of linear algebraic equations where the unknown
variables are the reflection and transmission coefficients. It can be arranged in a matrix form as

$$\bar{A}x = b$$  \hspace{1cm} (3.48)

where \(x\) is the vector containing the unknown coefficients and \(b\) is a vector containing the contribution of the vacuum or direct (\(G_0\)) part of the Green’s tensor.

To find conditions for the guided modes of this system, we do the following. First of all, we assume a lossless material for the wire in order to provide the possibility for modes which preserve their properties after propagating any distance from the source. As a next step, we place the observation point \(r\) very far from the source position \(r'\), in fact requiring \(|r - r'| \to \infty\). This results in vanishing of the direct contribution of the source compared to the wire-assisted contributions, i.e., \(|G_{0ij}| \ll |G_{Rij}|, |G_{Tij}|\) for all \(\{i, j\}\) elements. Thus, (3.48) transforms into the homogeneous equation

$$\bar{A}x = 0.$$  \hspace{1cm} (3.49)

Now, to have non-zero solutions infinitely far away from the source (i.e., eigenmodes), we require (3.49) to have a nontrivial solution, which means that the determinant of the matrix \(A\) has to vanish. \(\det(A) = 0\) yields the mode equation at a given frequency \(\omega\) for each harmonic order \(n\). This mode equation is identical to that obtained by different methods in [49, 50]:

$$\frac{n^2 k_0^2}{R^2} \left( \frac{1}{k_{r1}^2} - \frac{1}{k_{r0}^2} \right)^2 = \left( \frac{1}{k_{r1}^2} \frac{J_n'(k_{r1} R)}{J_n(k_{r1} R)} - \frac{1}{k_{r0}^2} \frac{H_n'(k_{r0} R)}{H_n(k_{r0} R)} \right) \times \left( \frac{k_0^2}{k_{r1}^2} \frac{J_n'(k_{r1} R)}{J_n(k_{r1} R)} - \frac{k_0^2}{k_{r0}^2} \frac{H_n'(k_{r0} R)}{H_n(k_{r0} R)} \right)$$  \hspace{1cm} (3.50)

where \(R\) is the wire radius, \(n\) is the index for the cylindrical order and \(k_z\) is the longitudinal component of the wave vector \(k\), i.e., the one which is parallel to the symmetry axis of the wire. \(k_0\) is the magnitude of the wave vector in vacuum and \(k_1 = \sqrt{\epsilon} k_0\) is the same in the wire, \(\epsilon\) being the relative electric permittivity of the metal. The radial components of the wave vector outside and inside the wire are, respectively, \(k_{r0} = \sqrt{k_0^2 - k_z^2}\) and \(k_{r1} = \sqrt{k_1^2 - k_z^2}\). \(J_n(x)\) and \(H_n(x)\) are, respectively, Bessel and Hankel functions of the first kind, and where they have a prime as an upper index, they have to be differentiated by their argument.

If one expresses all radial wave vector components with \(k_z\), the roots of (3.50) yield the allowed values for \(k_z = k_z(\omega; n)\) for each transversal mode \(n\). These are the surface plasmon modes. In Fig. 3.3 the allowed values of \(k_z\) are plotted as function of wire radius \(R\) for the different cylindrical harmonic orders. One recognizes that with the shrinking of \(R\), all solutions belonging to the \(n \neq 0\) modes have a cutoff and only the one belonging to \(n = 0\) persists. The reason behind this is that the \(n = 0\) cylindrical mode has no ‘winding’, i.e., it is independent of \(\phi\). The corresponding value of \(k_z\) diverges as \(1/R\) for \(R \to 0\). This divergence of \(k_z\) is associated with a strong confinement of the field around the wire: When \(k_z\) diverges, \(k_{r0} = \sqrt{k_0^2 - k_z^2}\) attains a large imaginary value. Since the radial confinement is mainly determined by \(H_n(k_{r0} r) \sim \exp(ik_{r0} r)\), the divergence of \(k_z\) means that the field is confined to a radial distance proportional to \(R\). Unlike dielectric waveguides, conducting waveguides thus allow for a strong confinement of the electromagnetic field around the waveguide.
Figure 3.3: $k_z$ components of the guided modes of a nanowire ($\epsilon = -50$), as the function of the wire radius $R$ and cylindrical harmonic order $n$. When the radius decreases, all modes with a non-zero $n$ have a cutoff, whereas the $n = 0$ mode has an increasing $k_z$, that is, an increasingly strong confinement. $R$ is scaled with the vacuum radiation wavelength $\lambda_0$.

As we will show later, this feature enables a strong coupling between atoms and propagating plasmon modes.

Note that, although the continuum model works very well on the length scales and dimensions we use (much larger than the distance between the atoms inside the metal), eventually it breaks down as $R \to 0$ since at and below radii comparable to the inter-atomic distance in the metal the atomic nature of the wire becomes important.

3.5 Interaction of a single emitter with a nano-wire

The starting point of our investigations involving interactions of quantum emitters mediated by surface plasmon eigenmodes of one or more nano-wires is the coupling of a single emitter to plasmons. In the following, we would like to characterize the interaction of a single atom with the surface plasmon eigenmodes of a nano-wire, assuming the configuration depicted in Fig. 3.4. We have already mentioned in Sec. 3.4 and, in more detail, in Appendix 2 that for a cylindrically symmetric system one can expand the Green’s tensor as

$$\bar{G}(r, r', \omega) = \sum_{n=0}^{\infty} \int_{-\infty}^{\infty} dk_z \bar{G}^{(n)}(r, r', \omega; k_z)$$

Examining the structure of $\bar{G}^{(n)}(r, r', \omega; k_z)$, one sees that (in the absence of losses) it has singularities exactly at $k_z = \pm k_z(\omega; n)$. These singularities originate from the $n$-th plasmonic mode of the nano-wire system.
For the numerical calculations we used $\epsilon = -50 + 0.6i$ as a typical value for electric permittivity inside the wire which corresponds to the electric permittivity of silver at around $\lambda = 1\mu m$ (∣85).}

\subsection{3.5.1 Width of the plasmon resonances}

In real systems, finite material losses are present. Taking into account the medium absorption, i.e. a small but finite value of $\epsilon''$, the discrete cylindrical modes turn into broadened resonances. This means that in case of losses there is no longer a single, well-defined $k_z$ but continuously many $k_z$ values peaked around $k_z(\omega; n)$ that contribute to the $n$-th plasmon mode. The consequences of this can be seen much clearer if the wire radius is much smaller than the optical wavelength, because then we can interpret the propagation of the plasmon mode as a 1D problem, looking at a single transverse mode only. The material losses induce a distribution of components with different $k_z$ values in the propagating plasmonic mode, each acquiring a phase factor $\exp(ik_zz)$ while propagating a distance $z$. This, in turn, results in a dephasing of the components upon propagation.

Fig. 3.5 shows the shape of the resonance belonging to the $n = 0$ cylindrical mode around $k_z = \pm k_z(\omega; n)$, the value corresponding to the lossless case. The lineshape is well approximated with a Lorentzian function. The HWHM of the resonance peak scales linearly with $\epsilon''$, as one would expect. In Fig. 3.6 the value of the HWHM is plotted, which assumes larger and larger values as the wire radius $R$ gets smaller. One recognizes a transition from a slower $R^{-1/2}$ to a faster $R^{-3/2}$ dependence of the width of the plasmon resonances at around $R \sim 10^{-1}\lambda_0$.

So, in case of $\epsilon'' \neq 0$, decreasing the wire radius is accompanied not only by a decreasing transverse mode area, but also by increasing propagation losses. The two effects are connected because the smaller the transverse mode area gets, the larger part of the propagating field will be concentrated inside the metal which induces increased propagation losses. We will see later that for a decreasing value of $R$ the detrimental effects of propagation losses will overcome the positive effect of an increased coupling. Thus, there exists a radius at which the tradeoff between coupling strength and propagation losses is optimal.
Figure 3.5: \( \text{Im} \left[ \hat{r}^T \tilde{G}^{(n=0)}(\mathbf{r}_A, \mathbf{r}_A, \omega; k_z) \hat{r} \right] \), in case of a single-mode, lossy wire \((\epsilon = 50 + 0.6i)\). \( \hat{r} \) is the unit vector in the radial direction. The broadened plasmonic resonance peaks show up at around the plasmonic longitudinal wavenumbers \( \pm k_{pl} \) (predicted by (3.50)), in the evanescent region \((k_{pl} > k_0)\). The distances and wave numbers are scaled with the vacuum radiation wavelength \( \lambda_0 \).

Figure 3.6: HWHM of the plasmonic resonance peaks shown in Fig.3.5 as a function of wire radius \( R \), in a log-log plot. The width and the radius are scaled by vacuum radiation wavelength \( \lambda_0 \). The electric permittivity is \( \epsilon = -50 + 0.6i \). As the wire gets thinner, the broadening of the resonance increases, i.e., the propagation losses increase.
3.5. INTERACTION OF A SINGLE EMITTER WITH A NANO-WIRE

3.5.2 Spontaneous emission and level shift of an atom near a nanowire

It is well known that a dielectric body near an atom influences its spontaneous emission rate. The interaction strength of a atom with the different modes of the electromagnetic field is inversely proportional to the square root of the effective mode volume. That also means that the probability of spontaneous emission into modes with the smallest effective mode volume will be the largest.

In the following we will take a look at the spontaneous emission rate of a two-level atom near the surface of a nanowire. We have seen before that it is possible to have a wire with a single, strongly confined mode at a given frequency, that is, a mode with a very small effective cross section area. Thus, we expect that if we place the atom close enough to the wire, its spontaneous emission rate will increase considerably [49, 50].

To ascertain this, we construct the reduced master equation for the system consisting of a single two-level atom in the vicinity of a cylindrical metallic wire, following the procedure described in 3.3 using the Hamiltonian (3.11) with \( N = 1 \).

\[
\dot{\hat{\rho}} = i \delta \omega \left[ \hat{\sigma}^\dagger \hat{\sigma}, \hat{\rho} \right] - \frac{\Gamma_{tot}}{2} \left( \hat{\sigma}^\dagger \hat{\sigma} \hat{\rho} + \hat{\rho} \hat{\sigma}^\dagger \hat{\sigma} - 2 \hat{\sigma} \hat{\sigma}^\dagger \hat{\rho} \right) \tag{3.52}
\]

where \( \hat{\sigma} = |g\rangle \langle e| \) and \( \hat{\sigma}^\dagger = |e\rangle \langle g| \) are the atomic operators flipping the state of the atom between ground state \( |g\rangle \) and excited state \( |e\rangle \). \( \delta \omega \) is the effective Lamb shift and \( \Gamma_{tot} \) is the total decay rate for a single atom:

\[
\Gamma_{tot} = \frac{2\omega_A^2 d_i d_j}{\hbar \epsilon_0 c^2} \text{Im} [G_{ij}(r_A, r_A, \omega_A)]
\]

\[
\delta \omega = \frac{d_i d_j}{\hbar \epsilon_0 \pi} \text{P} \int_0^\infty d\omega \frac{\omega^2}{c^2} \frac{\text{Im} [G_{ij}(r_A, r_A, \omega)]}{\omega - \omega_A},
\]

\( \omega_A \) and \( r_A \) being the transition frequency and position of the atom, respectively. \( d \) is the dipole moment of the atomic transition.

Fig. 3.7 shows the results of a numerical simulation for \( \Gamma_{tot} \) for different atom - wire axis distances and wire radii. According to these results, the interaction of the atom with the surface plasmon mode gives rise to a significant Purcell effect, enhancing the spontaneous emission rate by orders of magnitude. Thus, the atom interacts with the surface plasmon mode in the strong coupling regime. It is important to note that the interaction of the atom with the wire strongly depends on the direction of the polarization vector \( d \). The coupling to the plasmons is strongest when \( d \) points in the radial direction ([50]). In all simulations we used this configuration.

We have yet to justify the Markov approximation. In Fig. 3.8 we show \( \Gamma_{tot} \), as a function of different atom-wire distances and frequency. To specify the frequency dependence of the relative permittivity \( \epsilon(\omega) \) we use the Drude model and fit it to the data provided in [85] for silver. Since we stay at optical frequencies, we approximated the imaginary part to have the constant value it has.
Figure 3.7: Spontaneous decay rate of a single emitter relative to the vacuum spontaneous decay rate as a function of emitter - wire axis distance $r_A$ and wire radius $R$, both scaled by vacuum radiation wavelength $\lambda_0$. We use $\epsilon = -50 + 0.6i$. Because of the strong interaction of the emitter with the wire eigenmodes, there is a considerable increase in the spontaneous decay rate at small radii and emitter-wire distances. Since $\Gamma_{\text{tot}} \to \infty$ for $r_A \to R$, we have introduced a cutoff at a small $r_A - R$ for better visibility.

at the frequency ($\omega_0 = 1 \mu m$) and only the real part to vary,

$$\epsilon'(\omega) = 1 - \frac{2\pi 51}{(\omega/\omega_0)^2}$$

$$\epsilon''(\omega) = 0.6$$

which is a good approximation at optical frequencies. The results show that $\Gamma_{\text{tot}}$ has a very large width in frequency, much more than $0.5\omega_A$, which means that the recurrence time (which is proportional to the inverse of the width) is several orders of magnitude smaller than the characteristic time scale of the atomic decay. This justifies the use of the Markov approximation.

3.5.3 Sub-wavelength nanowire

We have seen in the preceding section that close proximity to the wire causes a significant increase of the total spontaneous decay rate of a single quantum emitter. However, it would also be important to know how much of the spontaneous decay is directed into the surface plasmon modes themselves.

The Green tensor formalism describes the interaction of one or many emitters with their surroundings in a compact form where contributions of different effects are difficult to distinguish. Thus, on first glance, the result does not enable us to separately determine the rate of spontaneous emission into electromagnetic modes associated with the nanowire plasmons and that into the rest of the reservoir. In the following we will show, however, that with some further calculations we can
3.5. INTERACTION OF A SINGLE EMITTER WITH A NANO-WIRE

Figure 3.8: Spontaneous decay rate of a single emitter relative to the vacuum spontaneous decay rate as a function of frequency $\omega$ and emitter-wire axis distance $r_A$, in case of a lossy wire with radius $R = 0.01\lambda_0$. For the dispersion of the relative complex electric permittivity $\epsilon(\omega)$ we use a Drude model, specified in (3.55). $\omega$ and $r_A$ are scaled by the frequency $\omega_0$ and the corresponding vacuum radiation wavelength $\lambda_0 = 2\pi c/\omega_0$, respectively. The large width in frequency verifies the applicability of the Markov approximation. Since $\Gamma_{tot} \to \infty$ for $r_A \to R$, we have introduced a cutoff at a small $r_A - R$ for better visibility.

still resolve the contributions of the different modes.

We can separate the interaction Hamiltonian

$$\hat{H}_I = \hat{H}_I^{tr} + \hat{H}_I^{ev}$$

into a part $\hat{H}_I^{ev}$ containing the evanescent modes ($|k_z| > k_0$) and a part $\hat{H}_I^{tr}$ containing the traveling-wave part ($|k_z| \leq k_0$) of the electromagnetic field. We do this because, as shown in Fig.3.3, the plasmons are evanescent in the radial direction ($|k_z| > k_0$), and thus do not contribute to the traveling-wave part of the $k$-spectrum. Moreover, for lossless media $\epsilon'' = 0$, one can verify from the structure of $\text{Im}[\tilde{G}^{(n)}(r_A, r_A, \omega; k_z)]$ that the surface plasmons are the only contribution in the evanescent region. In this way we can define traveling and evanescent creation and annihilation operators $\hat{a}_{\omega}^{tr, ev}, \hat{a}_{\omega}^{tr, ev\dag}$ with the commutation relations

$$\begin{bmatrix} \hat{a}_{\omega}^{tr, ev}, \hat{a}_{\omega'}^{tr, ev\dag} \end{bmatrix} = \delta_{tr, ev} \delta(\omega - \omega')$$
$$\begin{bmatrix} \hat{a}_{\omega}^{tr, ev}, \hat{a}_{\omega'}^{tr, ev\dag} \end{bmatrix} = 0.$$

We define the interaction Hamiltonian as

$$\hat{H}_I = -\int_0^\infty d\omega \, g^{tr}(\omega) \hat{a}_{\omega}^{tr\dag} \hat{\sigma} - \int_0^\infty d\omega \, g^{ev}(\omega) \hat{a}_{\omega}^{ev\dag} \hat{\sigma} + H.c.$$
where \( g^{ev, tr}(\omega) \) is the coupling strength to the evanescent and traveling modes, respectively, at frequency \( \omega \) which will be specified later. \( \hat{\sigma}^\dagger \) is the atomic raising operator between ground state \( |g\rangle \) excited state \( |e\rangle \). Writing up the Heisenberg equation for \( \hat{a}^{ev, tr}_\omega \) and \( \hat{\sigma} \), and using that the traveling and evanescent operators commute, we get

\[
\dot{\hat{a}}^{ev, tr}_\omega = -i\omega \hat{a}^{ev, tr}_\omega(t) + \frac{i}{\hbar} g^{ev, \text{tr}}(\omega) \hat{\sigma}(t) \tag{3.59}
\]

\[
\dot{\hat{\sigma}} = -i\omega_A \hat{\sigma}(t) - \frac{i}{\hbar} \int_0^\infty d\omega \left(g^{tr}(\omega) \hat{a}^{\text{tr}}_\omega(0) + g^{ev}(\omega) \hat{a}^{ev}_\omega(0)\right) e^{-i\omega t} \hat{\sigma}_z(t). \tag{3.60}
\]

We formally integrate (3.59)

\[
\hat{a}^{ev, tr}_\omega(t) = e^{-i\omega t} \hat{a}^{\text{tr}}(0) + \frac{i}{\hbar} \int_0^t d\tau \hat{\sigma}(\tau) e^{-i\omega(t-\tau)} \tag{3.61}
\]

and substitute it into (3.60):

\[
\dot{\hat{\sigma}} = -i\omega_A \hat{\sigma}(t) - \frac{i}{\hbar} \int_0^\infty d\omega \left(g^{\text{tr}}(\omega) \hat{a}^{\text{tr}}_\omega(0) + g^{ev}(\omega) \hat{a}^{ev}_\omega(0)\right) e^{-i\omega t} \hat{\sigma}_z(t)
\]

\[
+ \frac{1}{\hbar^2} \int_0^\infty d\omega \left(|g^{\text{tr}}(\omega)|^2 + |g^{ev}(\omega)|^2\right) \int_{-\infty}^t d\tau \hat{\sigma}(\tau) e^{-i\omega(t-\tau)} \hat{\sigma}_z(t). \tag{3.62}
\]

Note that in (3.62) there are no cross-terms because the traveling creation and annihilation operators commute with the evanescent ones.

We can repeat the same procedure using the Green function formalism. For the equation of motion of \( \mathbf{\tilde{f}}_\omega(r) \) and \( \hat{\sigma} \) we obtain

\[
\dot{\mathbf{\tilde{f}}}_\omega(r) = -i\omega \mathbf{\tilde{f}}_\omega(r) + \omega^2 \sqrt{\frac{\epsilon''(r, \omega)}{\hbar \pi \epsilon_0}} \mathbf{\tilde{G}}^*(r, r_A, \omega) \mathbf{d}\hat{\sigma} \tag{3.63}
\]

\[
\dot{\hat{\sigma}} = -i\omega_A \hat{\sigma}(t) - \frac{i}{\hbar} \int_0^\infty d\omega i \sqrt{\frac{\hbar \omega^2}{\pi \epsilon_0}} \int d^3r' \sqrt{\epsilon''(r', \omega)} \mathbf{d}^T \mathbf{\tilde{G}}(r_A, r', \omega) \mathbf{\tilde{f}}_\omega(r') \hat{\sigma}_z(t). \tag{3.64}
\]

By formal integration, we express the solution of (3.63) and substitute it into (3.64). Using the Green function property (3.25) for nonmagnetic materials we obtain the following expression:

\[
\dot{\hat{\sigma}} = -i\omega_A \hat{\sigma} - \frac{i}{\hbar} \mathbf{\tilde{f}}^{(+)\text{free}}_\omega(r_A, t) \cdot \mathbf{d}\hat{\sigma}_z(t)
\]

\[
+ \frac{1}{\hbar^2} \int_0^\infty d\omega \frac{\hbar \omega^2}{\pi \epsilon_0 c^2} \mathbf{d}^T \text{Im} \left[ \mathbf{\tilde{G}}(r_A, r_A, \omega) \right] \mathbf{d} \int_{-\infty}^t d\tau \hat{\sigma}(\tau) e^{-i\omega(t-\tau)} \hat{\sigma}_z(t), \tag{3.65}
\]

which is equivalent to (3.62). Both the source and the observation points are in vacuum so \(|k| =
Comparing (3.62) and (3.66), separating the evanescent and traveling contributions, and exploiting \( k_c \) currents, also to non-radiative losses which, in turn, have a contribution to \( \epsilon_{\text{metallic plane with a single oscillating charge above it}} \). As shown in Ref. [86], evanescent region: apart from the resonance peaks, we observe a broad, low-amplitude background but finite peaks with a finite width. Moreover, they will no longer be the only contribution in the absence of losses. In case of a lossy wire, the plasmon resonances are not singularities anymore but finite peaks with a finite width. (3.67) \( g^r(\omega) \) according to Fermi’s golden rule, the square of the coupling strength to a given mode of the field is proportional to the spontaneous decay rate into that mode. Because of this and Eqs. (3.67) and (3.68), we can define the decay rate into the plasmon mode associated with the \( n \)-th cylindrical order

\[
\Gamma_{pl}(\omega) = \sum_{n=0}^{\infty} \frac{2}{k_0} \int_{0}^{k_0} dk_z \sum_{n=0}^{\infty} \frac{\hbar \omega^2}{\pi \epsilon_0 c^2} d^3 \text{Im} \left[ \tilde{G}^{(n)}(r_A, r_A, \omega; k_z) \right] d = \sum_{n=0}^{\infty} \Gamma_{pl}^{(n)}(\omega). \tag{3.69}
\]

Note that the above decomposition into traveling and plasmonic components is strictly valid only in the absence of losses. In case of a lossy wire, the plasmon resonances are not singularities anymore but finite peaks with a finite width. Moreover, they will no longer be the only contribution in the evanescent region: apart from the resonance peaks, we observe a broad, low-amplitude background that is due to dissipative local circulating currents, analogous to those present in case of an infinite metallic plane with a single oscillating charge above it. As shown in Ref. [86], \( e'' \neq 0 \) results in non-radiative losses which, in turn, have a contribution to \( |g^{tr}(\omega)|^2 \), and, because of the circulating currents, also to \( |g^{ev}(\omega)|^2 \).

Our main interest lies in a thin, single-mode wire where we do not have higher-order plasmon modes (see Fig. 3.3). This allows us to resolve the plasmon mode from the circulating surface currents to a good approximation. The evanescent part of the \( n > 0 \) cylindrical orders contains only the circulating current background. As for the \( n = 0 \) contribution, one can verify that there is no peak-sitting-on-a-background behaviour in \( \text{Im} \left[ \tilde{G}^{(n=0)}(r_A, r_A, \omega; k_z) \right] \), but instead we have a single

\[
k_0 = \omega/c. \]
peak, corresponding to the plasmon mode, with an almost perfect Lorentzian shape. So, in the
$n = 0$ contribution we have no, or negligible circulating surface current contribution. Thus, in the
single-mode case the decay rate into the plasmon mode can be calculated as

$$\Gamma_{pl}(\omega) = 2 \int_{k_0}^{\infty} dk_z \frac{2\omega^2}{\hbar \epsilon_0 c^2} \text{d}^2 \mathbf{r} \text{Im} \left[ G^{(n=0)}(\mathbf{r}_A, \mathbf{r}_A; \omega, k_z) \right] \text{d}. \quad (3.70)$$

We plot the emission rate into the plasmons normalized to the overall decay rate as a function of
the atomic distance from the wire axis in Fig. 3.9. For small atom-wire distances, the spontaneous
decay is almost entirely directed into the plasmonic mode which reveals the extraordinarily strong
coupling achievable in near-field plasmonic systems. Furthermore, we observe that there is an
optimum distance at which this value is maximal. This is in accordance with the result in [49, 50].

Our description enables us to determine the origin of this effect. If the atom gets far from the
wire, the spontaneous emission rate into the confined plasmon modes decreases rapidly, while the
emission rate into the traveling modes ($0 < k_z < k_0$) stays more or less constant (practically zero
reflection from the wire and constant vacuum spontaneous emission rate) which results in an overall
decrease of $\Gamma_{pl}/\Gamma_{tot}$. Close to the wire the strong confinement of the plasmon modes enhances the
coupling to the plasmon mode such that the decay is directed almost exclusively into the guided
mode. However, if the atom gets too close to the wire the near field circulating current contribution
increases faster than the decay rate into the $n = 0$ plasmon mode. This, again, leads to an overall
decrease of $\Gamma_{pl}/\Gamma_{tot}$.
3.6 Plasmon-mediated interaction of two emitters: Dicke superradiance

If we place two emitters along the single-mode wire, both at the same distance from the surface, close enough to strongly couple to the single surface plasmon mode (as seen in Fig. 3.10), they will have a long-range interaction mediated by the plasmons. In the following, we will investigate this interaction.

![Figure 3.10: A pair of quantum emitters placed close to the surface of a thin, metallic nano-wire. Due to the strong atom-plasmon coupling, a long-range effective atom-atom coupling arises, resulting in a long-range Dicke superradiance phenomenon.](image)

The rotating-wave Hamiltonian in Schrödinger picture of two identical, two-level atoms interacting with the environment, i.e. with the electromagnetic field in presence of the nanowire, in the dipole approximation assumes the form

\[
\hat{H} = \int d^3r \int_0^\infty d\omega \hbar \omega \hat{f}_\omega^\dagger(\mathbf{r}) \hat{f}_\omega(\mathbf{r}) + \frac{\hbar}{2} \omega_A (\hat{\sigma}_{z1} + \hat{\sigma}_{z2}) \\
+ \left(-\hat{\sigma}_{1}^\dagger \hat{\mathbf{E}}^{(+)}(\mathbf{r}_1) \mathbf{d}_1 - \hat{\sigma}_{2}^\dagger \hat{\mathbf{E}}^{(+)}(\mathbf{r}_2) \mathbf{d}_2 + H.c.\right)
\]

where the indices 1 and 2 refer to the 1st and 2nd atom. The first term represents the field energy in the presence of the nanowire. The field is represented by the elementary excitations operators \( \hat{f}_\omega \) and \( \hat{f}_\omega^\dagger \), which are related to the electric field through Eq. (3.12) and (3.1). Since (3.1) involves the full Green function, the interaction of the field with the nanowire is already incorporated. This is one of the main advantages of the Green function formalism. Note that although describing the combined field-plus-nanowire system, the operators \( \hat{f}_\omega \) and \( \hat{f}_\omega^\dagger \) satisfy the free-field commutation relations (3.5) [81, 80]. The second term in the Hamiltonian represents the atomic energy and the second line describes the interaction between the atoms and the field excitations through (3.12) and (3.1). Similarly to the treatment in Sec. 3.5.2 for a single atom, we follow the procedure in Sec. 3.3 for \( N = 2 \) to get the equation of motion for the reduced, two-atom density matrix

\[
\dot{\hat{\rho}} = i \sum_{k,l=1}^{2} \delta\omega_{kl} \left[ \hat{\sigma}_{i}^\dagger \hat{\sigma}_{k} \hat{\rho} - \hat{\rho} \hat{\sigma}_{i}^\dagger \hat{\sigma}_{k} \right] - \sum_{k,l=1}^{2} \frac{\Gamma_{kl}}{2} \left( \hat{\sigma}_{i}^\dagger \hat{\sigma}_{k} \hat{\rho} + \hat{\rho} \hat{\sigma}_{i}^\dagger \hat{\sigma}_{k} - 2 \hat{\sigma}_{i} \hat{\sigma}_{k} \hat{\rho}^\dagger \right).
\]
Here $\delta\omega_{kk}$ are the single-atom Lamb shifts and $\delta\omega_{12}$ is the radiative dipole-dipole shift. Similarly, $\Gamma_{kk}$ is the single-atom decay rate $\Gamma_{tot}$ derived in Sec. 3.5.2 and $\Gamma_{12}$ is a contribution which describes the effective coupling between the atoms.

$$\Gamma_{12} = \frac{2\omega_A^2}{\hbar \epsilon_0 c^2} \mathbf{d}_1^T \text{Im} \left[ \hat{G}(\mathbf{r}_1, \mathbf{r}_2, \omega_A) \right] \mathbf{d}_2$$

(3.73)

$$\delta\omega_{12} = \frac{1}{\hbar \epsilon_0 \pi} \int_0^\infty d\omega \frac{\omega^2 \mathbf{d}_1^T \text{Im} \left[ \hat{G}(\mathbf{r}_1, \mathbf{r}_2, \omega) \right] \mathbf{d}_2}{\omega^2 - \omega_A}.$$  (3.74)

We would like to note that since the calculation of the dipole-dipole shift involves the integration of the Green tensor on the full spectrum, trying to perform it in the form of (3.74) can be a formidable task. It will thus be disregarded in the following discussion and be accounted for in a later chapter. By estimation, the shift mediated by the nanowire is typically at most of the order of the linewidth of the transition if the distance is larger than the plasmon wavelength.

Because of the effective interaction between the atoms, (3.72), we expect the emergence of collective decays. However, to observe these quantities, we have to find the diagonal basis for the two-atom system. We follow the method described in Sec. 3.3.1, with the assumption that $\Gamma_{11} = \Gamma_{22}$ (meaning that both of the atoms sit at the same distance from the wire surface) and knowing that $\Gamma_{12} = \Gamma_{21}$. We find that (3.72) is diagonal in the two-atom Dicke basis, namely, the states $|ee\rangle$, $|S\rangle = (|ge\rangle + |eg\rangle)/\sqrt{2}$, $|AS\rangle = (|ge\rangle - |eg\rangle)/\sqrt{2}$ and $|gg\rangle$. Writing up the equations of motion for the populations of these states we arrive at

$$\dot{\rho}^{ee}_{ee} = -2\Gamma_{11}\rho^{ee}_{ee}$$

(3.75)

$$\dot{\rho}^{S}_{S} = (\Gamma_{11} + \Gamma_{12})\rho^{ee}_{ee} - (\Gamma_{11} + \Gamma_{12})\rho^{S}_{S}$$

(3.76)

$$\dot{\rho}^{AS}_{AS} = (\Gamma_{11} - \Gamma_{12})\rho^{ee}_{ee} - (\Gamma_{11} - \Gamma_{12})\rho^{AS}_{AS}$$

(3.77)

$$\dot{\rho}^{gg}_{gg} = (\Gamma_{11} + \Gamma_{12})\rho^{S}_{S} + (\Gamma_{11} - \Gamma_{12})\rho^{AS}_{AS}.$$  (3.78)

We have here used the notation that the matrix elements of the two-particle density operator are $\rho_{gs}^{es} = \langle gs|\hat{\rho}|es\rangle$. Additionally, from the equations of the coherences, we perceive that the interaction shifts the resonance frequency of $|AS\rangle$ downwards (upwards) by $\delta\omega_{12}$, and that of $|S\rangle$ upwards (downwards) by the same amount, for example

$$\dot{\rho}^{S}_{gg} = i (\delta\omega_{11} + \delta\omega_{12}) \rho^{S}_{gg} - \frac{1}{2} (\Gamma_{11} + \Gamma_{12}) \left( \rho^{S}_{gg} - 2\rho^{ee}_{ee} \right)$$

(3.79)

$$\dot{\rho}^{AS}_{gg} = i (\delta\omega_{11} - \delta\omega_{12}) \rho^{AS}_{gg} - \frac{1}{2} (\Gamma_{11} - \Gamma_{12}) \left( \rho^{AS}_{gg} + 2\rho^{ee}_{ee} \right).$$  (3.80)

Fig. 3.11 shows the new collective atomic basis states in which the decay part of the master equation is diagonal, as well as the collective decay rates and dipole-dipole shifts. As we will show below, the presence of $\Gamma_{12}$ is responsible for a super- and subradiance effect related to the symmetric and antisymmetric transitions.
3.6. PLASMON-MEDIATED INTERACTION OF TWO EMITTERS: DICKE SUPERRADIANCE

Figure 3.11: Collective atomic decays and dipole-dipole shifts of a pair of interacting two-level atoms, described in the Dicke basis. States $|ee\rangle$ and $|gg\rangle$ mean both atoms to be in the excited and in the ground states, and $|S\rangle$ and $|AS\rangle$ are the symmetrized and antisymmetrized single-excitation states, respectively. If there is a strong effective coupling (high $\Gamma_{12}$) between the atoms, the difference between the collective decay rates becomes large, leading to a superradiance phenomenon.

$$\Gamma_{11} + \Gamma_{12}$$
$$\Gamma_{11} - \Gamma_{12}$$

$|ee\rangle$ $|gg\rangle$

$\delta\omega_{12}$ $\delta\omega_{12}$

$|S\rangle$ $|AS\rangle$

Figure 3.12: Variation of $\Gamma_{12}/\Gamma_{11}$ as a function of the distance $\Delta z$ between two emitters, in a lossless ($\epsilon = -50$), single-mode wire of radius $R = 0.01\lambda_0$. The emitter - wire axis distance is $r_A = 0.015\lambda_0$. The oscillations result in the alternation between super- and subradiance of the symmetric and antisymmetric atomic transitions. All distances are scaled by the vacuum radiation wavelength $\lambda_0$. 

71
Figure 3.13: Variation of $\Gamma_{12}/\Gamma_{11}$ as a function of the distance $\Delta z$ between two emitters, in a single-mode, lossy wire ($\epsilon = -50 + 0.6i$) of radius $R = 0.01\lambda_0$. The emitter - wire axis distance is $r_A = 0.015\lambda_0$. The oscillation is damped because of the dissipation of plasmons upon propagation. All distances are scaled by the vacuum radiation wavelength $\lambda_0$.

Fig. 3.12 and Fig. 3.13 show the value of the cross relaxation rate $\Gamma_{12}$ normalized to the single-atom decay rate $\Gamma_{11} = \Gamma_{22}$, where we have varied the distance between the two atoms by several vacuum wavelengths while keeping the atom - wire distance constant, with metal losses switched off and on, respectively. We observe an oscillatory behaviour of $\Gamma_{12}$ between $\pm\Gamma_{11}$, where the period of oscillations matches exactly the longitudinal wavelength of the surface plasmon mode. In essence, this effect is caused by the interference of the radiation emitted by the two atoms. If the distance between the atoms is an integer number of the plasmon wavelength the emission from the two atoms will interfere constructively in the symmetric state $|S\rangle$. The decay rate $\Gamma_{11} + \Gamma_{12}$ for $|S\rangle$ will thus increase relative to the single atom decay rate ($\Gamma_{12} > 0$). Similarly there will be destructive interference of the emission from the antisymmetric state $|AS\rangle$ giving rise to a decreased emission rate $\Gamma_{11} - \Gamma_{12}$. If on the other hand the distance between the atoms is a half integer number of wavelengths the constructive interference is from the antisymmetric state $|AS\rangle$ corresponding to $\Gamma_{12} < 0$. Note that the physical decay rates (eigenvalues of the decay matrix) given by $\Gamma_{11} \pm \Gamma_{12}$ are always non-negative despite the sign of $\Gamma_{12}$. Besides, one can see in Fig. 3.12 that the maximum value of $\Gamma_{12}$ is very close to $\Gamma_{11}$ which means an efficient enhancement/suppression of spontaneous decay of the corresponding transitions. Therefore, the atoms are indeed strongly coupled to the guided mode - not only is the overall decay rate greatly enhanced but almost the whole of the radiated energy is carried by the plasmons. If there are losses in the metal, however, the oscillations are damped as shown in Fig. 3.13.
3.6. PLASMON-MEDIATED INTERACTION OF TWO EMITTERS: DICKE SUPERRADIANCE

3.6.1 Surface plasmon propagation length and decay length of superradiance

Introducing losses in the metallic nanowire ($\varepsilon'' \neq 0$), we observe a broadening of the plasmonic resonance around $k_z = k_z(\omega; n)$ (predicted by the mode equation (3.50)). At the same time, while the amplitude of the oscillation in $\Gamma_{12}/\Gamma_{11}$ is constant in case of a lossless environment, it shows a decay with the increase of the inter-emitter distances by finite losses, as seen in Fig. 3.13.

The decay rate $\Gamma_{11}$ of a single emitter near the metallic nanowire, as well as the coupling $\Gamma_{12}$ between two emitters are determined by the imaginary part of the dyadic Green’s function, as seen in (3.27). To calculate the Green’s tensor for a cylindrically symmetric system (see Appendix 2) one has to perform an integral and a summation over an analytic expression:

$$\tilde{G}(r, r', \omega) = \int_{-\infty}^{\infty} dk_z \sum_{n=0}^{\infty} \tilde{G}^{(n)}(r, r', \omega; k_z)$$

(3.81)

where $k_z$ is the wave vector component parallel to the axis of symmetry and $n$ is the index of the cylindrical order. From the structure of $\tilde{G}^{(n)}(r, r', \omega; k_z)$ we find that if the radial and polar coordinates of the source point $r'$ and observation point $r$ are pairwise the same, i.e., $r = r'$ and $\phi = \phi'$

$$\tilde{G}^{(n)}(r, r', \omega; k_z) = e^{ik_z|z-z'|} \tilde{G}^{(n)}(r, r, \omega; k_z).$$

(3.82)

We also know, because of the translational symmetry of the system in the $z$ direction, that $\tilde{G}^{(n)}(r, r', \omega; k_z)$ is symmetric in the argument $k_z$. Thus, for atoms polarized in the radial direction, we can write

$$\text{Im} \left[ G_{rr}(r, r', \omega) \right] = \int_{-\infty}^{\infty} dk_z \cos(k_z \Delta z) \text{Im} \left[ \sum_{n=0}^{\infty} G_{rr}^{(n)}(r, r, \omega; k_z) \right]$$

(3.83)

where $G_{rr}(r, r', \omega) = \hat{r}^T \cdot \tilde{G}(r, r', \omega) \cdot \hat{r}$, with $\hat{r}$ being the radial unit vector. Additionally, we denote $\Delta z = |z - z'|$. From this point on in the discussion, for the sake of simplicity, we omit the spatial and frequency variables in the arguments of the Green’s functions, using the notation $G_{rr}^{(n)}(k_z)$.

Let us consider the case when the wire radius is small enough so that only the $n = 0$ plasmon mode is supported (see Fig. 3.3) since this is the regime where strong atom-plasmon coupling is achieved. In case of finite losses in the metal ($\varepsilon'' \neq 0$), we get a broadening in $\text{Im} \left[ G_{rr}^{(n)}(k_z) \right]$ around $k_z = \pm k_z(\omega; n)$, as seen in Fig. 3.3 in case of a thin wire, for $n = 0$.

As we can see in Fig. 3.14, one can neatly fit a Lorentzian function to this peak. Thus, we have an analytic approximation for $\text{Im} \left[ G_{rr}^{(0)}(k_z) \right]$:

$$\text{Im} \left[ G_{rr}^{(0)}(k_z) \right] \approx A \gamma^2 \left[ \frac{1}{\gamma^2 + (k_z - k_z^{SP})^2} + \frac{1}{\gamma^2 + (k_z + k_z^{SP})^2} \right]$$

(3.84)

where $A$ and $\gamma$ are the fitting parameters and $k_z^{SP} = k_z(\omega; 0)$.
CHAPTER 3. COLLECTIVE BEHAVIOUR OF QUANTUM EMITTERS COUPLED TO AN ENGINEERED RESERVOIR

Figure 3.14: Blue line: $\text{Im}\left[ G_{tt}^{(0)}(k_z) \right]$ around the plasmonic resonance as a function of $k_z \lambda_0$, in case of wire radius $R = 0.01 \lambda_0$, atom - wire axis distance $r_A = 0.015 \lambda_0$, electric permittivity $\epsilon = -50 + 0.6i$ (value at $\lambda_0 = 1 \mu m$). Red circles: Lorentzian fit on the peak, $A/(1 + (k_z - k_z^{SP})^2/\gamma^2)$ where we get as parameters $A = 129.54$, $k_z^{SP} = 15.137/\lambda_0$, $\gamma = 0.093$.

Considering the plasmonic part only, we approximate $\text{Im}\left[ G_{tt}^{(0)}(k_z) \right]$ with the Lorentzians (3.84) so we can easily perform the required integrations in (3.83) for $n = 0$ analytically.

$$I^L = \frac{1}{2} \int_{-\infty}^{\infty} dk_z \left( e^{ik_z \Delta z} + e^{-ik_z \Delta z} \right) A \gamma^2 \left[ \frac{1}{\gamma^2 + (k_z - k_z^{SP})^2} + \frac{1}{\gamma^2 + (k_z + k_z^{SP})^2} \right].$$ (3.85)

We get the result for $I^L$ by means of complex integration. Choosing the integration contours properly, we can rewrite the integral over the real axis as closed contour complex integrals and use Cauchy’s integral theorem. Since $\Delta z > 0$, for the term containing $\exp(i k_z \Delta z)$ we have to deform the countour upwards, and for the one containing $\exp(-i k_z \Delta z)$ we close the contour on the lower complex half plane:

$$\int_{-\infty}^{\infty} dk_z \left\{ \frac{e^{ik_z \Delta z}}{\gamma^2 + (k_z - k_z^{SP})^2} \right\} = \frac{A \gamma^2}{2i} \int_{\gamma} d\phi \frac{e^{ik_z \Delta z}}{\gamma^2 + (k_z + k_z^{SP})^2} \left( \frac{1}{k_z - k_z^{SP} - i \gamma} - \frac{1}{k_z + k_z^{SP} + i \gamma} \right),$$ (3.86)

Summing up the contributions of the poles that fall inside the contours we get

$$I^L = 2A \gamma \pi \cos(k_z^{SP} \Delta z) e^{-\gamma \Delta z}.$$ (3.87)
Figure 3.15: The dashed blue line indicates the behaviour of $\text{Im}[G^{(0)}_{rr}(k_z)]$ having a plasmonic peak in the near-evanescent region. The green line is the sum of higher-order contributions ($n = 1...20$) showing a broad, far-evanescent background of dissipative local currents - as a function of $k_z \lambda_0$. The plots are made using $R = 0.01 \lambda_0$ for the wire radius, $r_A = 0.015 \lambda_0$ for the atom - wire axis distance and $\epsilon = -50 + 0.6i$ for relative electric permittivity (value at $\lambda_0 = 1\mu m$).

With this result, the Lorentzian-fitted plasmonic contributions to $\Gamma_{11}$ and $\Gamma_{12}$ assume the simple, approximate form

$$\Gamma_{11}^L = \frac{2\omega_A^2 d^2}{\hbar \epsilon_0 c^2} 2A\gamma \pi$$
$$\Gamma_{12}^L = \frac{2\omega_A^2 d^2}{\hbar \epsilon_0 c^2} 2A\gamma \pi \cos(k_z S P \Delta z)e^{-\gamma \Delta z}.$$  \hspace{1cm} (3.88)

The results show that the amplitude of the atom-atom coupling decays at a rate identical to the broadening of the plasmonic resonance, i.e., the decay length of the coupling and the absorption length for the plasmons are the same.

However, even in the case of a thin wire, this is not the only contribution to the Green’s tensor: Fig.3.15 shows contributions in the traveling-wave region ($|k_z| < k_0$) as well as in the far-evanescent region; the latter is due to the presence of dissipative local currents. To prove the validity of our analytic approximation, we compared the analytic results with those of the numerical integration of the full problem, as seen in Figs.3.16 and 3.17. One can observe that $\Gamma_{12}^L/\Gamma_{11}^L$ follows $\Gamma_{12}/\Gamma_{11}$ which also contains the traveling-wave and far-evanescent contributions. This confirms that, indeed, the plasmonic contribution is far the most significant factor in the coupling between the emitters in case of a thin, single-mode wire.
Figure 3.16: The atom-atom coupling normalized by the single-atom decay rate ($\Gamma_{12}/\Gamma_{11}$, blue dashed line) is well approximated by $\Gamma_{12}^L/\Gamma_{11}^L$ (red line) obtained by fitting a Lorentzian on the plasmonic peak. The green dash-dotted line shows the exponential decay of the oscillations ($e^{-\gamma\Delta z}$) as a function of inter-emitter distance $\Delta z/\lambda_0$. The calculations were done using $R = 0.01\lambda_0$, $r_A = 0.015\lambda_0$, $\epsilon = -50 + 0.6i$ (value at $\lambda_0 = 1\mu m$), $k_{zP}^S = 15.137/\lambda_0$ and $\gamma = 0.093$.

Figure 3.17: A magnified view of Fig. 3.16 at one of the peaks in the oscillation of $\Gamma_{12}/\Gamma_{11}$ for the better illustration of the difference between the exact and the Lorentz-approximated result.
Deviations of the Lorentz-approximated result from $\Gamma_{12}/\Gamma_{11}$ are more distinctly observable at the peaks, the reason for which is that the additional contributions (traveling waves and local dissipative currents) in $\text{Im}[G_{rr}(k_z)]$ enhance the value of $\Gamma_{11}$ but not that of $\Gamma_{12}$ i.e., their influence is mainly local so they only negligibly contribute to the atom-atom coupling. As a result, $\Gamma_{12}/\Gamma_{11}$ will not assume as high peak values as $\Gamma^L_{12}/\Gamma^L_{11}$ does. Fig. 3.16 also shows that the decay length of the superradiance phenomenon over the inter-emitter distance $\Delta z$ for the full problem is, as expected, the same as the absorption length of a single plasmon.

### 3.6.2 Optimizing the atom-atom coupling

Finite material losses, i.e. $\epsilon'' \neq 0$ give rise to another effect related to the discussion in Section 3.5.3. If we put an emitter too close to the wire surface, non-radiative losses become increasingly important. Thus, although the coupling strength to the plasmons increases, the non-radiative channel will take over. Therefore, as Fig. 3.18 shows, there is, again, an optimal distance of the emitters, where the contrast of the super- and subradiance is maximal.

![Figure 3.18: Magnitude of an extremal value of $\Gamma_{12}/\Gamma_{11}$ as the function of the atom - wire axis distance $r_A$, scaled by the vacuum radiation wavelength $\lambda_0$, at wire radius $R = 0.01\lambda_0$ and inter-atomic distance $\Delta z = 1.01\lambda_0$. We use $\epsilon = -50 + 0.6i$. There is an optimal $r_A$ where the coupling to the guided mode is maximal, resulting in a minimal damping of the oscillations of $\Gamma_{12}/\Gamma_{11}$.](image_url)

The efficiency of the coupling between two emitters separated by a fixed distance $\Delta z$ depends on several factors. In order to maximize the single-atom plasmon coupling the atom should be placed as close as possible to the wire surface. Since at some point non-radiative losses of the single atom increase more rapidly when approaching the surface, there is an optimum atom-wire distance for a given wire radius $R$. 

77
On the other hand the propagating plasmons also experience absorption originating from the same material losses. These absorption losses depend on the structure of the mode function and thus also on the wire radius $R$. We have examined whether for a fixed distance $d$ along the wire there is a wire radius allowing an optimal compromise between these two effects. In Fig. 3.19 we plot the enveloping curve of the maxima of $\Gamma_{12}/\Gamma_{11}$ as a function of inter-atomic distance, in case of three different wire radii. In each case we used a wire-emitter distance that allowed an optimal atom-plasmon coupling. One can see that at a smaller radius $R$, the achievable maximum of $\Gamma_{12}/\Gamma_{11}$ is higher. However, as the propagation distance increases, the value of the envelope decreases faster compared to the case of a larger $R$. According to this, if the emitters are close to each other, it is better to have a wire with a smaller radius. On the other hand, if they are further apart the optimal radius would be a larger one. There is thus an optimal wire radius for a given inter-atomic distance, but there is no single optimal $R$ for all cases.

The superradiance effect is sensitive to the positioning of the atoms, so the experimental realization with a given atoms distance may be technically demanding. A possible method could be to use atoms trapped in very deep trapping potentials possibly created by an evanescent standing wave field surrounding the nanowire [87]. Alternatively one could use impurities in a solid state material acting as artificial atoms. Ideally such impurities could be implanted at a well determined position [88], but alternatively the effects discussed here could also be observed by post-selecting wires where, e.g., the random position of impurities match the desired distance.
3.7 Application for a phase gate

We have seen in Sec. 3.6 that a thin nano-wire, acting as a quasi-1D reservoir, ensures strong effective atom-atom interaction between two quantum emitters even if they are several wavelengths apart. Thus, the emitters, besides having a strong interaction, are also individually addressable with external light fields - in contrast with, for example, optical cavities where, although the atoms are strongly coupled to the cavity mode, they cannot be spatially resolved by an external resonant field.

As a possible application, we are going to analyze in the following the realization of a deterministic quantum phase gate between two lambda atoms coupled by a single surface plasmon mode of the nanowire, as shown in Fig. 3.20.

![Figure 3.20: Realization of a deterministic quantum phase gate, by coupling the $|e\rangle - |g\rangle$ transitions of two lambda atoms to $n = 0$ surface plasmon mode, inducing superradiance in the two-atom system, and applying external classical $2\pi$ pulses.](image)

We assume that only the $|g\rangle \rightarrow |e\rangle$ transition of each emitter is coupled to the single guided mode of the wire. If the two emitters are several wavelengths apart they can individually be addressed by lasers coupling the $|g\rangle \rightarrow |e\rangle$ transition. The phase gate works by exploiting the large difference in the decay rates between the super- and subradiant states shown in Fig. 3.13.

Let us first assume that we can ignore the decay of the excited level $|e\rangle$ in the $|g\rangle - |e\rangle$ two level system. In this case it is well known that a resonant $2\pi$ pulse, forcing the system to do a full Rabi oscillation, gives an additional $\pi$ phase to the atomic system. On the other hand, if the decay rate of the excited state $\Gamma_{eg}$ is much stronger than the resonant Rabi frequency, i.e. $\Omega \ll \Gamma$, the driving field cannot induce Rabi oscillations between the two levels, and the drive merely introduces a scattering rate $\propto \Omega^2/\Gamma_{eg}$, which vanishes in the limit of weak driving or strong decay. The idea behind the phase gate is that due to the subradiant effect we saw in Sec. 3.6 the interaction of a nearby emitter can change the excited state from being decaying into being non-decaying, thereby allowing the first emitter to pick up a $\pi$ phase shift conditioned on the state of the other.
Figure 3.21: Effective level scheme of two, externally driven, interacting 3-level lambda atoms, seen in Fig. 3.20. $\Omega_S$ and $\Omega_{AS}$ are, respectively, the symmetric and antisymmetric superpositions of $\Omega_1$ and $\Omega_2$. Because of the strong coupling to a single plasmonic mode, a high-contrast superradiance effect is present in the system, inducing a slow decay in the symmetric transition and a fast one in the antisymmetric transition.

Specifically, consider the level scheme of the two-atom system shown in Fig. 3.21. The ground and excited state of atoms 1 and 2 are coupled by real resonant classical drive fields with a strength parametrized by the Rabi frequencies $\Omega_1$ and $\Omega_2$. Thus, the interaction Hamiltonian now contains the emitter-reservoir interaction as well as the effect of the two classical external fields:

$$\hat{H}_I = -\sum_{j=1}^{2} \left\{ |e\rangle_j \langle s| d_{e\bar{s}} \hat{E}^{(+)}(\mathbf{r}_j) + |e\rangle_j \langle g| d_{e\bar{g}} \hat{E}^{(+)}(\mathbf{r}_j) + \Omega_j |e\rangle_j \langle g| + \text{H.c.} \right\}$$  \hspace{1cm} (3.89)

where we choose $d_{e\bar{g}}$ to point in the radial direction so that the $|g\rangle \rightarrow |e\rangle$ transitions in both atoms are maximally coupled to the plasmonic modes of the wire, whereas $d_{e\bar{s}}$ points in the tangential direction, i.e., the $|s\rangle \rightarrow |e\rangle$ transitions are decoupled from the plasmons. In this way, only the decay rates of collective states $|eg\rangle$, $|ge\rangle$ and $|ee\rangle$ are enhanced. We approximate the decay rate of the other states by their vacuum values because their alteration due to the presence of the wire is negligibly small.

The classical fields $\Omega_1$ and $\Omega_2$ couple the states $|gg\rangle$, $|eg\rangle$, $|ge\rangle$ and $|ee\rangle$ of the combined system but it is desirable to express the interaction in the basis involving the states $|S\rangle = (|eg\rangle + |ge\rangle)/\sqrt{2}$ and $|AS\rangle = (|eg\rangle - |ge\rangle)/\sqrt{2}$, because these states have a simpler decay dynamics, c.f., Eqs. (3.76) and (3.77).
In this case one gets the couplings shown in Fig. 3.21 with the effective coupling fields

\begin{align}
\Omega_S &= \frac{1}{\sqrt{2}}(\Omega_1 + \Omega_2) \\
\Omega_{AS} &= \frac{1}{\sqrt{2}}(\Omega_1 - \Omega_2).
\end{align}

Assume now that the \(\Delta z\) distance between the atoms is such that the symmetric state \(|S\rangle\) is subradiant, whereas the antisymmetric state \(|AS\rangle\) is superradiant \(\Gamma_{AS} \gg \Gamma_S\) and that \(\Omega_2 = \Omega_1\) corresponding to \(\Omega_{AS} = 0\) and \(\Omega_S = \sqrt{2}\Omega_1\). Choosing the driving strength \(\Omega_S\) to be in between the two decay rates

\(\Gamma_{AS} \gg \Omega_S \gg \Gamma_S\)

ensures that transition to the double excited state \(|ee\rangle\) is blocked by the strong decay \(\Gamma_{AS}\), whereas we can perform a \(2\pi\) Rabi oscillation on the two level system given by \(|gg\rangle\) and \(|S\rangle\) (see Fig. 3.21). This ensures that we achieve a phase change of \(\pi\) on the \(|gg\rangle\) state. Starting from the states \(|gs\rangle\) and \(|sg\rangle\) there is essentially only a single-atom interaction with the wire, since there is no coupling to the state \(|s\rangle\). Therefore, the excited state has always a fast decay \(\Gamma_{11} \sim \Gamma_{AS}/2\) such that the driving is too weak to excite the atoms. Furthermore the state \(|ss\rangle\) is completely unaffected by the classical light pulses and in the ideal limit the entire process will have the truth table

\begin{align*}
|ss\rangle &\rightarrow |ss\rangle \\
|sg\rangle &\rightarrow |sg\rangle \\
|gs\rangle &\rightarrow |gs\rangle \\
|gg\rangle &\rightarrow -|gg\rangle.
\end{align*}

In reality the finite ratio \(\Gamma_{AS}/\Gamma_S\) will limit how well one can fulfill the condition in Eq. (3.92) and this will limit the fidelity \(F\) of the gate. Choosing the Rabi frequency too large will allow scattering from the transitions which are supposed to be blocked by the fast decay of the excited states with decay rates \(\propto \Gamma_{eg}\). The population getting into the fast decaying states can be estimated as \(\Omega_1^2/\Gamma_S^2 \propto \Omega_{1eg}^2/\Gamma_{eg}^2\). To get the magnitude of population decaying out of these states during the interaction, the full population getting into the states must be multiplied by \(\Gamma_{eg} T_{\text{pulse}}\) where \(T_{\text{pulse}}\) is the duration of the external \(2\pi\) pulse. Thus, the imperfection resulting from this process scales as

\[X_1 = \frac{\Omega_1^2}{\Gamma_{eg}^2} \Gamma_{eg} T_{\text{pulse}} \propto \frac{\Omega_1}{\Gamma_{eg}}\]

because, being a \(2\pi\) pulse, \(T_{\text{pulse}} \propto 1/\Omega_1\).

On the other hand, choosing the external driving field too small will lead to a decay from the slowly-decaying excited state during the \(2\pi\) pulse on the \(|gg\rangle - |S\rangle\) transition. The whole population
starting in \( |gg\rangle \) goes through the state \( |S\rangle \), and the decay out of it scales as \( \Gamma_S T_{\text{pulse}} \) resulting in

\[
X_2 = \Gamma_S T_{\text{pulse}} \propto \frac{\Gamma_S}{\Omega_1}.
\]

(3.94)

For the total imperfection we get

\[
1 - F \sim X_1 + X_2 = \frac{\Omega_1}{\Gamma_{eg}} + \frac{\Gamma_S}{\Omega_1}.
\]

(3.95)

Minimizing (3.95) with respect to \( \Omega_1 \), we find that the minimal imperfection scales as

\[
1 - F_{opt} \sim \sqrt{\frac{\Gamma_S}{\Gamma_{eg}}}.
\]

(3.96)

To confirm this rough scaling analysis we show in Fig. 3.22 the result of a full numerical simulation of the density matrix equation for the system.

Figure 3.22: Fidelity of a maximally entangled state created by the phase gate optimized with respect to the drive strength. The phase gate fidelity error scales roughly as \( \sqrt{\Gamma_S/\Gamma_{eg}} \) (dashed line) in the limit \( F \to 1 \).

We can write the fidelity of the gate operation as the square of the absolute value of the overlap between the desired final atomic state and the one we get in the end:

\[
F = |\langle \psi_{\text{ideal}} | \psi \rangle|^2 = \langle \psi_{\text{ideal}} | \psi \rangle \langle \psi | \psi_{\text{ideal}} \rangle
\]

(3.97)
3.7. APPLICATION FOR A PHASE GATE

which is the expectation value of the final density operator taken in the desired atomic state:

\[ F = \langle \psi_{\text{ideal}} | \hat{\rho} | \psi_{\text{ideal}} \rangle. \]  

(3.98)

Using \( |\psi_{\text{initial}}\rangle = 1/2(|ss\rangle + |sg\rangle + |gs\rangle + |gg\rangle) \) as a starting state, we show in Fig. 3.22 the error \( 1 - F \) where \( F \) is optimized over the driving strength \( \Omega \) as a function of the ratio of the decay rates \( \Gamma_S/\Gamma_{eg} \). Here \( |\psi_{\text{ideal}}\rangle = 1/2(|ss\rangle + |sg\rangle + |gs\rangle - |gg\rangle) \) is a maximally entangled state created using the phase gate combined with ideal \( \pi/2 \) pulses on the \( |g\rangle - |s\rangle \) transitions. The dashed line shows the scaling derived above \( 1 - F = c \sqrt{\Gamma_S/\Gamma_{eg}} \), with the constant \( c \) chosen to match the behavior as \( F \to 1 \).

The analysis of the phase gate has so far not been tied to any particular system, but applies to any system where there is a large difference in the decay rates of the super- and sub-radiant states. We now turn to the specific implementation in terms of atoms coupled through the \( n = 0 \) plasmon mode. As we have seen above, the subradiant state does not have a completely vanishing decay rate and, accordingly, this will limit the fidelity of the gate. In Fig. 3.23, we show the optimal fidelity of the gate operation, optimized by varying \( \Omega_S \), as a function of the atom - wire axis distance \( r_A \). For the simulation, we used \( R = 0.003 \lambda_0 \) and \( \Delta z = 0.08 \lambda_0 \).

![Figure 3.23: Optimal fidelity of a maximally entangled state generated with the phase gate as a function of the atom - wire axis distance \( r_A \). \( r_A \) is scaled with the vacuum radiation wavelength \( \lambda_0 \). The two-atom states \( |ss\rangle, |sg\rangle, |gs\rangle \) and \( |gg\rangle \) are initially equally populated. We have used the parameters \( R = 0.003 \lambda_0 \), \( \Delta z = 0.08 \lambda_0 \), \( \epsilon = -50 + 0.6i \) in the simulation.]

The results show a maximum fidelity of 80% for the parameters above. There are two major detrimental effects which reduce the superradiance-subradiance contrast and, consequently, the gate fidelity, namely, the coupling to free space and the wire losses. Due to the free space coupling, a
small amount of the radiated energy will be scattered into the far-field region, resulting in a non-zero contribution to $\Gamma_S$. The wire losses induce traveling-wave and circulating current dissipation leading to an additional, local loss of energy. This also contributes to $\Gamma_S$. It is possible to suppress the spontaneous emission into free space, for example, by placing the atoms into a photonic bandgap material. This would considerably enhance the gate fidelity only if the free-space contribution to $\Gamma_S$ is dominant with respect to the wire-loss contribution. Fig. 3.24 shows the ratio $\Gamma_{wire}^S / \Gamma_{free}^S$ as a function of $\Delta z' / \lambda_0$, for distances $\Delta z$ corresponding to the minima of $\Gamma_{12}/\Gamma_{11}$. As one can see, to increase the inter-emitter distance $\Delta z$ while keeping $\Gamma_{wire}^S / \Gamma_{free}^S < 1$, we need to decrease $\Delta z'$. For the given parameters, this means that, for instance, for $d \approx 2 \lambda_0$ we have to have $\Delta z' \leq 0.1$.

### 3.8 Collective behaviour of three emitters coupled by plasmons

Increasing the number of atoms coupled to the surface plasmons results in an increasingly rich atomic subspace of the Hilbert space. Apart from changing the number of atoms in the system, one can also manipulate the collective behaviour by engineering the reservoir, i.e., the effective couplings between the atoms. Allowing the presence of classical external fields, the interaction Hamiltonian assumes the form

$$
\hat{H}_I = -\sum_{j=1}^{3} \left\{ |e\rangle_j \langle d_j| \hat{E}^{(+)}(\mathbf{r}_j) + |e\rangle_j \langle g| \Omega_j + \text{H.c.} \right\}
$$

(3.99)
where $\hat{E}^{(+)}(r_j)$ is the electric field operator defined in (3.12) and $\Omega_j$ are resonant external classical fields at the position of the $j$-th atom.

In the following, we consider the physical phenomena arising in a system that consists of three quantum emitters, coupled either by a single nano-wire or three of them in the shape of a triangle. We shall see that the change in the coupling geometry leads to substantial modification of the collective behaviour.

Note that because our analytic predictions heavily rely on the periodicity of $\Gamma_{12}$ in the absence of losses, as seen in Fig. 3.12 and Eq. (3.88), we set the ohmic losses to zero in the following. Introducing losses will only produce minor changes in the qualitative behaviour of the system.

### 3.8.1 Three emitters coupled by a single wire

Let us have a look at the system where three two-level atoms along a thin wire are coupled to the $n = 0$ surface plasmon mode, as illustrated in Fig. 3.25.

![Figure 3.25](image)

Figure 3.25: Setup for three identical emitters coupled by a single, thin metallic nano-wire, all of them located at a distance $r_A$ from the symmetry axis of the wire, having a radius $R$. The distance between the $m$-th and $n$-th atom is denoted by $\Delta z_{mn}$. Besides exchanging excitations with the mediation of surface plasmon modes of the wire, one can also manipulate them by external classical electromagnetic fields $\Omega_{1,2,3}$.

In order to find the diagonal basis and collective decay rates we set the classical Rabi frequencies mentioned above to zero and follow the procedure described in Sec. 3.3 with $N = 3$. As a result we get the master equation with the usual shifts, single-atom decay rates and coupling terms $\delta \omega_{kl}, \Gamma_{kl}, k,l = 1, 2, 3$. As usual, we start from the diagonal basis for the non-interacting atomic system:

$$\{ |ggg\rangle, |egg\rangle, |geg\rangle, |eeg\rangle, |ege\rangle, |gee\rangle, |eee\rangle \}.$$  

Having three atoms, we have to find the diagonal basis in the single-, double-, and triple-excitation manifolds from which the triple-excitation diagonal basis state is $|eee\rangle$. The elements of the single-
and double-excitation bases depend on the actual arrangement of the atoms, so one has to calculate them for each particular setup individually, following the treatment in Sec. 3.3. For a general spacing distribution between the atoms, one usually gets complicated expressions for the collective decay rates, none of which is usually subradiant. Because of the periodic behaviour of $\Gamma_{12}$ seen in Sec. 3.6 there are only certain atomic arrangements where subradiant states emerge. Therefore, instead of trying to give a general solution for the diagonal basis and the collective decay rates, we discuss particular configurations where one is likely to find entangled, subradiant states.

Let us first consider a setup where the inter-emitter distances are

$$\Delta z_{12} = p\lambda_{SP}$$
$$\Delta z_{23} = q\lambda_{SP}$$

(3.101)

where $\Delta z_{kl}$ is the distance between atoms $k$ and $l$ and $\lambda_{SP} = 2\pi/k_z^{SP}$ is the longitudinal wavelength associated with the $n = 0$ plasmon mode and $p, q$ are positive integer numbers. Also, let us assume that the atoms are at the same distance from the wire surface. Consequently, the single-atom decay rates and atom-atom couplings are ($\Gamma_{12} > 0$)

$$\Gamma_{11} = \Gamma_{22} = \Gamma_{33}$$
$$\Gamma_{12} = \Gamma_{23} = \Gamma_{13} \approx \Gamma_{11}.$$  

(3.102)

Following the notation in Sec. 3.3.1 for a diagonal basis we get

$$|1, 0\rangle = |ggg\rangle$$
$$|1, 1\rangle = \frac{1}{\sqrt{3}} (|egg\rangle + |geg\rangle + |gee\rangle)$$
$$|2, 1\rangle = \frac{1}{\sqrt{2}} (|egg\rangle + |gee\rangle)$$
$$|3, 1\rangle = \frac{1}{\sqrt{6}} (|egg\rangle + 2|geg\rangle - |gee\rangle)$$

$$|1, 2\rangle = \frac{1}{\sqrt{3}} (|eeg\rangle + |ege\rangle + |gee\rangle)$$
$$|2, 2\rangle = \frac{1}{\sqrt{2}} (-|eeg\rangle + |gee\rangle)$$
$$|3, 2\rangle = \frac{1}{\sqrt{6}} (-|eeg\rangle + 2|ege\rangle - |gee\rangle)$$

(3.103)

for which states the emerging collective decay rates are

$$|1, 0\rangle \rightarrow 0$$
$$|1, 1\rangle \rightarrow (\Gamma_{11} + 2\Gamma_{12})$$
$$|1, 2\rangle \rightarrow (2\Gamma_{11} + 2\Gamma_{12})$$
$$|2, 1\rangle \rightarrow (\Gamma_{11} - \Gamma_{12})$$
$$|2, 2\rangle \rightarrow (2\Gamma_{11} - \Gamma_{12})$$
$$|3, 2\rangle \rightarrow (2\Gamma_{11} - \Gamma_{12})$$

$$|1, 3\rangle \rightarrow 3\Gamma_{11}.$$  

(3.104)

We stress again that for the given inter-atomic distances $\Gamma_{12} > 0$. As one immediately sees, under the given circumstances the states $|2, 1\rangle$ and $|3, 1\rangle$ of the single-excitation manifold are subradiant, entangled states, the former containing two-partite, the latter containing three-partite entanglement.
We can vary the relative phase between the original basis state components making up the subradiant states by changing the spacing between the atoms. For example, we can place them at distances

\[
\Delta z_{12} = (1 + p)\lambda^{SP} \quad \Delta z_{23} = \left(1 + \frac{q}{2}\right)\lambda^{SP}, \quad p, q = 0, 1, 2, ...
\]  

(3.105)

The couplings in this case are \((\Gamma_{12} > 0)\)

\[
\Gamma_{12} = -\Gamma_{23} = -\Gamma_{13} \approx \Gamma_{11}.
\]  

(3.106)

We get subradiant diagonal basis states again solely in the single-excitation manifold, the basis states of which are

\[
|1,1\rangle = \frac{1}{\sqrt{3}} (-|egg\rangle - |geg\rangle + |gge\rangle) \quad |2,1\rangle = \frac{1}{\sqrt{2}} (|egg\rangle + |gge\rangle) \quad |3,1\rangle = \frac{1}{\sqrt{6}} (-|egg\rangle + 2|geg\rangle + |gge\rangle),
\]  

(3.107)

having identical decay rates to the former configuration:

\[
|1,1\rangle \rightarrow (\Gamma_{11} + 2\Gamma_{12}) \quad |2,1\rangle \rightarrow (\Gamma_{11} - \Gamma_{12}) \quad |3,1\rangle \rightarrow (\Gamma_{11} - \Gamma_{12}).
\]  

(3.108)

Choosing the distances to be

\[
\Delta z_{12} = \Delta z_{23} = \left(\frac{1}{2} + p\right)\lambda^{SP}, \quad p = 0, 1, 2, ...
\]  

(3.109)

and so the couplings \(\Gamma_{12} = \Gamma_{23} = -\Gamma_{13} \approx -\Gamma_{11}\), we can set the components of \(|3,1\rangle\) in phase with each other, having the diagonal basis states of the single-excitation manifold

\[
|1,1\rangle = \frac{1}{\sqrt{3}} (|egg\rangle - |geg\rangle + |gge\rangle) \quad |2,1\rangle = \frac{1}{\sqrt{2}} (-|egg\rangle + |gge\rangle) \quad |3,1\rangle = \frac{1}{\sqrt{6}} (|egg\rangle + 2|geg\rangle + |gge\rangle),
\]  

(3.110)

having the same collective decay rates as \((3.108)\).
CHAPTER 3. COLLECTIVE BEHAVIOUR OF QUANTUM EMITTERS COUPLED TO AN ENGINEERED RESERVOIR

Placing the emitters appropriately, one can also decouple atoms from the rest. For example, having

\[ \Delta z_{12} = (1 + p) \lambda^{SP} \]
\[ \Delta z_{23} = \left( \frac{1}{4} + q \right) \lambda^{SP}, \quad p, q = 0, 1, 2, ... \]  

(3.111)

results in couplings

\[ \Gamma_{12} \approx \Gamma_{11} \]
\[ \Gamma_{23} = \Gamma_{13} = 0. \]  

(3.112)

This leads to the decoupling of the third atom from the others:

\[ |1, 0 \rangle = |ggg \rangle \]
\[ |1, 1 \rangle = \frac{1}{\sqrt{2}} \left( |-egg \rangle + |gee \rangle \right) \]
\[ |1, 2 \rangle = \frac{1}{\sqrt{2}} \left( |ege \rangle + |gee \rangle \right) \]
\[ |2, 1 \rangle = \frac{1}{\sqrt{2}} \left( |egg \rangle + |geg \rangle \right) \]
\[ |3, 1 \rangle = |gee \rangle \]
\[ |3, 2 \rangle = |eeg \rangle \]
\[ |2, 2 \rangle = \frac{1}{\sqrt{2}} \left( |ege \rangle + |gee \rangle \right) \]
\[ |1, 3 \rangle = |eeg \rangle \]
\[ |3, 3 \rangle = |eee \rangle \]  

(3.113)

where, as we can see, the state of the third atom factors out, leaving the system with symmetric and antisymmetric two-partite entangled states, similar to those in Sec. 3.6. For the collective decay rates we get

\[ |1, 0 \rangle \to 0 \quad |1, 2 \rangle \to (2\Gamma_{11} - \Gamma_{12}) \]
\[ |1, 1 \rangle \to (\Gamma_{11} - \Gamma_{12}) \quad |2, 2 \rangle \to (2\Gamma_{11} + \Gamma_{12}) \]
\[ |2, 1 \rangle \to (\Gamma_{11} + \Gamma_{12}) \quad |3, 2 \rangle \to 2\Gamma_{11} \]
\[ |3, 1 \rangle \to \Gamma_{11} \quad |1, 3 \rangle \to 3\Gamma_{11} \]  

(3.114)

getting a Dicke subradiance for the antisymmetric \(|1, 1\rangle\) state and a superradiance for \(|2, 1\rangle\). If we want to make the symmetric state subradiant, all we have to do is place the emitters as

\[ \Delta z_{12} = \left( \frac{3}{2} + p \right) \lambda^{SP} \]
\[ \Delta z_{23} = \left( \frac{1}{4} + q \right) \lambda^{SP}, \quad p, q = 0, 1, 2, ... \]  

(3.115)

getting couplings

\[ \Gamma_{12} \approx -\Gamma_{11} \]
\[ \Gamma_{23} = \Gamma_{13} = 0 \]  

(3.116)
and so, for the diagonal basis in the single-excitation manifold we get

\[
\begin{align*}
|1, 1\rangle &\rightarrow (\Gamma_{11} + \Gamma_{12}) \\
|2, 1\rangle &\rightarrow (\Gamma_{11} - \Gamma_{12}) \\
|3, 1\rangle &\rightarrow \Gamma_{11}
\end{align*}
\]  

(3.117)

where now the symmetric state is subradiant. We can similarly decouple the second or the first atom and perform the operations described above on the rest of the emitters.

Note that in all cases, because of the periodic behaviour of \(\Gamma_{12}\) for a lossless wire, changing the relative positions of the atoms by integer multiples of \(\lambda^{SP}\) will affect neither the structure of the diagonal basis nor the collective decay rates.

A question one would probably ask at this point is whether there is a way to reliably prepare the atomic ensemble in these subradiant, entangled collective states. For achieving this goal, the combination of resonant external electromagnetic fields with the intrinsic dissipative processes in the system can be an efficient tool.

Consider for example the case when the atoms are placed according to (3.101). Here, the system has two subradiant states, both in the single-excitation manifold: \(|2, 1\rangle\) and \(|3, 1\rangle\). Exciting the atoms into the state \(|eee\rangle\), letting it decay and afterwards post-selecting would not be a good choice because even if they don’t decay to \(|ggg\rangle\), they will end up in a mixture of \(|2, 1\rangle\) and \(|3, 1\rangle\).

Writing up the matrix for the part of the Hamiltonian (3.99) that contains the interaction with the real classical fields \(\Omega_j\) in the basis (3.103) and setting

\[
\begin{align*}
\Omega_1 &= \Omega_3 \\
\Omega_2 &= 0
\end{align*}
\]  

(3.118)

we get

\[
H^d_{cl} = \begin{pmatrix}
0 & \frac{2\Omega_1}{\sqrt{3}} & 0 & -\sqrt{\frac{2}{3}}\Omega_1 & 0 & 0 & 0 & 0 \\
\frac{2\Omega_1}{\sqrt{3}} & 0 & 0 & 0 & \frac{4\Omega_1}{3} & 0 & \frac{\sqrt{2}\Omega_1}{\sqrt{3}} & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
-\sqrt{\frac{2}{3}}\Omega_1 & 0 & 0 & 0 & \frac{\sqrt{2}\Omega_1}{\sqrt{3}} & 0 & 0 & 0 \\
0 & \frac{4\Omega_1}{3} & 0 & \frac{\sqrt{2}\Omega_1}{\sqrt{3}} & 0 & 0 & 0 & \frac{2\Omega_1}{\sqrt{3}} \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & \frac{\sqrt{2}\Omega_1}{\sqrt{3}} & 0 & -\frac{4\Omega_1}{3} & 0 & 0 & 0 & -\sqrt{\frac{2}{3}}\Omega_1 \\
0 & 0 & 0 & 0 & \frac{2\Omega_1}{\sqrt{3}} & 0 & -\sqrt{\frac{2}{3}}\Omega_1 & 0
\end{pmatrix}
\]  

(3.119)
CHAPTER 3. COLLECTIVE BEHAVIOUR OF QUANTUM EMITTERS COUPLED TO AN ENGINEERED RESERVOIR

Figure 3.26: Level scheme in the diagonal basis for the three-atom system when the atoms are coupled by a single wire and their relative distance to each other is $\lambda^{SP}$. For this configuration there are two subradiant, entangled states, one of which can be isolated by choosing the external fields $\Omega_1 = \Omega_3$, $\Omega_2 = 0$. Thus, by optical pumping, the atomic population can be driven into the subradiant, entangled $|2, 1\rangle$.

The linkage between the collective atomic states induced by (3.119) is depicted in Fig. 3.26. Because of the choice of Rabi frequencies according to (3.118), the subradiant $|2, 1\rangle$ state is the only one unaffected by the classical fields in the single-excitation manifold. Thus, simply by optical pumping with the appropriate external fields one can bring the atomic population into a subradiant state where the atom in the middle stays in its ground state and the other two are maximally entangled.

3.8.2 Triangle configuration

We mentioned before that altering the coupling geometry between atoms we can influence the properties of the collective atomic states. Let us now take three nano-wires instead of a single one and arrange the system as seen in Fig. 3.27. In this geometry, a single wire couples only two atoms.

Changing the reservoir, we now have to work with a Green’s tensor different from the one we used for the single wire. However, in this case the cylindrical symmetry of the environment is lost so it is difficult to find the harmonic vector wave functions with which we could exactly fulfill the boundary conditions at the wire surfaces.

Nevertheless, one can make the following approximation. Let us assume that none of the wires is affected by the electromagnetic field reflected from another wire, that is, without the atoms between them, one wire is not affected by the presence of the others. This is a good approximation because we are concerned with plasmonic fields, and they are evanescent in the radial direction, i.e., they have an exponential decrease as the distance from the wire increases - and also they are by far the
3.8. COLLECTIVE BEHAVIOUR OF THREE EMITTERS COUPLED BY PLASMONS

Figure 3.27: a) Coupling three emitters with three thin metallic wires so that each of the atoms is only coupled to two wires at a time. The atoms can be manipulated by resonant external classical fields $\Omega_{1,2,3}$. b) Top view of the same setup.

dominating field contribution in case of thin wires. Thus, for the reflected Green’s tensors we can write

$$\bar{G}_R(r, r', \omega) = \sum_{j=1}^{3} \bar{G}_{R,j}(r, r', \omega)$$

(3.120)

where $\bar{G}_{R,j}$ is the single-wire reflected Green’s tensor for the $j$-th wire.

As in the preceding subsection, we will show examples for collective atomic behaviour with subradiant states in the case of different inter-atomic spacings. We assume that each of the atoms is at the same distance from both of the adjacent wires. Thus, the single-atom decay rates $\Gamma_{kk}$ for the three-wire geometry will be twice the value we got in the single-wire case but, at the same time, the $\Gamma_{12}$ atom-atom couplings will be the same as previously because it is still a single wire that connects two atoms. So now, instead of $\pm \Gamma_{11}$ the limiting values for $\Gamma_{12}$ are

$$-\frac{1}{2} \Gamma_{11} \leq \Gamma_{12} \leq \frac{1}{2} \Gamma_{11}$$

(3.121)

Let us first consider the setup where the spacing of the atoms is

$$\Delta z_{12} = \Delta z_{23} = \Delta z_{13} = (\frac{1}{2} + p)\lambda^{SP}, \quad p = 0, 1, 2, ...$$

(3.122)

For a diagonal basis we get the same as (3.103) but with collective decay rates

$$|1, 0\rangle \rightarrow 0 \quad |1, 2\rangle \rightarrow (2\Gamma_{11} - 2\Gamma_{12})$$
$$|1, 1\rangle \rightarrow (\Gamma_{11} - 2\Gamma_{12}) \quad |2, 2\rangle \rightarrow (2\Gamma_{11} + \Gamma_{12})$$
$$|2, 1\rangle \rightarrow (\Gamma_{11} + \Gamma_{12}) \quad |3, 2\rangle \rightarrow (2\Gamma_{11} + \Gamma_{12})$$
$$|3, 1\rangle \rightarrow (\Gamma_{11} + \Gamma_{12}) \quad |1, 3\rangle \rightarrow 3\Gamma_{11}$$

(3.123)

from which we see that in this geometry the system has a single subradiant state, the symmetric $|1, 1\rangle$ from the single-excitation manifold. $|1, 1\rangle$ has the attractive feature of being a three-partite entangled state, a so-called $W$ state, well-known and used in quantum information theory [89].
Changing the length of the sides of the triangle appropriately we can add phases to the components of $|1, 1\rangle$ while keeping it subradiant, for example let us set

$$\Delta z_{12} = \Delta z_{23} = (1 + p)\lambda^{SP}$$
$$\Delta z_{13} = (1/2 + q)\lambda^{SP}, \quad p, q = 0, 1, 2, ... \quad (3.124)$$

As a result, for the states in the single-excitation manifold we get

$$|1, 1\rangle = \frac{1}{\sqrt{3}}(-|egg\rangle - |geg\rangle + |gge\rangle)$$
$$|2, 1\rangle = \frac{1}{\sqrt{2}}(|egg\rangle + |gge\rangle)$$
$$|3, 1\rangle = \frac{1}{\sqrt{6}}(-|egg\rangle + 2|geg\rangle + |gge\rangle), \quad (3.125)$$

with the respective decay rates in (3.123).

We can also try to decouple one of the atoms from the rest as we did in the case of a single wire: this can be done by choosing one of the triangle sides to be an odd multiple of $\lambda^{SP}/4$. However, because each of the atoms interacts with two wires we will not find any subradiant states in these cases unlike we did for a single wire.

Finally, we would like to propose a scheme for bringing the atomic population into the entangled subradiant W-state in case of the arrangement seen in (3.122). Since now, unlike for the single wire, $|1, 1\rangle$ is the only subradiant state, one way of achieving this could be to excite the system into $|eee\rangle$, let it decay and then post-select.

However, using resonant external fields is a more efficient scheme. Although we cannot perform optical pumping, by way of isolating the subradiant state with a convenient choice of field amplitudes, as we did for the single-wire case, we can still exploit the fact that from the excited states only $|1, 1\rangle$ is slowly decaying and all the others are short-lived. Setting the Rabi frequencies of the classical fields interacting with the atoms to be

$$\Omega_1 = \Omega_2 = \Omega_3 \quad (3.126)$$
results in a matrix for the classical part of the interaction Hamiltonian (3.99)

\[
H_{cl}^{cl} = \begin{pmatrix}
0 & \sqrt{3}\Omega_1 & 0 & 0 & 0 & 0 & 0 & 0 \\
\sqrt{3}\Omega_1 & 0 & 0 & 0 & 2\Omega_1 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & \Omega_1 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & -\Omega_1 & 0 \\
0 & 2\Omega_1 & 0 & 0 & 0 & 0 & 0 & \sqrt{3}\Omega_1 \\
0 & 0 & \Omega_1 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & -\Omega_1 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & \sqrt{3}\Omega_1 & 0 & 0 & 0
\end{pmatrix}
\] (3.127)

written up in the basis (3.103). The matrix (3.127) results in the linkage pattern depicted in Fig. 3.28. We choose \(\Omega_1\) so that the effective field \(\sqrt{3}\Omega_1\) acting on the transition \(|ggg\rangle - |1,1\rangle\) be a \(\pi\) pulse.

![Level scheme in the diagonal basis for the three-atom system when the atoms are coupled by a three wires arranged in a triangular fashion.](image)

Figure 3.28: Level scheme in the diagonal basis for the three-atom system when the atoms are coupled by a three wires arranged in a triangular fashion. The relative distance of the neighbouring atoms is \(\lambda_{SP}/2\). In this case we find a single subradiant state, the symmetric \(|1,1\rangle\) which is a W state. Setting the external fields \(\Omega_1 = \Omega_2 = \Omega_3\), we can bring the population into \(|1,1\rangle\) by aid of a \(\pi\) pulse which has an amplitude that saturates the slowly decaying transition but cannot saturate the other, fast-decaying transitions. Post-selection eliminates the imperfections occurring during the process.

According to similar considerations as for the phase gate in Sec. 3.7, we must set the amplitude \(\sqrt{3}\Omega_1^0\) of the pulse to be

\[
\Gamma_{11} - \Gamma_{12} \ll \sqrt{3}\Omega_1^0 \ll 2\Gamma_{11} - \Gamma_{12}
\] (3.128)

so that the \(|ggg\rangle - |1,1\rangle\) transition is saturated but the fast-decaying \(|2,1\rangle\) state is only marginally populated. The imperfections arising from the scattering into higher-excited states can be eliminated by post-selection.
3.9 Chain of atoms: realization of a spin-boson model

The plasmon-mediated interaction between spins along a nanowire could also be used to implement interesting and important many-body models involving a one-dimensional continuum of bosonic modes. The role of the latter is played here by the guided plasmon modes. Of particular interest is e.g. the spin-boson model \[^{[90]}\], in which a chain of spins is coupled to the one-dimensional continuum of bosonic modes through an interaction of the type

\[
\hat{H}_{SB} = \int_{0}^{\infty} d\omega \sum_{j} \left( \hat{\sigma}_{j} + \hat{\sigma}_{j}^{\dagger} \right) \left( g(\omega)\hat{a}_{\omega} + g^{*}(\omega)\hat{a}_{\omega}^{\dagger} \right). \tag{3.129}
\]

Different from the atom-plasmon coupling in the interaction Hamiltonian of (3.11) discussed throughout this paper, this Hamiltonian contains counter-rotating terms of the form $\hat{\sigma}^{\dagger}\hat{a}^{\dagger}$ and $\hat{\sigma}\hat{a}$. Typically these terms can be neglected for electromagnetic modes oscillating at optical frequencies, which constitutes the so-called rotating-wave approximation (RWA). It is, however, possible to compensate the fast oscillations by using effective two-photon Raman transitions rather than single-photon ones, as suggested in Ref. \[^{[91]}\]. Consider e.g. the $F = 1/2 - F = 1/2$ coupling scheme shown in Fig. 3.29, where the lower state $|g\rangle$ is coupled to another state $|s\rangle$ in the ground-state manifold via a two-photon Raman transition through an excited state $|e_{+}\rangle$ involving a right circular polarized external drive field of Rabi-frequency $\Omega_{+}$ and the linear polarized quantized plasmon field $E$.

![Figure 3.29: Raman coupling scheme for the realization of a spin boson model.](image)

In the rotating wave approximation the atom can make a transition from $|g\rangle$ to $|s\rangle$ either by emitting a photon into $E$ via the transition $|g\rangle \rightarrow |e_{+}\rangle \rightarrow |s\rangle$ or by absorbing a photon from $E$ via the transition $|g\rangle \rightarrow |e_{-}\rangle \rightarrow |s\rangle$. In addition the state $|s\rangle$ is coupled through another excited state $|e_{-}\rangle$ via a two-photon Raman transition involving a left circular polarized external drive field $\Omega_{-}$ and again the linear polarized plasmon field $E$. For each single-photon transition the rotating-wave approximation is well justified.
since we are considering optical fields. The complete interaction Hamiltonian in RWA thus reads

\[ \hat{H}_I = \sum_j \left\{ \hbar \Omega^+_j \hat{\sigma}^+_j \hat{d}^+_j + \Omega^-_j \hat{\sigma}^-_j \hat{d}^-_j + H.c. \right\} \tag{3.130} \]

\[ + \sum_j \left( \hat{\sigma}^-_j \hat{d}^-_j \hat{E}^+(z_j) + \hat{\sigma}^+_j \hat{d}^+_j \hat{E}^-(z_j) \right). \]

Here \( \hat{\sigma}^\pm_j = |\mu\rangle_j \langle \nu| \) are the atomic flip operators between states \(|\nu\rangle\) and \(|\mu\rangle\) of the \(j\)th atom at position \(z_j\), and \(\hat{d}_\pm\) are the vector dipole moments of the transitions \(|s\rangle - |e_+\rangle\) and \(|g\rangle - |e_-\rangle\) respectively.

It is assumed that the Raman transitions are in two-photon resonance. On the other hand the single-photon transitions shall have a large detuning \(\Delta_\pm\) such that the excited states can be adiabatically eliminated. To illustrate the adiabatic elimination procedure let us consider only a single atom at position \(z_j\), and \(\hat{d}_\pm\) are the vector dipole moments of the transitions \(|s\rangle - |e_+\rangle\) and \(|g\rangle - |e_-\rangle\) respectively.

It is assumed that the Raman transitions are in two-photon resonance. On the other hand the single-photon transitions shall have a large detuning \(\Delta_\pm\) such that the excited states can be adiabatically eliminated. To illustrate the adiabatic elimination procedure let us consider only a single atom at position \(z_j\), and \(\hat{d}_\pm\) are the vector dipole moments of the transitions \(|s\rangle - |e_+\rangle\) and \(|g\rangle - |e_-\rangle\) respectively.

The equation of motion of the atomic wavefunction \(|\psi(t)\rangle = c_g(t) |g\rangle + c_e(t) |e\rangle + c_s(t) |s\rangle\) reads

\[ \frac{d}{dt} \begin{pmatrix} c_g(t) \\ c_e(t) \\ c_s(t) \end{pmatrix} = i \begin{pmatrix} 0 & \Omega^+_1 & 0 \\ \Omega^-_1 & \Delta & \Omega^-_2 \\ 0 & \Omega^-_2 & 0 \end{pmatrix} \begin{pmatrix} c_g(t) \\ c_e(t) \\ c_s(t) \end{pmatrix}. \]

If \(|\Delta| \gg |\Omega_{1,2}|\) we can neglect the time derivative in the second line (adiabatic elimination of the excited state) and approximate \(c_e(t) \approx \Omega_1 c_g(t)/\Delta + \Omega_2 c_s(t)/\Delta\). Substituting this result in the above equations yields

\[ \frac{d}{dt} \begin{pmatrix} c_g(t) \\ c_s(t) \end{pmatrix} = i \begin{pmatrix} 0 & \Omega^+_1 \Omega^-_2/\Delta \\ \Omega^-_1 \Omega^+_2/\Delta & 0 \end{pmatrix} \begin{pmatrix} c_g(t) \\ c_s(t) \end{pmatrix}. \]

Since the individual optical Rabi-frequencies have a time dependence \(\Omega_{1,2} \sim e^{-i\omega_{1,2}t}\), the two-photon Rabi-frequency \(\Omega_1^+ \Omega_2^-/\Delta\) has a slow frequency. Generalizing the above discussion to the two parallel \(\Lambda\) transition of Fig.3.29 and replacing one of the two fields of each \(\Lambda\) transition by the common quantized probe field \(\hat{E}\) leads to the effective Hamiltonian

\[ \hat{H}_{\text{eff}} \sim \sum_j \left( \Omega^+_j \hat{d}^+_j + \Omega^-_j \hat{d}^-_j \hat{E}^-(z_j) + \frac{\Omega^+_j \Omega^-_j}{\Delta} \hat{d}^+_j \hat{E}^-(z_j) + H.c. \right). \tag{3.131} \]

which exactly corresponds to that of the spin-boson model.
CHAPTER 3. COLLECTIVE BEHAVIOUR OF QUANTUM EMITTERS COUPLED TO AN ENGINEERED RESERVOIR

3.10 Dipole-dipole shifts

In the preceding sections of this chapter we mainly discussed the collective decay rates arising from the effective interaction between emitters, and their consequences. However, we have not yet said much about the Lamb and dipole-dipole shifts. The reason for this is that while we can determine the decay rates and couplings from calculating the required Green’s tensor component at a given frequency, the energy shifts additionally require a principal value integration that has to be performed over the whole spectral range. This is generally a rather difficult task since in most cases we do not have an analytic expression for the Green’s tensor.

Below we present a method that makes it much easier to tackle these additional difficulties and we subsequently apply it for the case of a pair of emitters coupled by the $n = 0$ surface plasmon mode of a nano-wire.

3.10.1 General method

Let us consider a system of $N$ two-level quantum emitters, characterized by the lowering and raising operators $\hat{\sigma}_j$ and $\hat{\sigma}_j^\dagger$ coupled to a common electromagnetic reservoir. Under conditions that permit the dipole-, rotating wave-, and Markov approximation, following the steps described in Sec. 3.3 leads to a master equation for the atoms of the following form

$$\dot{\hat{\rho}} = \frac{i}{\hbar} \sum_{m,n=1}^{N} \delta\omega_{mn}[\hat{\sigma}_m^\dagger \hat{\sigma}_n, \hat{\rho}] - \sum_{m,n=1}^{N} \frac{\Gamma_{mn}}{2} \left( \hat{\sigma}_m^\dagger \hat{\sigma}_n \hat{\rho} + \hat{\rho} \hat{\sigma}_m^\dagger \hat{\sigma}_n - 2 \hat{\sigma}_n \hat{\rho} \hat{\sigma}_m^\dagger \right).$$ (3.132)

The first, Hermitian term of the RHS contains the $\delta\omega_{mn}$ Lamb shifts and $\delta\omega_{m\neq n}$ dipole-dipole shifts. In the dissipative term we find the single-atom decay rates $\Gamma_{nn}$ and the inter-atomic couplings $\Gamma_{m\neq n}$. Their explicit form is:

$$\Gamma_{mn}(\omega_A) = \frac{2\omega_A^2 d_m d_n^*}{\hbar \epsilon_0 c^2} \text{Im} \left[ G_{ij}(r_m, r_n, \omega_A) \right]$$ (3.133)

$$\delta\omega_{mn}(\omega_A) = \frac{d_m d_n^*}{\hbar \epsilon_0 \pi} \int_0^\infty \frac{d\omega}{c^2} \frac{\omega^2}{\omega - \omega_A} \text{Im} \left[ G_{ij}(r_m, r_n, \omega) \right].$$ (3.134)

Here, $G_{ij}(r_m, r_n, \omega)$ is the $\{i, j\}$ component of the Green’s tensor for the electromagnetic field including the interaction with a passive medium, evaluated at frequency $\omega$ and at positions $r_m$ and $r_n$. Deriving (3.133) and (3.134) we also applied a Markov approximation. This is possible as long as the calculated decay rates $\Gamma_{mn}(\omega)$ and shifts $\delta\omega_{mn}(\omega)$ depend only slowly on frequency $\omega$, i.e. do not change appreciably over frequency ranges of the order of $\Gamma_{mn}$ and $\delta\omega_{mn}$. It should be kept in mind that even if the spectral response of the medium is flat, retardation effects can cause a spectral dispersion of the Green’s tensor at two different positions $r_m$ and $r_n$ with a characteristic width given by $c/|r_m - r_n|$. [82]
3.10. DIPOLE-DIPOLE SHIFTS

If we allow magnetic media (see Appendix 2), \( \vec{G}(r_m, r_n, \omega) \) fulfills the Maxwell-Helmholtz wave equation assuming the form

\[
\left[ \nabla \times \frac{1}{\mu(r, \omega)} \nabla \times - \frac{\omega^2}{c^2} \epsilon(r, \omega) \right] \vec{G}(r, r', \omega) = \vec{I} \delta(r - r'),
\]

(3.135)

with the proper boundary conditions. \( \epsilon(r, \omega) \) and \( \mu(r, \omega) \) are the relative electric permittivity and magnetic permeability, respectively. In case of transition frequencies \( \omega_A \) for which the rotating wave approximation is valid and which are far from the ultraviolet domain, we can use the full (vacuum plus material part) Green’s tensor for calculating \( \Gamma_{mn} \). However, for calculating \( \delta\omega_{mn} \) one has to perform an integral over the whole spectrum. Since the atom-field coupling is treated in a non-relativistic way, this approach does not take into account the high-frequency contributions correctly. A well known consequence of this is that the vacuum level shifts (Lamb shift) obtained from (3.134) are incorrect. Here a rather involved calculation based on relativistic quantum field theory is required, taking into account all possible transitions of the emitter and including proper renormalizations.

However, if we are interested only in the changes produced by the presence of a medium, this problem can be circumvented. Usually, the medium tailors the reservoir modes only within a certain frequency range and becomes transparent in the high-frequency domain where relativistic effects typically occur. Thus, calculating the effects relative to the case of free-space vacuum, i.e., using \( \vec{G}^{med} = \vec{G} - \vec{G}^{vac} \) instead of the full \( \vec{G} \), the above equations give correct expressions also for the Lamb or dipole-dipole shifts relative to vacuum. In other words, calculating only the material-induced level shift introduces an automatic renormalization and lets us get rid of the ultraviolet divergences.

Even though ultraviolet divergencies are eliminated in (3.134) by considering only the changes due to the medium, the expression still contains an integral which is rather difficult to calculate. In the following, we give a general method for simplifying this expression. For this we use a generalized Kramers-Kronig relation. Note that in the discussion below, for simplicity and transparency, we omit the spatial coordinates in the argument of \( \vec{G} \), and indicate it only when necessary.

Kramers-Kronig relations for the Green’s tensor

The full Green’s tensor \( \vec{G}(\omega) \) does not have any poles on the upper complex half-plane because of causality. Thus, the Kramers-Kronig relations (see Appendix 3) apply, e.g.

\[
\text{Re}[\vec{G}(\omega_A)] = \frac{1}{\pi} \text{P} \int_{-\infty}^{\infty} d\omega \frac{\text{Im}[\vec{G}(\omega)]}{\omega - \omega_A}.
\]

(3.136)

Note that the Green’s function inherits the symmetry \( \vec{G}(-\omega^*) = \vec{G}^*(\omega) \) from \( \epsilon(\omega) \) and \( \mu(\omega) \). For real \( \omega \) this means that \( \text{Re}[\vec{G}(\omega)] \) is an even, \( \text{Im}[\vec{G}(\omega)] \) is an odd function of \( \omega \). Thus, equation
\( (3.136) \) can also be written in the form
\[
\text{Re}[\bar{G}(\omega_A)] = \frac{2}{\pi} \int_0^\infty d\omega \frac{\omega \text{Im}[\bar{G}(\omega)]}{\omega^2 - \omega_A^2}.
\]  (3.137)

An important step in the derivation of the Kramers-Kronig relation is the integration over the semicircle (\( \nearrow \) contribution) in the complex upper half plane. As \( \bar{G}(\omega) \to \bar{G}_\text{vac}(\omega) \) for large \(|\omega|\), we use the vacuum Green’s tensor \((92)\)
\[
\bar{G}_\text{vac}(\mathbf{r},\omega) = e^{i\omega c r/4\pi r} \left[ \left( 1 - \frac{1}{i\omega c r} - \frac{1}{\omega^2 c^2 r^2} \right) \hat{\mathbf{r}} \otimes \hat{\mathbf{r}} + \frac{\delta(r)}{3\omega^2 c^2} \right]
\]  (3.138)
for the integration over the semicircle of the contour. At the same time we extend the semicircle to infinity, i.e., \(|\omega| \to \infty\). At large \(|\omega|\), \(\bar{G}_\text{vac}(\omega)\) goes as
\[
\lim_{|\omega| \to \infty} \bar{G}_\text{vac}(\mathbf{r},\omega) = e^{i\omega c r/4\pi r} \left( 1 - \hat{\mathbf{r}} \otimes \hat{\mathbf{r}} \right).
\]  (3.139)
To perform the \( \nearrow \) integral we use the parametrization \( \omega = |\omega|e^{i\alpha} \) and approach the two points where the \( \nearrow \) part joins the real axis (\( \delta \leq \alpha \leq \pi - \delta \)). We then find
\[
I = |\omega| \int_{\nearrow} d(e^{i\alpha}) e^{i\omega r} |\omega| e^{i\alpha} = |\omega| \int_{1+1i\delta}^{1+1i\delta} d(e^{i\alpha}) e^{i\omega |\omega| e^{i\alpha}} = -\frac{2e}{r} \sin\left( \frac{|\omega|}{c} r \right) e^{-\frac{|\omega|}{c} r \delta}.
\]  (3.140)
Taking \(|\omega|\) to infinity and \(\delta\) to zero, we get
\[
\lim_{|\omega| \to \infty} \lim_{\delta \to 0} I = 0.
\]  (3.141)
Because the integrand on any point of the \( \nearrow \) contour part goes exponentially fast to 0, the integral will vanish on this part of the contour even if we multiply the integrand with a polynomial of \( \omega \). In particular we find that the variant of the Kramers-Kronig relation
\[
\frac{\omega_A^2}{c^2} \text{Re}[\bar{G}(\omega_A)] = \frac{2}{\pi} \int_0^\infty d\omega \frac{\omega^2 \text{Im}[\bar{G}(\omega)]}{\omega^2 - \omega_A^2}
\]  (3.142)
holds as well. For the reasons stated above, \((3.136)\) and \((3.142)\) - being true for \(\bar{G}\) and \(\bar{G}_\text{vac}\) - are valid for the material contribution \(\bar{G}_\text{med} = \bar{G} - \bar{G}_\text{vac}\) also.

Medium contribution to the Lamb and dipole-dipole shift

We can rewrite the principal value integral in \((3.141)\) as
\[
I_1 = \frac{2}{\pi} \int_0^\infty d\omega \frac{\omega^2 \text{Im}[G_{ij}(r_m, r_n, \omega)]}{\omega^2 - \omega_A^2} (\omega + \omega_A),
\]  (3.143)
where we replaced the full $\tilde{G}$ by $\tilde{G}^{med} = \tilde{G} - \tilde{G}^{vac}$, i.e. the contribution of the medium. We can now substitute the variant of the Kramers-Kronig relation \((3.142)\) into \((3.143)\) which yields:

$$I_1 = \frac{\pi \omega_A^2}{2 c^2} \text{Re}\{G_{ij}^{med}(\omega_A)\} + \text{Im}\left\{ \frac{1}{\omega + \omega_A - i\delta} \right\}.$$  \hspace{1cm} (3.144)

Now, we try to evaluate the principal value from the second term of \((3.144)\). In order to do this we will transfer the integral from the real axis to the imaginary axis. We shall see that in addition to losing the principal value from the front of the integral, we get a much better-behaved integrand for purely imaginary frequencies: the oscillations in the components of the Green’s tensor become exponentially decreasing functions. As $\tilde{G}(\omega)$ is analytic on the upper complex half-plane, the integrand on the RHS of \((3.144)\) has poles at $\omega = \pm \omega_A$. Introducing small losses to the system, i.e., shifting the poles to the upper complex half-plane and using the identity

$$\frac{1}{\omega - (\pm \omega_A + i\delta)} = \frac{1}{\omega + \omega_A - i\delta} + i\pi \delta(\omega - \omega_A),$$

we can resolve the principal value term

$$\mathcal{P}\left(\frac{\omega_A}{\omega - \omega_A} \right) = \frac{1}{2} \left( \frac{1}{\omega - \omega_A - i\delta} - i\pi \delta(\omega - \omega_A) \right).$$

Thus, the second term in \((3.144)\) assumes the form

$$I_2 = -\frac{\pi}{2} \text{Im}\left\{ \frac{\omega_A^2}{c^2} G_{ij}^{med}(\omega_A) \right\} + 2 \text{Im}\left\{ \int_0^\infty d\omega \frac{\omega^2}{c^2} G_{ij}^{med}(\omega) \left( \frac{1}{\omega - \omega_A - i\delta} - \frac{1}{\omega + \omega_A - i\delta} \right) \right\}. \hspace{1cm} (3.147)$$

Because the integral goes from 0 to $\infty$ on the real axis, we can create a closed contour in the upper right quarter of the complex plane. The integral over the curved part (C) of the contour disappears so we can write

$$\int_0^\infty \rightarrow \oint - \int_C - \int_{i\infty}^0 \rightarrow \oint - \int_{i\infty}^0. \hspace{1cm} (3.148)$$

Using the parametrization $\omega = i\kappa$ on the imaginary axis, we get

$$I_2 = \frac{\pi \omega_A^2}{2 c^2} \text{Re}\{G_{ij}^{med}(\omega_A)\} - \text{Im}\left\{ \int_0^\infty d\kappa \frac{\omega_A^2}{c^2} \frac{iG_{ij}^{med}(i\kappa)}{2} \left( \frac{1}{i\kappa - \omega_A} - \frac{1}{i\kappa + \omega_A} \right) \right\}. \hspace{1cm} (3.149)$$
Substituting \((3.149)\) into \((3.144)\) and then that into \((3.134)\), we get for the medium contribution to the Lamb and dipole-dipole shift

\[
\delta \omega_{mn}(\omega_A) = \frac{d_m}{\hbar \pi \epsilon_0} \left[ \frac{\omega_A^2}{c^2} \operatorname{Re}\{G_{ij}^\text{med}(\mathbf{r}_m, \mathbf{r}_n, \omega_A)\} \right. \\
+ \int_0^\infty \frac{d\kappa}{c^2} \operatorname{Re}\{G_{ij}^\text{med}(\mathbf{r}_m, \mathbf{r}_n, i\kappa)\} \frac{\omega_A}{\kappa^2 + \omega_A^2} \right].
\]

(3.150)

In this form we no longer have to worry about the principal value. As an additional benefit, transferring the integration to the imaginary axis makes the Green’s tensor much better behaved (exponential decay instead of oscillations) which is very useful when calculating the shift by numerical means.

**Origin of the integral term**

The expression for the dipole-dipole shift found in the literature (for example, \[93, 94, 95\]) usually involves only the first term of the right hand side of \((3.150)\) or, equivalently, the shift is proportional to the real part of the electric field at the atomic frequency. This is, however, an approximation that relies on the assumption that only frequencies around \(\omega_A\) contribute to the dipole-dipole shift considerably. We can obtain this result if we only keep the first two terms in \((3.146)\), arguing that the other terms have their greatest contribution at around \(\omega = -\omega_A\) which is not contained by the region of integration in \((3.143)\). However, there is a fundamental reason why in general the integral term in \((3.150)\) must be present. Starting from \((3.144)\), obtained from the principal value integral in \((3.134)\), we can write

\[
\mathbb{P} \int_0^\infty \frac{d\omega}{c^2} \frac{\omega^2}{\omega - \omega_A} \operatorname{Im}\{G_{ij}^\text{med}(\omega)\} = \frac{\pi \omega_A^2}{2 c^2} \operatorname{Re}\{G_{ij}^\text{med}(\omega_A)\} \\
+ \omega_A \mathbb{P} \int_0^\infty \frac{d\omega}{c^2} \frac{\omega^2}{\omega^2 - \omega_A^2} \operatorname{Im}\{G_{ij}^\text{med}(\omega)\}. 
\]

(3.151)

Resolving the integral term on the RHS of Eq. \((3.151)\), we get two terms, one of which is identical to half of the term on the LHS of the equation. Thus, one obtains

\[
\mathbb{P} \int_0^\infty \frac{d\omega}{c^2} \frac{\omega^2}{\omega - \omega_A} \operatorname{Im}\{G_{ij}^\text{med}(\omega)\} = \frac{\pi \omega_A^2}{2 c^2} \operatorname{Re}\{G_{ij}^\text{med}(\omega_A)\} \\
- \mathbb{P} \int_0^\infty \frac{d\omega}{c^2} \frac{\omega^2}{\omega + \omega_A} \operatorname{Im}\{G_{ij}^\text{med}(\omega)\}. 
\]

(3.152)
Changing the integration variable on the RHS of Eq. \((3.152)\) to \(\tilde{\omega} = -\omega\) and using the fact that \(\text{Im}\{G_{ij}(\omega)\}\) is an odd function of \(\omega\), we get
\[
\mathbb{P} \int_{0}^{\infty} d\omega \frac{\omega^2 \text{Im}\{G_{ij}^{med}(\omega)\}}{c^2 \omega - \omega_A} = \pi \frac{\omega_A^2}{c^2} \text{Re}\{G_{ij}^{med}(\omega_A)\}
\]
\[
-\mathbb{P} \int_{-\infty}^{0} d\tilde{\omega} \frac{\tilde{\omega}^2 \text{Im}\{G_{ij}^{med}(\tilde{\omega})\}}{c^2 \tilde{\omega} - \omega_A}
\]
which, rearranged, gives us the variant of the Kramers-Kronig relation \((3.142)\)
\[
\mathbb{P} \int_{-\infty}^{\infty} d\omega \frac{\omega^2 \text{Im}\{G_{ij}^{med}(\omega)\}}{c^2 \omega - \omega_A} = \pi \frac{\omega_A^2}{c^2} \text{Re}\{G_{ij}^{med}(\omega_A)\}.
\]
Thus, one sees that the discussed term in \((3.152)\) is necessary in order to fulfill the Kramers-Kronig relations.

As a final step, we check the equivalence of the integral term in the RHS of \((3.150)\) with that of \((3.152)\). In order to do this, we start from the latter, transforming the integral as
\[
\int_{0}^{\infty} \to \oint - \int_{C} - \int_{i\infty}^{0}.
\]
Since there are no poles present on the upper right quarter of the complex plane, the total contour integral as well as the integral over the curved part \(C\) is zero and we get
\[
\text{Im}\left[ \int_{0}^{\infty} d\omega \frac{\omega^2 G_{ij}^{med}(\omega)}{c^2 \omega + \omega_A} \right] = \text{Im}\left[ \int_{0}^{\infty} d\kappa \frac{\kappa^2 i G_{ij}^{med}(i\kappa)}{c^2 i\kappa + \omega_A} \right].
\]
Making the denominator real in the first term of the integral on the RHS of \((3.149)\) one arrives to the expression
\[
\text{Im}\left[ \int_{0}^{\infty} d\kappa \frac{\kappa^2 i G_{ij}^{med}(i\kappa)}{c^2 i\kappa + \omega_A} \right] = \int_{0}^{\infty} \frac{\kappa^3 \text{Im}\left[ G_{ij}^{med}(i\kappa) \right]}{c^2} + \frac{\omega_A \kappa^2}{c^2} \text{Re}\left[ G_{ij}^{med}(i\kappa) \right] \).
\]
Similarly, for the second term,
\[
\text{Im}\left[ \int_{0}^{\infty} d\kappa \frac{\kappa^2 i G_{ij}^{med}(i\kappa)}{c^2 i\kappa - \omega_A} \right] = \int_{0}^{\infty} \frac{\kappa^3 \text{Im}\left[ G_{ij}^{med}(i\kappa) \right]}{c^2} - \frac{\omega_A \kappa^2}{c^2} \text{Re}\left[ G_{ij}^{med}(i\kappa) \right] \).
\]
From the property \(\bar{G}(-\omega^*) = \bar{G}^*(\omega)\) follows that \(\text{Im}\left[ G_{ij}^{med}(i\kappa) \right] = 0\) if \(\kappa\) is a real number. As a
result of the considerations above, we arrive to the equations

\[
\text{Im} \left[ \int_0^\infty \frac{d\omega}{c^2} \frac{\omega^2}{\omega + \omega_A} G_{ij}^{\text{med}}(\omega) \right] = -\text{Im} \left[ \int_0^\infty d\kappa \frac{\kappa^2}{c^2} iG_{ij}^{\text{med}}(i\kappa) \right] = \text{Im} \left[ \int_0^\infty d\kappa \frac{\kappa^2}{c^2} iG_{ij}^{\text{med}}(i\kappa - \omega_A) \right]
\]

which means that the expressions (3.150) are equivalent and our calculations are consistent.

As we will see later on for the setup discussed here, the second term in (3.151) is indeed negligible for emitter separations much larger than the characteristic length (i.e., the wavelength). However, when the emitters get close enough, the static \((\omega = 0)\) contribution in \(\delta \omega_{12}\) increases substantially, and we cannot neglect the integral term any more. Also there may be situations such as the coupling of quantum emitters over macroscopic distances through a negative-index material where the question of neglecting it requires a more careful consideration.

### 3.10.2 A pair of emitters near a nano-wire

In the following, we will apply the method introduced above to a particular example. From the interaction of two emitters, each coupled to the basic \((n = 0)\) surface plasmon (SP) mode of a metallic wire of sub-wavelength radius, emerges a Dicke superradiance effect that depends on the inter-emitter distance as described in Sec. 3.6. Expressing the states of the atomic system in the well-known Dicke basis, as seen in Fig. 3.11, the collective atomic decay rates become \(\Gamma_{11} \pm \Gamma_{12}\). This means that the effective interaction between the atoms modifies the original single-atom decay rate \(\Gamma_{11}\) by the coupling \(\Gamma_{12}\) and the singly excited levels also get a \(\pm \delta \omega_{12}\) interaction-induced dipole-dipole shift (see equations (3.133) and (3.134)).

Trying to directly calculate (3.134) introduces difficulties since one has to deal with a principal value integral. This is especially a problem if we have to perform the integral numerically (which is usually the case by non-trivial geometries), because we have to know the behavior of the \(\tilde{G}^{\text{med}}(\omega)\) around \(\omega = \omega_A\). As described in Appendix 22, the reflected part of the Green’s tensor can be expressed as

\[
\tilde{G}_R(r, r', \omega) = \int_{-\infty}^{\infty} dk_z \sum_{n=0}^{\infty} \tilde{G}_R^{(n)}(r, r', \omega; k_z)
\]

and in this case \(\tilde{G}^{\text{med}, (n)}(r, r', \omega; k_z) = \tilde{G}_R^{(n)}(r, r', \omega; k_z)\). Although we know the analytic form of \(\tilde{G}^{\text{med}, (n)}(r, r', \omega; k_z)\), it is a rather complicated function and we cannot analytically integrate it.

Using the method described in the previous section, however, circumvents these difficulties and lets us perform the much simpler integration in (3.150) where, in addition we have to substitute purely imaginary frequencies into the Green’s tensor, by which we get a more well-behaved function.

We calculated the dipole-dipole shift resulting from the presence of the wire using the full scattered Green’s tensor. For a maximal atom-plasmon coupling, we set the polarization of both atoms in the radial direction, i.e., we considered the Green’s tensor element \(G_{rr}^{\text{med}, (n)} = \tilde{r}^T \cdot \tilde{G}^{\text{med}, (n)} \cdot \tilde{r}\).

We compared the result to an analytic approximation used by \cite{94}, based on a single-plasmon
3.10. DIPOLE-DIPOLE SHIFTS

resonance model, a derivation of which, using our approach, is given in Appendix 4:

\[ \delta \omega_{12}^{\text{appr}} = -\frac{2\pi d^2 \omega_A^2}{\hbar \epsilon_0 c^2} A(\omega_A) \gamma(\omega_A) e^{-\gamma \Delta z} \sin(k_{pz} \Delta z). \]  

(3.161)

Here \( A \) and \( \gamma \) are the amplitude and width of the Lorentzian fit to the plasmonic resonances (see Appendix 4) and \( k_{pz} \) is the longitudinal component of the wave vector of the plasmon mode. \( \omega_A \) is the frequency and \( d \) is the dipole strength of the atomic transition, and \( \Delta z \) is the inter-emitter distance.

Figure 3.30: Dipole-dipole shift \( \delta \omega_{12} \) due to the presence of the wire and the \( \Gamma_{12} \) coupling between the atoms, scaled by the total single atom decay rate \( \Gamma_{11} \), as a function of inter-atomic distance \( \Delta z \) in units of the vacuum radiation wavelength \( \lambda_0 \). \( \delta \omega_{12}^{\text{appr}} \) is a good analytic approximation of \( \delta_{12} \), for distances comparable to \( \lambda_0 \) and higher. Here, \( \delta \omega_{12}/\Gamma_{11} \) shows an oscillating behaviour shifted by \( \pi/2 \) with respect to \( \Gamma_{12}/\Gamma_{11} \). For inter-emitter distances comparable to the wire radius, there is a substantial increase in \( \delta \omega_{12} \) and it begins to strongly deviate from \( \delta \omega_{12}^{\text{appr}} \) (see Fig. 3.31).

Fig. 3.30 shows the results of the calculations. For inter-emitter distances larger than the vacuum radiation wavelength, \( \delta \omega_{12}^{\text{appr}} \) is a good approximation to the exact wire-induced dipole-dipole shift. It only deviates from the sinusoidal behaviour when the inter-emitter distance becomes comparable to the wire radius: in this case, the atoms begin to be affected by the three dimensional nature of the wire and the quasi-1D coupling approximation (i.e., \( \delta \omega_{12}^{\text{appr}} \)) is no longer valid. However, because the wire is quite thin, this typically happens at distances well below the vacuum radiation wavelength which means that in this regime the emitters are already strongly interacting through the vacuum as well. So we can safely say that the analytic approximation works well if the inter-emitter distance is above the vacuum radiation wavelength of the emitters. In the regime where the exact calculations are well approximated by \( \delta \omega_{12}^{\text{appr}} \), \( \delta \omega_{12}/\Gamma_{11} \) oscillates with the same period as \( \Gamma_{12}/\Gamma_{11} \) only with an additional \( \pi/2 \) relative phase shift [94]. This means that for inter-emitter distances yielding extrema
CHAPTER 3. COLLECTIVE BEHAVIOUR OF QUANTUM EMITTERS COUPLED TO AN 
ENGINEERED RESERVOIR

Figure 3.31: The first term and the second, integral term of the dipole-dipole shift, as seen in \((3.150)\) and the actual wire-induced dipole-dipole shift \(\delta \omega_{12}\), scaled by the total single-emitter decay rate \(\Gamma_{11}\). Because of the increasing static (\(\omega = 0\)) contributions in the Green’s tensor, for small inter-emitter distances the integral term becomes comparable to the first term, thus becoming no longer negligible.

for \(\Gamma_{12}/\Gamma_{11}\), i.e., where the symmetric or antisymmetric transition is superradiant, \(|S\rangle\) and \(|AS\rangle\) are degenerate. On the other hand, when \(\Gamma_{12}/\Gamma_{11} = 0\), this degeneracy is lifted by \(|\delta \omega_{12}\rangle\) being maximal. The extrema of \(|\delta \omega_{12}\rangle\) are \(0.5\Gamma_{11}\) at most. The decay of the amplitude of the oscillations for both \(\delta \omega_{12}/\Gamma_{11}\) and \(\Gamma_{12}/\Gamma_{11}\) is caused by ohmic losses in the metal, represented by \(\gamma\). Thus, the interaction always makes a distinction between the symmetric and the antisymmetric transition: either by the different decay rates, or by the lifted degeneracy of \(|S\rangle\) and \(|AS\rangle\).

As seen in Fig 3.31, the closer the emitters are to each other, the more substantial the integral term of \((3.150)\) becomes. This is in accordance with the arguments made in Sec. 3.10.1, namely that the decrease of the inter-emitter distance enhances the static contribution of the Green’s tensor. For small enough distances, the integral term is not negligible any more.
Part III

Appendix
.1 Field quantization in the presence of absorbing, magnetic media

Below, we shortly present the quantization scheme involving a Green’s function formalism, as discussed in [80, 81]. In order to quantize the electromagnetic field in the presence of linear, absorbing magnetodielectric matter, we start with the material-assisted Maxwell equations and replace the classical fields with operators:

\[
\nabla \cdot \hat{\mathbf{B}}(\mathbf{r}, \omega) = 0 \tag{1}
\]
\[
\nabla \cdot \hat{\mathbf{D}}(\mathbf{r}, \omega) = 0 \tag{2}
\]
\[
\nabla \times \hat{\mathbf{E}}(\mathbf{r}, \omega) = i\omega \hat{\mathbf{B}}(\mathbf{r}, \omega) \tag{3}
\]
\[
\nabla \times \hat{\mathbf{H}}(\mathbf{r}, \omega) = -i\omega \hat{\mathbf{D}}(\mathbf{r}, \omega) \tag{4}
\]

where, as usual,

\[
\hat{\mathbf{D}}(\mathbf{r}, \omega) = \epsilon_0 \hat{\mathbf{E}}(\mathbf{r}, \omega) + \hat{\mathbf{P}}(\mathbf{r}, \omega) \tag{5}
\]
\[
\hat{\mathbf{H}}(\mathbf{r}, \omega) = \kappa_0 \hat{\mathbf{B}}(\mathbf{r}, \omega) - \hat{\mathbf{M}}(\mathbf{r}, \omega) \tag{6}
\]

\(\epsilon_0\) is the vacuum electric permittivity and \(\kappa_0 = 1/\mu_0\) is the inverse of the vacuum magnetic permeability. In accordance with the fluctuation-dissipation theorem, if there is absorption in the system then, inevitably, noise arises as well. Therefore, we add electric and magnetic noise terms (associated with electric and magnetic losses) to the material-related field operators:

\[
\hat{\mathbf{P}}(\mathbf{r}, \omega) = \epsilon_0 (\epsilon(\mathbf{r}, \omega) - 1) \hat{\mathbf{E}}(\mathbf{r}, \omega) + \hat{\mathbf{P}}_N(\mathbf{r}, \omega) \tag{7}
\]
\[
\hat{\mathbf{M}}(\mathbf{r}, \omega) = \kappa_0 (1 - \kappa(\mathbf{r}, \omega)) \hat{\mathbf{B}}(\mathbf{r}, \omega) + \hat{\mathbf{M}}_N(\mathbf{r}, \omega) \tag{8}
\]
where $\hat{P}_N(r, \omega)$ and $\hat{M}_N(r, \omega)$ are the noise polarization and noise magnetization, related to the electric and magnetic losses, respectively. $\epsilon(r, \omega)$ is the relative electric permittivity and $\kappa(r, \omega)$ is the inverse of the relative magnetic permeability. Constructing the wave equation for the electric field from the equations above, we get

$$\nabla \times \kappa(r, \omega) \nabla \times \hat{E}(r, \omega) - \frac{\omega^2}{c^2} \epsilon(r, \omega) \hat{E}(r, \omega) = i \omega \mu_0 \hat{j}_N(r, \omega)$$

(9)

where on the RHS the source term is the noise current $\hat{j}_N(r, \omega)$ contains the contribution of the electric as well as the magnetic losses:

$$\hat{j}_N(r, \omega) = -i \omega \hat{P}_N(r, \omega) + \nabla \times \hat{M}_N(r, \omega).$$

(10)

The solution of the wave equation (9) can be expressed with the classical Green’s tensor of the system:

$$\hat{E}(r, \omega) = i \omega \mu_0 \int d^3 r' G(r, r', \omega) \hat{j}_N(r', \omega)$$

(11)

with which (9) can be transformed to the Maxwell-Helmholtz wave equation

$$\left[ \nabla \times \kappa(r, \omega) \nabla \times - \frac{\omega^2}{c^2} \epsilon(r, \omega) \right] G(r, r', \omega) = \delta(r - r')$$

(12)

with the proper boundary conditions. A more detailed discussion of the equation and the Green’s tensor can be found in Appendix 2. Being the noise source in (9), $\hat{j}_N$ represents elementary excitations of the system. Thus, we assign bosonic vector fields $\hat{f}^e_{\omega}(r)$ and $\hat{f}^m_{\omega}(r)$ for the noise polarization and noise magnetization, respectively:

$$\begin{align*}
\hat{P}_N(r, \omega) &= i \sqrt{\frac{\hbar c_0}{\pi}} \epsilon''(r, \omega) \hat{f}^e_{\omega}(r) \\
\hat{M}_N(r, \omega) &= \sqrt{-\frac{\hbar \kappa_0}{\pi}} \kappa''(r, \omega) \hat{f}^m_{\omega}(r),
\end{align*}$$

(13)

(14)

with $\epsilon''(r, \omega) = \text{Im}[\epsilon(r, \omega)]$ and $\kappa''(r, \omega) = \text{Im}[\kappa(r, \omega)]$. It can be shown that the noise polarization and noise magnetization are indeed related to different basic variables ([81] and Ref. 24 therein) and they cannot be represented with a single, common bosonic field. Hence, they fulfill the commutation relations

$$\begin{align*}
\left[ \hat{f}^e_{\omega}(r), \hat{f}^{\dagger q}_{\omega'}(r') \right] &= \delta_{\omega, \omega'} \delta_{q, \omega'} \delta(r - r') \\
\left[ \hat{f}^m_{\omega}(r), \hat{f}^{\dagger q}_{\omega'}(r') \right] &= 0,
\end{align*}$$

(15)

(16)
1. FIELD QUANTIZATION IN THE PRESENCE OF ABSORBING, MAGNETIC MEDIA

with the parameters \( p, q = \{e, m\} \). We can express the noise current with the electric and magnetic bosonic vector field operators as

\[
\hat{j}_N = \mathbf{\omega} \sqrt{\frac{\hbar \varepsilon_0}{\pi}} \epsilon(r, \omega) \mathbf{\hat{f}}_e^p(r) + \nabla \times \sqrt{-\frac{\hbar \kappa_0}{\pi}} \kappa''(r, \omega) \mathbf{\hat{f}}_m^p(r).
\]

(17)

Note that we have the freedom to choose the phase factors for \( \mathbf{\hat{P}}_N \) and \( \mathbf{\hat{M}}_N \). We chose them so that the coefficients of \( \mathbf{\hat{f}}_e^p \) and \( \mathbf{\hat{f}}_m^p \) in (17) are real.

Having defined the operators that create and annihilate elementary excitations of the combined system composed of the electromagnetic field and the lossy medium, we can write for the Hamiltonian

\[
\hat{H}^F = \sum_{p=e,m} \int d^3r \int_0^{\infty} d\omega \mathbf{\hat{f}}_p^\dagger(\omega) \mathbf{\hat{f}}_p(\omega).
\]

(18)

Thus, all quantities of the electromagnetic field can be expressed by aid of the operators \( \mathbf{\hat{f}}_e^p, m \) and their adjoints. For instance, the electric field assumes the form in the Schrödinger picture

\[
\mathbf{\hat{E}}(r) = \int_0^{\infty} d\omega \mathbf{\hat{E}}(r, \omega) + \text{H.c.}
\]

(19)

where, substituting the expression for the noise current into [11] we can write

\[
\mathbf{\hat{E}}(r, \omega) = i \sqrt{\frac{\hbar}{\varepsilon_0 \pi}} \int d^3r' \mathbf{\bar{G}}(r, r', \omega) \cdot \left\{ \frac{\mathbf{\omega}^2}{c^2} \sqrt{\epsilon''(r', \omega)} \mathbf{\hat{f}}_e^p(r') + \frac{\mathbf{\omega}}{c} \nabla r' \times \left[ \sqrt{-\kappa''(r', \omega)} \mathbf{\hat{f}}_m^p(r') \right] \right\}.
\]

(20)

As for the magnetic field, using [4], we can construct

\[
\mathbf{\hat{B}}(r, \omega) = \sqrt{\frac{\hbar}{\varepsilon_0 \pi}} \int d^3r' \nabla r' \times \mathbf{\bar{G}}(r, r', \omega) \cdot \left\{ \frac{\mathbf{\omega}^2}{c^2} \sqrt{\epsilon''(r', \omega)} \mathbf{\hat{f}}_e^p(r') + \frac{1}{c} \nabla r' \times \left[ \sqrt{-\kappa''(r', \omega)} \mathbf{\hat{f}}_m^p(r') \right] \right\}
\]

(21)

and, from this, express the magnetic field operator in Schrödinger picture, as

\[
\mathbf{\hat{B}}(r) = \int_0^{\infty} d\omega \mathbf{\hat{B}}(r, \omega) + \text{H.c.}
\]

(22)

One can also prove that for \( \mathbf{\hat{E}}(r) \) and \( \mathbf{\hat{B}}(r) \) the fundamental equal-time commutation relations

\[
\left[ \mathbf{\hat{E}}_i(r), \mathbf{\hat{E}}_j(r') \right] = \left[ \mathbf{\hat{B}}_i(r), \mathbf{\hat{B}}_j(r') \right] = 0
\]

(23)

\[
\left[ \epsilon_0 \mathbf{\hat{E}}_i(r), \mathbf{\hat{B}}_j(r') \right] = -i\hbar \delta_{ij} \partial^i_k \delta(r - r')
\]

(24)

are preserved when the fields are expressed with the operators \( \mathbf{\hat{f}}_e^p, m(r) \).

Note that in order for (11) to be a unique solution of Maxwell’s equations without a homogeneous term even in the case of no medium in the system, we assume that infinite distance from the relevant regions of space there exists some absorbing medium. In practice this means that one first has to
perform the calculations with a small but finite value of $\epsilon''$ as $|r| \to \infty$ and only take $\epsilon'' \to 0$ afterwards (see [83, 84]).
.2 Green’s tensor

In this Appendix we discuss the derivation and some properties of the classical Green’s tensor, based on [80, 81, 92, 96, 97, 98].

The Green tensor describes the response of the medium to a single oscillating point dipole source, fulfilling the Maxwell-Helmholtz equation derived in Appendix 1:

\[
\begin{aligned}
&\nabla \times \kappa(r, \omega) \nabla \times -\frac{\omega^2}{c^2} \varepsilon(r, \omega) \right) \vec{G}(r, r', \omega) = \vec{I} \delta(r - r')
\end{aligned}
\]  

(25)

where \( \varepsilon(r, \omega) \) is the relative electric permittivity and \( \kappa(r, \omega) = 1/\mu(r, \omega) \) is the inverse of the relative magnetic permeability of the medium.

.2.1 Some properties of the Green’s tensor

Since the relative electric permittivity and magnetic permeability obey the relations \( \varepsilon(r, -\omega) = \varepsilon^*(r, \omega^*) \) and \( \mu(r, -\omega) = \mu^*(r, \omega^*) \), through (25) the Green’s tensor inherits this property:

\[
\vec{G}(r, r', -\omega) = \vec{G}^*(r, r', \omega^*).
\]  

(26)

This also implies that for real frequencies

\[
\begin{aligned}
\text{Re} \left[ \vec{G}(r, r', -\omega) \right] &= \text{Re} \left[ \vec{G}(r, r', \omega) \right] \\
\text{Im} \left[ \vec{G}(r, r', -\omega) \right] &= -\text{Im} \left[ \vec{G}(r, r', \omega) \right].
\end{aligned}
\]  

(27)

The reciprocity relation for exchanging the position of source and observation point

\[
\vec{G}(r, r', \omega) = \vec{G}^T(r', r, \omega)
\]  

(28)

is also generally true. Furthermore, from (25), using the relations (26) and (28), it is possible to show that

\[
\begin{aligned}
&\int d^3s \frac{\omega^2}{c^2} \varepsilon''(s, \omega) \vec{G}(r, s, \omega) \vec{G}^t(s, r, \omega) - \text{Im} \left[ \vec{G}(r, r') \right] \\
&= \int d^3s \kappa''(s, \omega) \left[ \vec{G}(r, s, \omega) \times \nabla_s \right] \cdot \left[ \nabla_s \times \vec{G}^t(r', s, \omega) \right]
\end{aligned}
\]  

(29)

where, in Cartesian coordinates,

\[
\begin{aligned}
\left[ \nabla_s \times \vec{G}(r, s, \omega) \right]_{ij} &= \epsilon_{ikl} \partial_k G_{lj}(r, s, \omega) \\
\left[ \vec{G}(r, s, \omega) \times \nabla_s \right]_{ij} &= \epsilon_{jkl} \partial_k G_{il}(r, s, \omega).
\end{aligned}
\]  

(30)
.2.2 Integral representation of the Green’s tensor

To derive the integral representations for the Green’s tensor we first assume a homogeneous, non-magnetic (\( \mu(\omega) = 1 \)), unbounded medium. Subsequently we also extend the results to magnetic materials. Note that in the following discussion we are going to omit the argument \( \omega \) in the functions, nevertheless we remain in the frequency domain throughout.

It is simpler to solve the Maxwell-Helmholtz wave equation for the scalar Green’s function, and then to transform the solution into the dyadic Green’s function than dealing with the tensorial Maxwell-Helmholtz equation directly. Thus, we have

\[
\left[ \nabla^2 + k_0^2 \right] g(r - r') = -\delta(r - r') \tag{31}
\]

where \( k_0 = \omega \sqrt{\epsilon(\omega)} / c \) and the Green’s function \( g(r, r') \) depends on the difference of its arguments due to the translational invariance in the medium. The relation

\[
\left[ \hat{\mathbf{l}} + \frac{1}{k_0^2} \nabla \otimes \nabla \right] g(r - r') = \tilde{G}(r, r') \tag{32}
\]

defines the connection between the scalar Green’s function and the Green’s tensor. Setting \( r' = 0 \), the solution of (31) in Fourier space is

\[
g(k) = \frac{1}{k^2 - k_0^2}. \tag{33}
\]

Transforming it back we get

\[
g(r) = \frac{1}{(2\pi)^3} \int d^3 k e^{i k \cdot r} g(k). \tag{34}
\]

Note that if we allow for homogeneous magnetic materials in the system, we see from (25) that we can easily obtain the Green’s tensor by multiplying the original, non-magnetic Green’s tensor with \( \mu(\omega) \) and replacing in it \( \epsilon(\omega) \) everywhere by \( \mu(\omega) \epsilon(\omega) \).

At this point there are several directions one can take. If the system is indeed unbounded, we can perform the integral analytically and transform \( g(r) \) into \( \tilde{G}(r) \) using the relation (32). On the other hand, if we aim for the Green’s tensor of a system that consists of multiple layers of different, homogeneous media arranged in a given structure, we only integrate over some of the components of \( k \) so that expanding the integrand in the basis of harmonic functions we are able to fulfill the boundary conditions between the layers. We have to choose the harmonics in a way that they mimic the symmetries of the system.

The boundary condition equations connect the solutions of (25) in the separate layers:

\[
\hat{n}_l \times \tilde{G}(r, r', \omega)_{r=R_l^{-}} = \hat{n}_l \times \tilde{G}(r, r', \omega)_{r=R_l^{+}} \tag{35}
\]

\[
\kappa_l(\omega) \hat{n}_l \times \nabla \times \tilde{G}(r, r', \omega)_{r=R_l^{-}} = \kappa_{l+1}(\omega) \hat{n}_l \times \nabla \times \tilde{G}(r, r', \omega)_{r=R_l^{+}} \tag{36}
\]
where $\hat{n}_l$ is the unit vector perpendicular to the surface between the $l$-th and $l+1$st layer and $R_l$ represents a point on this surface. The conditions (35) and (36) are the Green’s tensor equivalents of the expressions

$$\hat{n}_l \times \mathbf{E}(r, \omega)_{r=R_l^-} = \hat{n}_l \times \mathbf{E}(r, \omega)_{r=R_l^+}$$  \hspace{1cm} (37)

$$\kappa_l(\omega)\hat{n}_l \times \mathbf{H}(r, \omega)_{r=R_l^-} = \kappa_{l+1}(\omega)\hat{n}_l \times \mathbf{H}(r, \omega)_{r=R_l^+}$$  \hspace{1cm} (38)

well-known from classical electrodynamics, $\mathbf{E}(r, \omega)$ and $\mathbf{H}(r, \omega)$ being the electric and magnetic field, respectively.

### Analytic Green’s tensor for unbounded vacuum

Adding infinitesimally small losses to the system, we have

$$g_0(k) = \frac{1}{k^2 - (k_0 + i\delta)^2}.$$  \hspace{1cm} (39)

The complex integration according to (34) results in the scalar Green’s function in real space:

$$g_0(r) = \frac{e^{ikr}}{4\pi r}.$$  \hspace{1cm} (40)

Finally, we get the Green’s tensor by using (32):

$$\mathbb{G}_0(r, \omega) = \frac{e^{ik_0 r}}{4\pi r} \left[ P(ik_0 r)\hat{\mathbb{r}} + Q(ik_0 r)\hat{\mathbb{r}} \otimes \hat{\mathbb{r}} \right] - \frac{\delta(r)}{3k_0^2}\hat{\mathbb{I}}$$  \hspace{1cm} (41)

where

$$P(z) = 1 - \frac{1}{z} + \frac{1}{z^2}$$

$$Q(z) = -1 + \frac{3}{z} - \frac{3}{z^2}.$$  \hspace{1cm} (42)

Note that if we want to calculate the decay rate of a single atom in vacuum, according to (3.27), we have to take $\text{Im}[\mathbb{G}(r)]$ with $r \to 0$. However, having it in spherical coordinates, in $r = 0$ the angles (and so the direction of $\hat{r}$) are not defined. Thus, taking the limit $r \to 0$, one also has to average $\hat{r} \otimes \hat{r}$ over the solid angle, performing the integral

$$\frac{1}{4\pi} \int d\Omega \ [\hat{r} \otimes \hat{r}]_{ij}$$  \hspace{1cm} (43)

where $[\hat{r} \otimes \hat{r}]_{ij}$ is the $\{i, j\}$ component of $\hat{r} \otimes \hat{r}$ in Cartesian coordinates. As a result we get

$$\text{Im} \left[ \mathbb{G}_0(0, \omega) \right] = \frac{k_0}{6\pi} \hat{\mathbb{I}}$$  \hspace{1cm} (44)
which, substituted into (3.27), produces the result of Fermi’s golden rule.

### Plane wave representation

We need this representation for cases when the system consists of planarly stratified layers of materials. Working in Cartesian coordinates, we perform the complex integration in (34) over \( k_z \) substituting (39). Deforming the contour for \( z > 0 \) upwards and for \( z < 0 \) downwards, we obtain

\[
g_0(r) = \frac{i}{(2\pi)^2} \int d^2k_\perp \frac{e^{ik_\perp \cdot r_\perp + ik_z |z|}}{2k_z}
\]

where \( k_z \) is not an integration variable anymore but has the value \( k_z = \sqrt{k_0^2 - k_\perp^2} \) and \( k_\perp = k_x^2 + k_y^2 \). Note that when applying (32) in order to get \( \bar{G}_0(r) \), the first derivative with respect to \( z \) has a discontinuity at \( z = 0 \). This, upon the second derivation with respect to \( z \) results in a Dirac delta term in the final expression:

\[
\bar{G}_0(r) = -\hat{z} \otimes \hat{z} \frac{\delta(r)}{k_0^2} + \left\{ \begin{array}{ll}
\frac{i}{8\pi^2} \int d^2k_\perp \frac{1}{k_z} \left[ \bar{I} - \frac{k \otimes k}{k_0} \right] e^{ik \cdot r} & z > 0 \\
\frac{i}{8\pi^2} \int d^2k_\perp \frac{1}{k_z} \left[ \bar{I} - \frac{K \otimes K}{k_0} \right] e^{iK \cdot r} & z < 0
\end{array} \right.
\]

where

\[
\begin{align*}
k &= k_x \hat{x} + k_y \hat{y} + k_z \hat{z} \\
K &= k_x \hat{x} + k_y \hat{y} - k_z \hat{z}.
\end{align*}
\]

Now, we construct an orthonormal system of unit vectors, starting with \( k/k_0 = \hat{k} \). The other two vectors are as follows:

\[
\begin{align*}
\hat{e}(k_z) &= \frac{\hat{k} \times \hat{z}}{k \times \hat{z}} = \frac{1}{k_\perp} (\hat{x}k_y - \hat{y}k_x) \\
\hat{h}(k_z) &= \frac{1}{k_0} \hat{e} \times k = -\frac{k_z}{k_0 k_\perp} (\hat{x}k_x + \hat{y}k_y) + \frac{k_\perp}{k_0} \hat{z}.
\end{align*}
\]

Being an orthonormal system, \( \bar{I} = \hat{e} \otimes \hat{e} + \hat{h} \otimes \hat{h} + \hat{k} \otimes \hat{k} \) from which follows that

\[
\bar{G}_0(r) = -\hat{z} \otimes \hat{z} \frac{\delta(r)}{k_0^2} + \left\{ \begin{array}{ll}
\frac{i}{8\pi^2} \int d^2k_\perp \frac{1}{k_z} \left[ \hat{e}(k_z) \otimes \hat{e}(k_z) + \hat{h}(k_z) \otimes \hat{h}(k_z) \right] e^{ik \cdot r} & z > 0 \\
\frac{i}{8\pi^2} \int d^2k_\perp \frac{1}{k_z} \left[ \hat{e}(-k_z) \otimes \hat{e}(-k_z) + \hat{h}(-k_z) \otimes \hat{h}(-k_z) \right] e^{iK \cdot r} & z < 0
\end{array} \right.
\]

where

\[
\begin{align*}
\hat{e}(-k_z) &= \hat{e}(k_z) \\
\hat{h}(-k_z) &= \frac{1}{k_0} \hat{e} \times K.
\end{align*}
\]
Constructing the Green’s tensor of a multilayer medium we can use the orthonormal, $k_z$-dependent unit vectors as building blocks demonstrated in the following example.

Green’s tensor for a three-layer planarly stratified medium

Let us assume that the system consists of three layers, marked with 0, 1 and 2, where 0 and 2 are only bounded from one side and 1, being between 0 and 2, is bounded from both sides. We use the notation $\tilde{G}_{(ij)}$ where $l$ and $j$ denote the region where our observation and source points are, respectively. We place the source in region $\bar{0}$, i.e., we set $j = 0$.

In region 0, the Green’s tensor is the sum of the direct (vacuum) contribution (51) and the reflected term:

$$\tilde{G}_{(00)}(\mathbf{r}, \mathbf{r}', \omega) = \tilde{G}_0(\mathbf{r}, \mathbf{r}', \omega) + \tilde{G}_R(\mathbf{r}, \mathbf{r}', \omega)$$

$$= \frac{i}{8\pi^2} \int d^2k_z \frac{1}{k_z} \left\{ \left[ \hat{e}(-k_z)e^{iK\mathbf{r}} + R^{TE}\hat{e}(k_z)e^{ik\mathbf{r}} \right] \hat{e}(-k_z)e^{-iK\mathbf{r}'} + \left[ \hat{h}(-k_z)e^{iK\mathbf{r}} + R^{TM}\hat{h}(k_z)e^{ik\mathbf{r}} \right] \hat{h}(-k_z)e^{-iK\mathbf{r}'} \right\} (z < z')$$

(54)

where $R^{TE}$ and $R^{TM}$ are reflection coefficients associated with the transverse electric and transverse magnetic field contributions, respectively. For region 1, we have reflected as well as transmitted terms:

$$\tilde{G}_{(10)}(\mathbf{r}, \mathbf{r}', \omega) = \frac{i}{8\pi^2} \int d^2k_z \frac{1}{k_z} \left\{ \left[ A_1\hat{e}(k_{1z})e^{iK_1\mathbf{r}} + B_1\hat{e}(-k_{1z})e^{iK_1\mathbf{r}} \right] \hat{e}(-k_z)e^{-iK\mathbf{r}'} + \left[ C_1\hat{h}(k_{1z})e^{iK_1\mathbf{r}} + D_1\hat{h}(-k_{1z})e^{iK_1\mathbf{r}} \right] \hat{h}(-k_z)e^{-iK\mathbf{r}'} \right\}.$$

(55)

For region 2, we only have transmitted terms:

$$\tilde{G}_{(20)}(\mathbf{r}, \mathbf{r}', \omega) = \frac{i}{8\pi^2} \int d^2k_z \frac{1}{k_z} \left\{ T^{TE}\hat{e}(-k_{2z})e^{iK_2\mathbf{r}}\hat{e}(-k_z)e^{-iK\mathbf{r}'} + T^{TM}\hat{h}(-k_{2z})e^{iK_2\mathbf{r}}\hat{h}(-k_z)e^{-iK\mathbf{r}'} \right\}$$

(56)

and the $k$-components in the different regions are

$$k_1 = k_x\hat{x} + k_y\hat{y} + k_{1z}\hat{z}$$

(57)

$$K_1 = k_x\hat{x} + k_y\hat{y} - k_{1z}\hat{z}$$

(58)

$$k_{1z} = \sqrt{k_1^2 - k_x^2 - k_y^2}.$$  

(59)

Note that, for the sake of simplicity, we omitted the tensor product symbols in the equations (54)-(56).

The reflection and transmission coefficients are obtained by solving the boundary condition
equations (35), (36), where the surface normal unit vector \( \hat{n}_l \) is now \( \hat{z} \).

**Cylindrical wave representation**

If the system we study has a cylindrical symmetry, we need to expand the Green’s tensor in the basis of cylindrical harmonic vector wave functions in order to fulfill the boundary conditions (98).

The cylindrical harmonic vector wave functions are given by

\[
\begin{align*}
M_{e,n}(k_r, k_z, \mathbf{r}) &= \nabla \times \left[ Z_n(k_r r) \cos(n \phi) e^{ik_z z} \right] \\
M_{o,n}(k_r, k_z, \mathbf{r}) &= \nabla \times \left[ Z_n(k_r r) \sin(n \phi) e^{ik_z z} \right] \\
N_{e,n}(k_r, k_z, \mathbf{r}) &= \frac{1}{k} \nabla \times M_{e,n}(k_r, k_z, \mathbf{r}) \\
N_{o,n}(k_r, k_z, \mathbf{r}) &= \frac{1}{k} \nabla \times M_{o,n}(k_r, k_z, \mathbf{r})
\end{align*}
\]

where \( \mathbf{r} = (r, \phi, z) \) and \( \mathbf{k} = (k_r, \phi_k, k_z) \), the wave number \( k = \omega \sqrt{\varepsilon} \), and \( k_r^2 = k^2 - k_z^2 \). The \( e \) and \( o \) denote even and odd, respectively. Note that \( k \) can be complex due to the non-vanishing imaginary part of the electric permittivity \( \varepsilon'' = \text{Im}[\varepsilon] \). The radial part \( Z_n(x) \) has to be either replaced by the Bessel function \( J_n(x) \), or, when the “(1)” appears as a superscript on \( M \) or \( N \), by \( H_n^{(1)}(x) \), i.e. the Hankel function of the first kind. We will see by the Green’s tensor expansion that this is needed for regularizing the functions around \( r = 0 \).

The Green’s tensor for an unbounded homogeneous non-magnetic material expanded in cylindrical harmonics assumes the form

\[
\tilde{G}_0(\mathbf{r}, \mathbf{r}', \omega) = -\frac{\hat{r} \hat{r} \delta(\mathbf{r} - \mathbf{r}')}{k^2} + \frac{i}{8\pi} \int_{-\infty}^{\infty} dk_z \sum_{n=0}^{\infty} \frac{2 - \delta_{n,0}}{k_r^2} \left[ M_{e,n}^{(1)}(k_r, k_z, \mathbf{r}) M_{e,n}^{(1)}(k_r, -k_z, \mathbf{r}') + N_{o,n}^{(1)}(k_r, k_z, \mathbf{r}) N_{o,n}^{(1)}(k_r, -k_z, \mathbf{r}') \right] \\
= \begin{cases} 
M_{e,n}^{(1)}(k_r, k_z, \mathbf{r}) M_{e,n}^{(1)}(k_r, -k_z, \mathbf{r}') + N_{o,n}^{(1)}(k_r, k_z, \mathbf{r}) N_{o,n}^{(1)}(k_r, -k_z, \mathbf{r}') & r \leq r' \\
M_{e,n}^{(1)}(k_r, k_z, \mathbf{r}) M_{e,n}^{(1)}(k_r, -k_z, \mathbf{r}') + N_{o,n}^{(1)}(k_r, k_z, \mathbf{r}) N_{o,n}^{(1)}(k_r, -k_z, \mathbf{r}') & r' \leq r 
\end{cases}
\]

The tensorial products between the even and odd cylindrical vector wave functions are defined according to \( M_{e,n} \mathbf{N}_{o,n} = M_{e,n}^{(1)} + M_{o,n}^{(1)} N_{o,n} \) and all other combinations similarly. For better readability, we have omitted the tensorial product symbol between them.

**Green’s tensor for a cylinder embedded in vacuum**

If we have a multilayered, cylindrically symmetric system where the material between the layer boundaries is homogeneous, we can proceed analogously to the plane-wave case: we use the harmonic vector functions as building blocks for the Green’s tensor. The description concerns non-magnetic media in the system, however, one can easily generalize the result to magnetic media as described earlier in this Appendix.
In this example we present the Green’s tensor of a system where a cylinder is embedded in vacuum and the source is outside the cylinder. Thus, we have two regions, we denote the vacuum with 0 and the inside of the cylinder with 1. As before, we denote the Green’s tensor for each region by \( \bar{G}_{(lj)} \) where \( l \) denotes the region index where the observation point is and \( j \) is the index of the region where the source is located.

Thus, if \( r < r' \), for region 0 we have the following superposition of a direct (vacuum) term and a reflected contribution:

\[
\bar{G}_{(00)}(r, r', \omega) = \bar{G}_0(r, r', \omega) + \bar{G}_R(r, r', \omega)
= -\frac{\hat{r} \cdot \delta(r - r')}{k^2} + \frac{i}{8\pi} \int_{-\infty}^{\infty} dk_z \sum_{n=0}^{\infty} \frac{2 - \delta_{n,0}}{k_{r_0}^2} \times \left[ (M_{\delta n}(k_{r_0}, k_z, r) + A_R M_{\delta n}^{(1)}(k_{r_0}, k_z, r) + B_R N_{\delta n}^{(1)}(k_{r_0}, k_z, r)) M_{\delta n}^{(1)}(k_{r_0}, -k_z, r') \\
+ (N_{\delta n}(k_{r_0}, k_z, r) + C_R N_{\delta n}^{(1)}(k_{r_0}, k_z, r) + D_R M_{\delta n}^{(1)}(k_{r_0}, k_z, r)) N_{\delta n}^{(1)}(k_{r_0}, -k_z, r') \right].
\]  

(65)

For the transmitted part, we have

\[
\bar{G}_{(10)}(r, r', \omega) = \frac{i}{8\pi} \int_{-\infty}^{\infty} dk_z \sum_{n=0}^{\infty} \frac{2 - \delta_{n,0}}{k_{r_0}^2} \times \left[ (A_T M_{\delta n}(k_{r_1}, k_z, r) + B_T N_{\delta n}^{(1)}(k_{r_1}, k_z, r)) M_{\delta n}^{(1)}(k_{r_0}, -k_z, r') \\
+ (C_T N_{\delta n}(k_{r_1}, k_z, r) + D_T M_{\delta n}^{(1)}(k_{r_1}, k_z, r)) N_{\delta n}^{(1)}(k_{r_0}, -k_z, r') \right].
\]  

(66)

where \( k_0 = \omega/c, k_1 = k_0\sqrt{\epsilon_1} \) and \( k_{r_{0,1}} = \sqrt{k_{0,1}^2 - k_z^2} \).

The reflection and transmission coefficients \( A_{R,T}, B_{R,T}, C_{R,T}, D_{R,T} \) can be specified by imposing the boundary conditions at the surface of the cylinder (\( r = R \)):

\[
\hat{r} \times \bar{G}(r, r')_{r=R^-} = \hat{r} \times \bar{G}(r, r')_{r=R^+}
\]

\[
\hat{r} \times \nabla \times \bar{G}(r, r')_{r=R^-} = \kappa_1 \hat{r} \times \nabla \times \bar{G}(r, r')_{r=R^+}.
\]  

(67)

(68)
3 Kramers-Kronig relations in general

First, let us look at the Kramers-Kronig relations in case of a general, complex-valued function $f(\omega)$ which is analytic in the upper complex half-plane. According to Cauchy’s theorem,

$$f(\omega_A) = \lim_{\delta \to 0^+} \frac{1}{2\pi i} \oint_C d\omega \frac{f(\omega)}{\omega - \omega_A - i\delta}$$

and the contour (containing the point $\omega_A + i\delta$ within) is closed in the upper complex half-plane. If we assume that the path integral of $f(\omega)$ is nonzero only over the real axis and disappears on the other parts of the contour (that is, on the complex plane) if we extend it to infinity, we can write the integral as

$$f(\omega_A) = \lim_{\delta \to 0^+} \frac{1}{2\pi i} \int_{-\infty}^{\infty} d\omega \frac{f(\omega)}{\omega - \omega_A - i\delta}.$$  \hspace{1cm} (70)

Since

$$\frac{1}{\omega - \omega_A - i\delta} = \mathbb{P} \left( \frac{1}{\omega - \omega_A} \right) + i\pi \delta(\omega - \omega_A),$$

$$f(\omega_A) = \frac{1}{2\pi i} \mathbb{P} \int_{-\infty}^{\infty} d\omega \frac{f(\omega)}{\omega - \omega_A} + \frac{1}{2} f(\omega_A).$$  \hspace{1cm} (72)

Rearranging the equation we get

$$f(\omega_A) = \frac{1}{\pi i} \mathbb{P} \int_{-\infty}^{\infty} d\omega \frac{f(\omega)}{\omega - \omega_A}.$$  \hspace{1cm} (73)

(73) also defines expressions connecting the real and imaginary parts of $f(\omega)$:

$$\text{Re}[f(\omega_A)] = \frac{1}{\pi} \mathbb{P} \int_{-\infty}^{\infty} d\omega \frac{\text{Im}[f(\omega)]}{\omega - \omega_A}$$

$$\text{Im}[f(\omega_A)] = -\frac{1}{\pi} \mathbb{P} \int_{-\infty}^{\infty} d\omega \frac{\text{Re}[f(\omega)]}{\omega - \omega_A}$$  \hspace{1cm} (74)

which are called the Kramers-Kronig relations.
.4 Analytic approximation of the decay and dipole-dipole shift for two emitters coupled to surface plasmons of a nanowire

The Green’s tensor for an infinitely long, cylindrical wire can be calculated as given in Appendix 2. Here, the material contribution $G^{\text{med}}$ is the scattered Green’s tensor component $\bar{G}_R$ and we can formally write

$$\bar{G}_R(\mathbf{r}_1, \mathbf{r}_2, \omega) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} G^{(n)}_{R}(\mathbf{r}_1, \mathbf{r}_2, \omega; k_z) \, dk_z \, dk_z$$

(75)

where we know $G^{(n)}_{R}(\mathbf{r}_1, \mathbf{r}_2, \omega; k_z) = \bar{G}^{\text{med},(n)}_{R}(\mathbf{r}_1, \mathbf{r}_2, \omega; k_z)$ analytically.

The atom-wire coupling is strongest if the dipole moment of the emitters point in the radial direction. If the cylindrical coordinates of two emitters only differ in their $z$ component (where $z$ is the symmetry axis of the wire) one finds

$$G^{\text{med}}_{rr}(\mathbf{r}_1, \mathbf{r}_2, \omega) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} G^{\text{med},(n)}_{rr}(\mathbf{r}_1, \mathbf{r}_1, \omega; k_z) \, dk_z \, dk_z$$

(76)

where $\Delta z = |z_2 - z_1|$ and $G^{\text{med}}_{rr} = \hat{r}^T \cdot \bar{G}^{\text{med}} \cdot \hat{r}$, $\hat{r}$ being the unit vector pointing in the radial direction.

In case of a thin wire (radius well below the vacuum radiation wavelength of the emitter), and small emitter-wire distance (in the order of magnitude of the radius), the plasmonic contribution becomes dominant in $\sum_n G^{\text{med},(n)}_{rr}(\mathbf{r}_1, \mathbf{r}_1, \omega; k_z)$ and it is contained in the $n = 0$ term. In this case, the imaginary part of the Green’s tensor is well approximated by two Lorentzian fits, centered at $\pm k_z^{SP}$, i.e., the $z$ component of the wave vector of the plasmonic mode. This approximation is valid for inter-emitter distances comparable to or larger than the vacuum radiation wavelength because in this case the only substantial channel that couples the emitters are the surface plasmon modes.

$$\text{Im} \left[ G^{\text{med},(0)}_{rr}(\mathbf{r}_1, \mathbf{r}_1, \omega; k_z) \right] \approx \frac{A(\omega)}{1 + \frac{|k_z - k_z^{SP}(\omega)|^2}{\gamma(\omega)^2}} + \frac{A(\omega)}{1 + \frac{|k_z + k_z^{SP}(\omega)|^2}{\gamma(\omega)^2}}$$

(77)

Because of the translational invariance of the wire in the $z$ direction, $G^{\text{med},(0)}_{rr}$ is symmetric in $k_z$. Thus, substituting in (76) we get

$$\text{Im} \left[ G^{\text{med}}_{rr}(\mathbf{r}_1, \mathbf{r}_2, \omega) \right] \approx 2\pi A \gamma \cos(k_z^{SP} \Delta z) e^{-\gamma \Delta z} = \text{Im} \left[ 2\pi i A \gamma e^{ik_z^{SP} \Delta z} e^{-\gamma \Delta z} \right].$$

(78)

Since the Kramers-Kronig relations uniquely define the real part of $G$ if we know the imaginary part, we can safely assume that

$$\text{Re} \left[ G^{\text{med}}_{rr}(\mathbf{r}_1, \mathbf{r}_2, \omega) \right] \approx \text{Re} \left[ 2\pi i A \gamma e^{ik_z^{SP} \Delta z} e^{-\gamma \Delta z} \right] = 2\pi A \gamma \sin(k_z^{SP} \Delta z) e^{-\gamma \Delta z}.$$

(79)

With this, we can now express the wire-induced single-emitter decay rate and emitter-emitter...
coupling, respectively

\[
\Gamma_{11}^{\text{appr}} = \frac{4d^2 \pi \omega_A^2}{\hbar \epsilon_0 c^2} A(\omega_A) \gamma(\omega_A) \tag{80}
\]

\[
\Gamma_{12}^{\text{appr}} = \frac{4d^2 \pi \omega_A^2}{\hbar \epsilon_0 c^2} A(\omega_A) \gamma(\omega_A)e^{-\gamma \Delta z} \cos(k_z^{SP}\Delta z) \tag{81}
\]

where \(d\) is the strength of the dipole transition of the atoms and \(\omega_A\) is the transition frequency.

For the wire-induced dipole-dipole shift, according to (3.150) we get the approximation

\[
\delta \omega_{12} \approx -\frac{2\pi d^2 \omega_A^2}{\hbar \epsilon_0 c^2} A(\omega_A) \gamma(\omega_A)e^{-\gamma \Delta z} \sin(k_z^{SP}\Delta z) \tag{82}
\]

According to the discussion in Sec. 3.10.1, for inter-emitter distances larger than the vacuum radiation wavelength the integral term in (82) can be neglected, so in the end we arrive to the analytic approximation

\[
\delta \omega_{12}^{\text{appr}} = -\frac{2\pi d^2 \omega_A^2}{\hbar \epsilon_0 c^2} A(\omega_A) \gamma(\omega_A)e^{-\gamma \Delta z} \sin(k_z^{SP}\Delta z). \tag{83}
\]

The approximate results can also be written as

\[
\Gamma_{12}^{\text{appr}} = \Gamma_{11}^{\text{appr}} \cos(k_z^{SP}\Delta z)e^{-\gamma \Delta z} \tag{84}
\]

\[
\delta \omega_{12}^{\text{appr}} = -\frac{\Gamma_{11}^{\text{appr}}}{2} \sin(k_z^{SP}\Delta z)e^{-\gamma \Delta z}. \tag{85}
\]
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