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Generation of Correlated Photon Pairs in a Quantum System with Broken Inversion Symmetry

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Zusammenfassung

Diese Diplomarbeit untersucht ein getriebenes Zwei-Niveau-System mit verletzter Inversionssymmetrie (intrinsische Dipolmomente der zwei Niveaus unterscheiden sich). Die physikalische Beschreibung der Wechselwirkung mit dem Laserfeld erfolgt in einem halbklassischen Bild, während bei der Wechselwirkung mit dem elektromagnetischen Vakuum die Quantennatur des Feldes miteinbezogen wird. Somit werden in den darauffolgenden Rechnungen die hier wichtigen Effekte der spontanen Emission berücksichtigt. Für die Lösung unseres Problems wählen wir einen störungstheoretischen Ansatz, sodass wir mit Hilfe einer Mastergleichng die Bewegungsgleichungen des Systems herleiten. Insbesondere wird unser Zwei-Niveau-System mit einem nichtresonanten Laser getrieben, der die Emission eines THz-Photons und eines optischen Photons induziert. In diesem Zusammenhang berechnen wir die Intensität-Intensität-Korrelationsfunktion der beiden Photonen, die ein Maß für die Wahrscheinlichkeit ist, ein Photon direkt nach dem anderen zu detektieren. Da diese beiden Photonen zusätzlich noch eine klassische Cauchy-Schwarz-Ungleichung verletzen, handelt es sich hier um ein nicht-klassisches korreliertes Photonenpaar, das im Gebiet der Quanteninformationstheorie Anwendung findet. Gammaglobulinmoleküle und bestimmte Quantenpunkte weisen z.B. verletzte Inversionssymmetrie auf und können so im Rahmen unseres Modells beschrieben werden. Für die numerischen Berechnungen greifen wir auf die Parameter von Gammaglobulin zurück.

Abstract

The aim of our work is to investigate a pumped two-level system with broken inversion symmetry (intrinsic dipole moments of the two levels differ). We choose a semiclassical description for the interaction with the laser and additionally take into account a quantized environment giving rise to effects of spontaneous emission. We further choose a perturbative approach to derive the equations of motion in a master equation approach. An off-resonant laser induces the emission of a THz-photon and the emission of an optical photon, whose quantum intensity-intensity correlation function we calculate. This function gives a measure for the probability of detecting one photon right after the other. Moreover, we observe the violation of a Cauchy-Schwarz inequality that brings us to the conclusion that we are dealing with a non-classical pair of correlated photons, which are important in the emerging field of quantum information science. This theoretical model may, for example, be applied to gamma globulin molecules or quantum dots. In this thesis we perform the numerical calculations with the parameters of gamma globulin molecules.

Contents

Introduction

The nature of light has long been a mystery to mankind. In 1704, Newton proposed the idea that light consisted of tiny particles leading to the *corpuscular theory of light* in his work "Opticks". However, 14 years earlier, in 1690, Huygens presented his theory of light in the form of *wave optics*. It was only in the 19th century that physicists dismissed Newton's idea after important experimental work by T. Young in 1801 on the *two-slit diffraction pattern* that was explained by the wave hypothesis. Further experimental and theoretical evidence for Huygens' wave hypothesis was given by the discovery of electromagnetic phenomena by Ampère (1820, 1825), Oersted (1820), Faraday (1831), the formulation of *classical electromagnetic theory* by Maxwell in 1864, and finally by the discovery of *electromagnetic waves* by Hertz in 1887. Until the late 19th century the physics community believed that physics had almost received its final shape. No ground-breaking discoveries were expected anymore. Maxwell's equations explained most of the electromagnetic phenomena including the propagation of light as an electromagnetic wave and *Newtonian mechanics* had been accepted as the law describing celestial mechanics. People were so convinced about the uniformity of physical theories that Max Planck was advised by his teacher Phillip Jolly not to study physics [1], since there wouldn't be much work for him to do in research afterwards. This changed in 1900, a particularly important year for physics. Prior to 1900, theory failed to explain the experimental data of *black-body radiation*. Only the description for long wavelengths seemed to be correct, whereas the description for short wavelengths was completely wrong, a circumstance known nowadays as the *ultraviolet catastrophe*. In 1900 Max Planck first derived his well-known law describing black-body radiation empirically and shortly after proposed a bold theoretical derivation, claiming that the oscillator energy is not continuous, but discrete. He also introduced the famous *Planck constant* h that has the unit of an action [2]. This work was presented in front of the German physics community on the 14th of December 1900, which is considered the birthday of Quantum Mechanics. This strange new theory describing the dynamics of the smallest particles is now considered so important that Nobel prize winner Leon Lederman said that Quantum Mechanics was responsible for one third of the American gross domestic product [3].

Planck's work was so fruitful that even 17 years later Einstein was still intrigued by the subject and published a new derivation of the formula. In the form of a rate equation, he proposed a phenomenological model describing the interaction of matter (in form of a two-level system) and radiation based on three fundamental processes: stimulated emission, stimulated absorption and *spontaneous emission*, where the latter process cannot be understood in semiclassical terms. Assuming that radiative energy is homogeneous and isotropic in space and assuming thermal equilibrium between the two levels, Einstein's model correctly reproduces Planck's law for black-body radiation.

In 1925-26, W. Heisenberg [4], E. Schrödinger [5] and P. A. M. Dirac [6] developed the rigorous formalism of matrix mechanics, wave mechanics, and unified quantum mechanics and M. Born proposed the probabilistic interpretation of the absolute square of the wave function. In 1927, P. A. M. Dirac finally formulated the *Quantum theory of radiation* [7], which describes the emission and absorption of radiation in a fundamental way and is consistent with Einstein's coefficients. This was the first time that the quantum of light, the *photon*, was given a precise definition, by noting that the dynamics of a single-mode free field is the same as of a quantized harmonic oscillator. Thus the theoretical background for the conception of *optical amplifiers* was available, leading C. H. Townes to construct the first maser (Microwave Amplification by Stimulated Emission of Radiation) in 1954 [8, 9] followed by the ruby laser (Light Amplification by Stimulated Emission of Radiation) by T. Maiman [10] and the helium-neon gas laser by A. Javan, W. Bennett and D. Herriott in 1960 [11]. With the advent and further development of the laser, physicists had a powerful tool to control quantum systems. The new field of *Quantum Optics* began to emerge, which M. O. Scully and M. S. Zubairy describe in the preface of their famous textbook [12], as "the union of quantum field theory and physical optics". For further detail see [13–16], which have very nice introductions on the history of quantum mechanics and partially inspired my own.

The next important question to answer was whether there was any experimental evidence of the *quantum nature of light*. At first glance, it seemed that interference experiments could be well explained by both classical and quantum theories of light. While the classical theory explains Young's double slit experiment with the interference of classical electromagnetic waves, the quantum theory predicts the same results by considering the interference of wave functions. It was not until the 1950s, however, that *Hanbury-Brown* and *Twiss* conducted their famous experiment measuring the intensity-intensity correlation function [17]. Physicists were then able to measure *second-order correlations*, which turned out to be key to determining the quantum nature of light. In classical coherence theory the second-order correlations satisfy a series of inequalities, while in quantum coherence theory the same second-order correlations might violate them. Thus we speak of light having a quantum nature, if its second-order correlation function violates a classical inequality [18]. The Hanbury-Brown and Twiss technique was initially designed to measure the size of astronomical objects, but has found many different areas of application [19]. The main areas are condensed matter [20, 21] and atomic physics [22].

With the rise and development of quantum mechanics and quantum optics, a new field of research emerged. Some scientists believed that with quantum effects, it was possible to send information at a speed faster than that of light, contradicting the principles of Einstein's theory of special relativity. The answer to this question can be reduced to another question, whether it is possible to clone an unknown quantum state or not. Wootters, Zurek [23] and Dieks [24] showed in the early 80s of the last century that this was not possible, a fact that is now known as the *no-cloning theorem*. This marked the beginning of quantum computation and information science [25]: "Quantum computation and quantum information is the study of the information processing tasks that can be accomplished using quantum mechanical systems" [25]. Although theoretical considerations are already at an advanced stage, the experimental realization is an ongoing challenge in present-day research. There are several candidates that allow *experimental realization*: photons and non-linear optical media, cavity quantum electrodynamics devices, ion traps, and nuclear magnetic resonance with molecules. With these techniques it is possible to partially control and study simple quantum systems and thus implement the theoretical ideas of quantum information science. While classical information theory defines the bit as the fundamental quantity with possible values of either "0" or "1", quantum information theory uses the qubit as fundamental quantity, which may not only acquire values of "0" and "1", but arbitrary quantum mechanical superpositions of both states. Together with the measurement process of quantum mechanics and quantum entanglement, this is the main difference responsible for all the peculiar effects of quantum computation and information science. Secure information transmission and new and more efficient algorithms are just a few examples of new possibilities [26], of which the Shor algorithm is the most prominent example. It has been shown that the problem of factoring an integer could be solved in polynomial time, which is a crucial advantage over the classical algorithm that just solves the problem in exponential time [27]. This classically intractable problem lies at the heart of modern cryptography and is thus of great interest. In order to use optical systems for implementation, there is a need for *correlated or even entangled photon pairs*. In that context, it has been shown that there is a strong relation between photon pairs that violate a Cauchy-Schwarz inequality and a pair of entangled photons [28]. Currently there are a series of experimental techniques available to produce entangled photons such as parametric down conversion [29], four-wave mixing [30, 31], and electromagnetically induced transparency [30]. An atomic memory for correlated photon states has even been experimentally realized, playing an essential role for quantum communication over long distances [32–34]. A quantum network [35] that allows quantum communication consists of interconnected nodes, which are made of different physical systems. Since every physical system has a different characterisitc frequency, it is necessary to produce entangled photons of different wavelengths. Although considerable advances have been achieved in the last years, there is still a lack of sources for tunable correlated photon pairs [36] of different frequencies [37], which we will explore here.

Therefore, it is of great interest to investigate quantum systems interacting with electromagnetic fields and to study different types of correlations between light. Not only does this contribute to fundamental research in the field of quantum optics, but it also helps to further develop all the possibilities of quantum communication.

The *subject of this work* is directly related to the field of quantum optics, the field that studies the light-matter interaction and has applications in quantum computation and information science. We give a *theoretical description* of a driven quantum system which emits light exhibiting properties that may only be explained in a quantum picture. In our model we choose a quantum mechanical description of matter, meaning that the energy levels of the system are quantized. For our purposes the approximation of a single two-level system is sufficient. We will often refer to it as atoms, although we regard any emitter with the necessary conditions. In order to account for quantum processes such as spontaneous emission, we consider the interaction of matter with a quantized electromagnetic vacuum field, which allows vacuum fluctuations [38]. The dynamics of the system are mainly due to a driving classical laser field. The framework we use to describe the physics of the system is non-relativistic quantum mechanics. In principle it is possible to make predictions about the dynamics by setting up a Hamiltonian, and a wave function and solving the time-dependent Schrödinger equation. However, for such a non-trivial system as ours this is a rather difficult task, so we will make use of different and more sophisticated techniques such as the master equation approach. The necessary conditions for our model are found in *single molecules and single quantum dots* for example. In addition to transition dipole moments between the energy levels, they also have an intrinsic and different dipole moment for each level. We stress the fact that we do not use the very common rotating wave approximation, but choose a *perturbative approach*, with which we are able to derive different non-linear processes of spontaneous emission, between which we will calculate magnitudes of intensity-intensity correlation. The laser used to drive the system is slightly *off-resonant*, which is a key ingredient for the non-linear spontaneous emission.

The particular focus of this thesis lies in the investigation of the intensity-intensity correlation between a photon in the THz-regime and an optical photon. It turns out that we deal with a strongly correlated non-classical photon pair of different frequencies that violates a classical Cauchy-Schwarz inequality and as mentioned before finds direct application in a quantum network. We also calculate the intensity of the THz-photons, which may be regarded as a source for tunable THz-radiation. Up to the present, there is still a lack of reliable THz-sources, since neither optical nor microwave techniques are efficiently applicable. Therefore it is an interesting subject of ongoing research [39–41].

We will start off with a physical description of our system by setting up a Hamiltonian in Ch. I. We consider a quantum two-level system that interacts with a *fully quantized electromagnetic environment* and a strong *driving classical field* in the well-known dipole approximation. The Hamiltonian takes into account the energy of the two-level system and the environment, the interaction energy of the coupling to the laser and the matter-environment interaction term, containing energy-conserving as well as energy non-conserving terms. At the same time we will introduce and demonstrate well-known concepts of matter-field interactions. In Ch. II we will carry out some manipulations on our Hamiltonian to simplify the next calculations. At first we will perform a *unitary transformation* going into an interaction-like picture. The purpose is to remove the explicit time-dependence from the most important terms and thus smoothen the way for perturbation theory that will approximate the Hamiltonian and further simplify it. Throughout Ch. III this Hamiltonian is used to derive the equations of motion following the *master equation approach* and by making use of the *Born-Markov approximation*. We will also define the different decay rates. In Ch. IV we are able to physically interpret the master equation of the system. Different processes of spontaneous emission are identified and discussed. Numerical calculations are explicitly done using the parameters of gamma globulin molecules and quantum dots, which are examples for systems with broken inversion symmetry. They are briefly discussed. The intensities of two different emission processes and their second-order intensity-intensity correlation function are calculated, which allows us to derive their statistical properties. Finally, we probe the system for quantum effects in the emission of light and specially observe the violation of a Cauchy-Schwarz inequality. Throughout the thesis we have omitted several technical calculations, which nevertheless are important for a complete understanding of this work and can be found in the appendix. We finish by giving a summary of the whole work, suggestions for the implications of this work and by listing the bibliography.

Conventions

Throughout the whole work we will stick to the same notation and conventions that are going to be defined here. We will make use of the Gaussian-cgs units and give a short introduction on the unit system. For a more rigorous introduction see [42]. The fundamental mechanical units are centimeters for length, grams for mass and seconds for time (cgs). Because of that we define new units for force (dyne)

$$[force] = [mass] \times \frac{[length]}{[time]^2} = \frac{\mathrm{g} \cdot \mathrm{cm}}{\mathrm{s}^2} = 1 \text{ dyne} = 10^{-5}\,\mathrm{N}$$

and for energy (erg)

$$[energy] = [force] \times [length] = \frac{\mathrm{g} \cdot \mathrm{cm}^2}{\mathrm{s}^2} = 1 \text{ erg} = 10^{-7}\,\mathrm{J}.$$

In the following we define a way of characterizing electromagnetic quantities and their relation to the SI-unit system. We set the following quantity equal to one

$$4\pi\varepsilon_0 = \frac{10^7}{c^2} \cdot \frac{\mathrm{C}^2}{\mathrm{N} \cdot \mathrm{s}^2} = 1,$$

where c is the speed of light in vacuum and ε_0 is the electric vacuum permittivity. To obtain the equivalent for 1 Coulomb (SI-units) in cgs-units, we have to solve for it

$$1\,\mathrm{C} = 10^{-\frac{7}{2}}\, c\,\mathrm{s}\,\sqrt{\mathrm{N}} = 2.998 \times 10^9 \mathrm{esu},$$

where we have defined the electrostatic unit (esu) as

$$1 \cdot \mathrm{cm}\sqrt{\mathrm{dyne}} = 1 \cdot \mathrm{esu}.$$

The last important change is the requirement that the magnetic field $\boldsymbol{B}$ should have the same units as the electric field $\boldsymbol{E}$. For that purpose we take the law of the Lorenz force in SI-units

$$F = q(\frac{\boldsymbol{v}}{c} \times \boldsymbol{B}c + \boldsymbol{E})$$

and redefine the magnetic field $\boldsymbol{B}$ (it absorbs the c)

$$F = q(\frac{\boldsymbol{v}}{c} \times \boldsymbol{B} + \boldsymbol{E}).$$

The magnetic and electric fields now have the same units in the Lorenz law above.

After having defined the unit system, we will now give a brief overview of the used physical constants, definitions, and the mathematical notation. We will also make use of the "braket" notation introduced by Dirac [43].

Physical Constants and definitions	
$\hbar = 1.054^{-27}\text{erg}\cdot\text{s}$	Reduced Planck constant
$e = 4.801 \times 10^{-10}\text{esu}$	Elementary charge
$c = 2.997 \times 10^{10}\text{cm/s}$	Speed of light in vacuum
$S^{+} = \|2\rangle\langle 1\|$	Atomic raising operator
$S^{-} = \|1\rangle\langle 2\|$	Atomic lowering operator
$S_z = \frac{1}{2}(\|2\rangle\langle 2\| - \|1\rangle\langle 1\|)$	Atomic inversion operator
H	Hamiltonian
$\wp_{ij} = e\langle i\|\boldsymbol{r}\|j\rangle$	Electric dipole moment operator
$a_{\boldsymbol{k}}$	Annihilation operator of the $\boldsymbol{k}$-th mode
$a_{\boldsymbol{k}}^{\dagger}$	Creation operator of the $\boldsymbol{k}$-th mode
$\Omega = \wp_{12}\cdot\boldsymbol{E}_0/\hbar$	Atomic Rabi frequency

Table I.: Brief overview of used physical constants [44]. $|i\rangle$ (with $i \in \{1,2\}$) are the states of a two-level system.

Mathematical notation	
$\boldsymbol{a}$	Three-vector (bold Latin letters)
$\langle\psi\|, \|\psi\rangle$	Bra and ket vectors
$\dot{x}$	Derivative with respect to time
$A^{\dagger}$	Hermitian conjugate of operator A
$\boldsymbol{a}\cdot\boldsymbol{b}$	Scalar product
$[A,B]$	Commutator
$h.c.$	Hermitian conjugate
$\langle A\rangle$	Expectation value of the operator A
$\delta(x)$	Dirac's delta function
P	Cauchy's principal value
$\hat{\epsilon}_{\boldsymbol{k}}$	Polarization unit vector of the $\boldsymbol{k}$-th mode

Table II.: Summary of mathematical notation, more details can be found in the appendices.

I. Physical description of the system

In the following section we will derive the Hamiltonian describing the dynamics of the considered system: an off-resonantly driven two-level system with both transition dipole moment and intrinsic dipole moments for each level, as illustrated in Fig. I.1. The system has broken inversion symmetry, meaning that the intrinsic dipole moments are different from each other, $|\wp_{11}| \neq |\wp_{22}|$. We start with classical matter-field interaction theory and work in a dipole approximation throughout the whole thesis. On that basis we will add features of the fully quantized theory, in order to describe the interaction of matter and the electromagnetic vacuum field. We will also add features of the semi-classical theory that gives a very good description of the interaction of matter with a driving electromagnetic field, the laser. In the end we will have a Hamiltonian of the following form $H = H_F + H_A + H_L + H_1$, where H_F stands for the electromagnetic field energy, H_A for the atomic energy, H_L for the atom-laser interaction energy and H_1 for the atom-vacuum interaction energy. We basically follow the treatment of M. O. Scully and M. S. Zubairy [12] that can also be found in many other standard textbooks on Quantum Optics.

I.1. Matter-field interaction

I.1.1. Classical interaction

We will start off from the well-known Hamiltonian describing the interaction of an electron of charge e, mass m and momentum $\boldsymbol{p}$ bound to a central electrostatic potential $V(r)$ immersed in an electromagnetic field described by the potentials $\boldsymbol{A}(\boldsymbol{r},t)$ and $U(\boldsymbol{r},t)$. The Hamiltonian H is given by

$$H = \frac{1}{2m}\left(\boldsymbol{p} - e\boldsymbol{A}(\boldsymbol{r},t)\right)^2 + eU(\boldsymbol{r},t) + V(r). \qquad \text{(I.1)}$$

The first approximation we make is the well-known dipole approximation, which is valid, if the field wavelength is much larger than the atomic size. In our case we drive the system with optical frequencies, which correspond roughly to a wavelength of 10^{-6}m. The order of magnitude of the size of big atoms and molecules is about 10^{-9}m. These

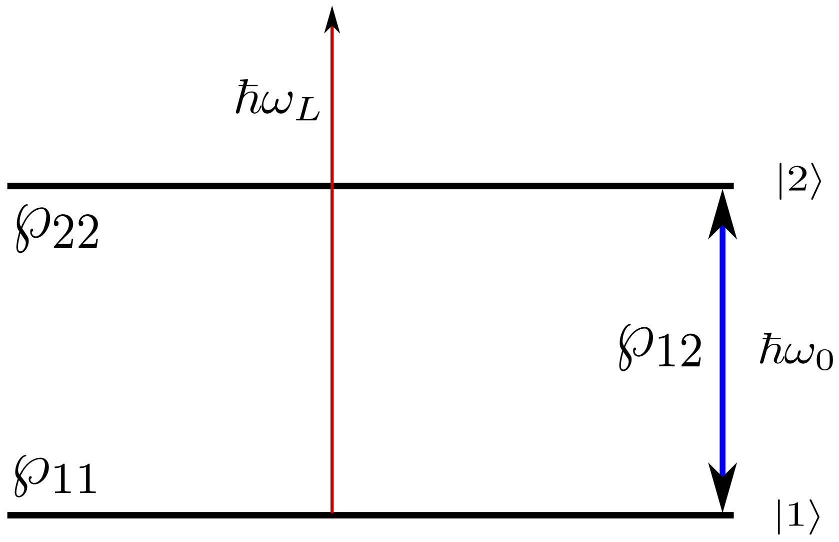

Figure I.1.: The figure illustrates our considered two-level system that is driven by an off-resonant laser of frequency ω_L. The transition frequency is ω_0, the transition dipole moment is $\wp_{12}$ and the intrinsic dipole moments for each level are $\wp_{11}$ and $\wp_{22}$.

are well-known values that support the validity of the approximation. The spatial dependence of the electromagnetic field can then be neglected, leading us to the following Hamiltonian

$$H = H_0 + H_1 = \frac{p^2}{2m} + V(r) - e\boldsymbol{r} \cdot \boldsymbol{E}(\boldsymbol{r}_0, t), \tag{I.2}$$

where the electromagnetic field is described by $\boldsymbol{E}(\boldsymbol{r}_0, t)$ and $\boldsymbol{r}_0$ is the position of the nucleus. $H_0 = p^2/(2m) + V(r)$ is the free part and $H_1 = -e\boldsymbol{r} \cdot \boldsymbol{E}(\boldsymbol{r}_0, t)$ is the interacting part of the Hamiltonian.

I.1.2. Quantum theory of interaction

In order to describe our system, we have to take into account the interaction with the vacuum modes, for which the quantization of the electromagnetic field is necessary. The occuring effects of quantum vacuum fluctuations are responsible for the processes of spontaneous emission and are thus important for our present work. Up to this point we just made semi-classical considerations, whereas now we are taking elements of a fully quantized theory into account.

First we consider the free part H_0. In a quantized theory it can be written as the sum of the energy of the two-level system H_A and the energy of the electromagnetic field H_F,

$$H_0 = H_A + H_F. \tag{I.3}$$

In a two-level system, the basis $\{|1\rangle, |2\rangle\}$ forms a complete set of orthonormal energy eigenstates, with respective energies $\hbar\omega_1$ and $\hbar\omega_2$. It then follows from the completeness relation

$$|1\rangle\langle 1| + |2\rangle\langle 2| = 1 \tag{I.4}$$

and the time-independent Schrödinger equation $H_A|i\rangle = E_i|i\rangle$, $i \in \{1, 2\}$ that

$$\begin{aligned} H_A &= H_A(|1\rangle\langle 1| + |2\rangle\langle 2|) = E_1|1\rangle\langle 1| + E_2|2\rangle\langle 2| \\ &= \frac{1}{2}(E_1 - E_2)(|1\rangle\langle 1| - |2\rangle\langle 2|) + \frac{1}{2}(E_1 + E_2)(|1\rangle\langle 1| + |2\rangle\langle 2|) \\ &= \hbar\omega_0 S_z + \frac{1}{2}(E_1 + E_2), \end{aligned} \tag{I.5}$$

where we have defined the atomic operator $S_z = (|2\rangle\langle 2| - |1\rangle\langle 1|)/2$ and the energy difference $\hbar\omega_0 = E_2 - E_1$. Since constant factors can be dropped in the Hamiltonian we are left with

$$H_A = \hbar\omega_0 S_z. \tag{I.6}$$

We see that the energy of the two-level system only depends on the atomic operator S_z. For a brief treatment of atomic operators and respective relations, check out the appendix A.1.

The energy of the free field H_F can be described as a sum of quantized harmonic oscillators

$$H_F = \sum_{\boldsymbol{k}} \hbar\omega_{\boldsymbol{k}} \left(a_{\boldsymbol{k}}^{\dagger} a_{\boldsymbol{k}} + \frac{1}{2} \right), \tag{I.7}$$

where each summand represents an electromagnetic field mode with frequency $\omega_{\boldsymbol{k}}$ and $a^\dagger_{\boldsymbol{k}}$, $a_{\boldsymbol{k}}$ represent the creation and annihilation operators of the $\boldsymbol{k}$-th mode, respectively. Here again we can drop constant terms, leaving us with

$$H_F = \sum_{\boldsymbol{k}} \hbar\omega_{\boldsymbol{k}} a^\dagger_{\boldsymbol{k}} a_{\boldsymbol{k}}. \tag{I.8}$$

Now we can turn to the interaction part H_1 of the Hamiltonian in Eq. (I.2). As derived in many standard books of quantum optics (e.g., in [46]), the quantized electric field has the following form

$$\boldsymbol{E}(\boldsymbol{r},t) = i\sum_{\boldsymbol{k}} \hat{\epsilon}_{\boldsymbol{k}} \varepsilon_{\boldsymbol{k}} a_{\boldsymbol{k}} e^{-i\omega_{\boldsymbol{k}}t + i\boldsymbol{k}\cdot\boldsymbol{r}} + h.c., \tag{I.9}$$

where the summation is carried out over all possible sets of values of the wave vector $\boldsymbol{k} = (k_x, k_y, k_z)$, $\hat{\epsilon}_{\boldsymbol{k}}$ is the polarization unit vector, and

$$\varepsilon_{\boldsymbol{k}} = \left(\frac{2\pi\hbar\omega_{\boldsymbol{k}}}{V}\right)^{\frac{1}{2}}, \tag{I.10}$$

which has the dimensions of an electric field and V is a quantization volume. We stress the fact that we are not using SI units, but cgs units.

We evaluate Eq. (I.9) in the dipole approximation ($e^{i\boldsymbol{k}\cdot\boldsymbol{r}} \approx 1$) and assume that the atom is situated at the origin. Further the creation and annihilation operators absorb the time-dependence leading to

$$\boldsymbol{E}(\boldsymbol{r},t) = i\sum_{\boldsymbol{k}} \hat{\epsilon}_{\boldsymbol{k}} \varepsilon_{\boldsymbol{k}} (a_{\boldsymbol{k}}(t) - a^\dagger_{\boldsymbol{k}}(t)). \tag{I.11}$$

Now we can expand the interacting part H_1 of the Hamiltonian in Eq. (I.2) by using the completeness relation (I.4) and the quantized electric field (I.11),

$$
\begin{aligned}
H_1 &= -e\Big(|1\rangle\langle 1| + |2\rangle\langle 2|\Big)\boldsymbol{r}\Big(|1\rangle\langle 1| + |2\rangle\langle 2|\Big)\cdot\boldsymbol{E} \\
&= -e\Big(|1\rangle\langle 1|\boldsymbol{r}|2\rangle\langle 2| + |2\rangle\langle 2|\boldsymbol{r}|1\rangle\langle 1|\Big)\cdot\boldsymbol{E} - e\Big(|1\rangle\langle 1|\boldsymbol{r}|1\rangle\langle 1| + |2\rangle\langle 2|\boldsymbol{r}|2\rangle\langle 2|\Big)\cdot\boldsymbol{E} \\
&= -\Big(\wp_{12}|1\rangle\langle 2| + \wp_{21}|2\rangle\langle 1|\Big)\cdot\boldsymbol{E} - \Big(\wp_{11}|1\rangle\langle 1| + \wp_{22}|2\rangle\langle 2|\Big)\cdot\boldsymbol{E} \\
&= -\Big(\wp_{12}|1\rangle\langle 2| + \wp_{21}|2\rangle\langle 1|\Big)\cdot\boldsymbol{E} - \frac{1}{2}(\wp_{11} + \wp_{22})\cdot\boldsymbol{E} - S_z(\wp_{22} - \wp_{11})\cdot\boldsymbol{E},
\end{aligned}
\tag{I.12}
$$

where we have defined the matrix elements of the electric dipole moment operator

$$
\wp_{ij} = e\langle i|\boldsymbol{r}|j\rangle \qquad\qquad i,j \in \{1,2\}. \tag{I.13}
$$

The diagonal parts $\wp_{11}$ and $\wp_{22}$ stand for the intrinsic electric dipole moments of each energy level, whereas $\wp_{12}$ and $\wp_{21}$ denominate the transition dipole moment operator between the two energy levels. The second term in the final expression of Eq. (I.12) may be neglected, because it does not contain any atomic operator and therefore vanishes in the equations of motion afterwards (see Eq. III.1). The third term contributes to the dephasing of the molecule and is neglected here, since we already keep a dephasing term later on. As we are dealing with a two-level system and we assume that the dipole moments are real, we know that $\wp_{12} = \wp_{21}$, leading us to

$$
\begin{aligned}
H_1 &= -(\wp_{12}|1\rangle\langle 2| + \wp_{21}|2\rangle\langle 1|)\cdot\boldsymbol{E} \\
&= -\wp_{12}(S^+ + S^-)\cdot\boldsymbol{E} \\
&= i\sum_{\boldsymbol{k}}\varepsilon_{\boldsymbol{k}}\hat{\epsilon}_{\boldsymbol{k}}\cdot\wp_{12}(S^+ + S^-)(a_{\boldsymbol{k}}^\dagger - a_{\boldsymbol{k}}),
\end{aligned}
\tag{I.14}
$$

where we have defined the atomic operators $S^+ = |2\rangle\langle 1|$ and $S^- = |1\rangle\langle 2|$.

This interaction Hamiltonian reveals already a lot about basic interaction processes that occur, as very well explained by Orszag in [47]. The term of the Hamiltonian proportional to $S^+a_{\boldsymbol{k}}$ corresponds to a process, in which the two-level system is excited from the ground state $|1\rangle$ to the excited state $|2\rangle$ and one photon is absorbed. The term proportional to $S^-a_{\boldsymbol{k}}^\dagger$ corresponds to the process in which the two-level system is deexcited from the upper state $|2\rangle$ to the ground state $|1\rangle$ together with the emission of a photon. The term proportional to $S^+a_{\boldsymbol{k}}^\dagger$ accounts for a process in which the system is excited and emits a photon and the fourth term, proportional to $S^-a_{\boldsymbol{k}}$, describes a process in which the

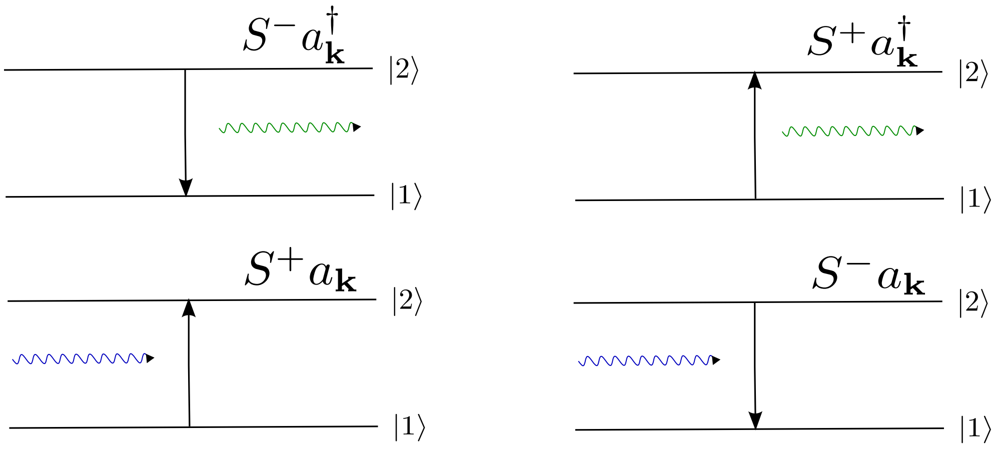

Figure I.2.: Interpretation of four terms in the interaction part of the Hamiltonian in Eq. (I.14). The two processes on the left side correspond to energy-conserving terms and the two processes on the right side correspond to energy non-conserving terms. From Ch. 8 in [47].

system is deexcited and absorbs a photon. The first two processes are clearly energy-conserving, whereas the latter ones are energy non-conserving. All four processes are illustrated in Fig. (I.2).

In the widely used rotating wave approximation, only energy conserving terms are taken into consideration. In our work, however, we chose a perturbative approach, which does not neglect the energy non-conserving terms, which may not be neglected for strong driving and which are important for the non-linear effects of spontaneous emission. In summary we have the following Hamiltonian H for a two-level system, which interacts with a quantized electromagnetic vacuum field:

$$\begin{aligned} H &= H_F + H_A + H_1 \\ &= \sum_{\boldsymbol{k}} \hbar\omega_{\boldsymbol{k}} a^\dagger_{\boldsymbol{k}} a_{\boldsymbol{k}} + \hbar\omega_0 S_z + i \sum_{\boldsymbol{k}} \varepsilon_{\boldsymbol{k}} \hat{\epsilon}_{\boldsymbol{k}} \cdot \wp_{12}(S^+ + S^-)(a^\dagger_{\boldsymbol{k}} - a_{\boldsymbol{k}}). \end{aligned} \tag{I.15}$$

H_F accounts for the energy of the environment, H_A for the energy of the atom, and H_1 describes the interaction of the atom with the environment.

I.1.3. Semiclassical theory of interaction

The next step is to consider the interaction with a laser, which will be described by a classical electromagnetic field. Again we basically follow the treatment of standard textbooks with the difference that we do consider broken inversion symmetry as in [39].

As in Eq. (I.2), we will work in a dipole approximation and the interaction Hamiltonian between the laser and the two-level system takes the following form:

$$H_L = -e\boldsymbol{r} \cdot \boldsymbol{E}(\boldsymbol{r}_0, t). \tag{I.16}$$

Using the completeness relation (I.4) and considering a driving classical field given by a linearly polarized monochromatic plane-wave field in the dipole approximation $\boldsymbol{E} = \boldsymbol{E}_0 \cos(\omega_L t)$ with the laser frequency ω_L and the amplitude $\boldsymbol{E}_0$, we may write Eq. (I.16) as

$$\begin{aligned} H_L &= -e(|1\rangle\langle 1| + |2\rangle\langle 2|)\boldsymbol{r}(|1\rangle\langle 1| + |2\rangle\langle 2|) \cdot \boldsymbol{E}_0 \cos(\omega_L t) \\ &= -\Big(\wp_{11}|1\rangle\langle 1| + \wp_{21}|2\rangle\langle 1| + \wp_{12}|1\rangle\langle 2| + \wp_{22}|2\rangle\langle 2|\Big) \cdot \boldsymbol{E}_0 \cos(\omega_L t) \\ &= -\wp_{12} \cdot \boldsymbol{E}_0(S^+ + S^-)\cos(\omega_L t) - \Big(\wp_{11}(\frac{1}{2} - S_z) + \wp_{22}(\frac{1}{2} + S_z)\Big) \cdot \boldsymbol{E}_0 \cos(\omega_L t) \\ &= -\wp_{12} \cdot \boldsymbol{E}_0(S^+ + S^-)\cos(\omega_L t) - \Big((\wp_{22} - \wp_{11})S_z + \frac{1}{2}\wp_{11} + \frac{1}{2}\wp_{22}\Big) \cdot \boldsymbol{E}_0 \cos(\omega_L t). \end{aligned} \tag{I.17}$$

In the above equation the fast oscillating terms that are not proportional to an atomic operator are dropped, since they vanish in the equations of motion afterwards (see Eq. (III.1)). This leads to

$$\begin{aligned} H_L &= -\wp_{12} \cdot \boldsymbol{E}_0(S^+ + S^-)\cos(\omega_L t) + (\wp_{11} - \wp_{22})S_z \cdot \boldsymbol{E}_0 \cos(\omega_L t) \\ &= -\hbar\Omega(S^+ + S^-)\cos(\omega_L t) + \hbar G S_z \cos(\omega_L t), \end{aligned} \tag{I.18}$$

where we have defined the constants

$$\Omega = \frac{\wp_{12} \cdot \boldsymbol{E}_0}{\hbar}, \tag{I.19}$$

$$G = \frac{(\wp_{11} - \wp_{22}) \cdot \boldsymbol{E}_0}{\hbar}. \tag{I.20}$$

Ω is called the Rabi frequency of the system. In the absence of diagonal dipole moment operators $\wp_{11}$ and $\wp_{22}$, i.e. when $G = 0$, the Rabi frequency is related to the frequency at which the electron oscillates between the two energy levels. We stress the fact that it is different from the frequency ω_L of the driving field. The effect was first observed in a mathematically similar system of a spin in a time-dependent magnetic field [48]. We see that the Rabi frequency Ω is proportional to the amplitude $\boldsymbol{E}_0$ of the driving field.

Thus if we want to control the Rabi oscillations of the system, we have to manipulate the intensity of our laser.

Now we are able to write down the total Hamiltonian we will work with,

$$\begin{aligned} H &= H_F + H_A + H_L + H_1 \\ &= \sum_{\boldsymbol{k}} \hbar\omega_{\boldsymbol{k}} a_{\boldsymbol{k}}^{\dagger} a_{\boldsymbol{k}} + \hbar\omega_0 S_z - \hbar\Omega(S^+ + S^-)\cos(\omega_L t) + \hbar G S_z \cos(\omega_L t) \\ &+ i\sum_{\boldsymbol{k}} (\boldsymbol{g_k}\cdot\boldsymbol{d})(S^+ + S^-)(a_{\boldsymbol{k}}^{\dagger} - a_{\boldsymbol{k}}), \end{aligned} \tag{I.21}$$

where the coupling constant $\boldsymbol{g_k}$ is defined as $\boldsymbol{g_k} = \varepsilon_{\boldsymbol{k}}\hat{\epsilon}_{\boldsymbol{k}}$ and $\boldsymbol{d} = \wp_{12}$.

In summary we have derived the Hamiltonian describing the system illustrated in Fig. I.1. H_F accounts for the energy of the environment; it is a sum over all the electromagnetic field modes, which are described by quantum harmonic oscillators. H_A takes into account the energy of the two-level system. H_L describes the interaction between the classical radiation field and the quantized matter, and H_1 accounts for the interaction between the two-level system and the quantized environment. Thus it is apparent that in our model, both semi-classical and fully quantized approaches are followed. The semi-classical theory is sufficient in explaining the interaction of the driving laser field, while the quantized theory is necessary to describe the electromagnetic vacuum field, which is needed to explain important quantum effects such as spontaneous emission.

II. Manipulation of the Hamiltonian

In the previous Chapter we described the physics of our system by setting up a Hamiltonian. Now we we would be in a position to solve the equations of motion of the system using the time dependent Schrödinger equation, but this turns out to be a difficult task that we do not follow. For that reason we perform a couple of manipulations on the Hamiltonian in order to simplify the calculations afterwards. At first we make a unitary transformation of the whole Hamiltonian to a rotating frame, bringing it to favourable form, with which we use a special form of perturbation theory described in App. B.1. The whole chapter is very technical and in the end we will have the final form of the Hamiltonian, with which we can derive the equations of motion.

II.1. Unitary transformation of the Hamiltonian

As derived in the previous section, the Hamiltonian describing our dynamical system is given by

$$\begin{aligned} H = &\sum_{\boldsymbol{k}} \hbar\omega_{\boldsymbol{k}} a_{\boldsymbol{k}}^\dagger a_{\boldsymbol{k}} + \hbar\omega_0 S_z - \hbar\Omega(S^+ + S^-)\cos(\omega_L t) \\ &+\hbar G S_z \cos(\omega_L t) + i\sum_{\boldsymbol{k}} (\boldsymbol{g}_{\boldsymbol{k}} \cdot \boldsymbol{d})(a_{\boldsymbol{k}}^\dagger - a_{\boldsymbol{k}})(S^+ + S^-). \end{aligned} \tag{II.1}$$

Next we perform the following unitary transformation of the whole Hamiltonian

$$\tilde{H} = e^{\frac{i}{\hbar}\tilde{H}_0 t}(H - \tilde{H}_0)e^{-\frac{i}{\hbar}\tilde{H}_0 t}, \tag{II.2}$$

where

$$\tilde{H}_0 = \sum_{\boldsymbol{k}} \hbar\omega_L a_{\boldsymbol{k}}^\dagger a_{\boldsymbol{k}} + \hbar\omega_L S_z. \tag{II.3}$$

This means that we work in a frame rotating with frequency ω_L. We define the terms in H_0 with the frequency ω_L of the laser and not with ω_0 of the transition or $\omega_{\boldsymbol{k}}$ of the modes. The derivation of the transformed Schrödinger equation (II.2) can be found in App. A.2.

In order to explicitly calculate this expression we have to resort to the very useful Hadamard lemma that can be found in many introductory books on quantum mechanics such as [49].

Hadamard's lemma. $e^{\alpha A} B e^{-\alpha A} = B + \alpha[A,B] + \frac{\alpha^2}{2!}[A,[A,B]] + \ldots$, *where* $\alpha \in \mathbf{R}$ *and* A,B *are linear operators.*

Now we are in a position to calculate the following important relations:

$$e^{i\omega_L t S_z} S^+ e^{-i\omega_L t S_z} = S^+ + i\omega_L t S^+ + \frac{(i\omega_L t)^2}{2!} S^+ + \ldots = S^+ e^{i\omega_L t}, \tag{II.4a}$$

$$e^{i\omega_L t S_z} S^- e^{-i\omega_L t S_z} = S^- - i\omega_L t S^- + \frac{(-i\omega_L t)^2}{2!} S^- + \ldots = S^- e^{-i\omega_L t}, \tag{II.4b}$$

$$e^{i\sum_{\boldsymbol{k}} a^\dagger_{\boldsymbol{k}} a_{\boldsymbol{k}} \omega_L t} a^\dagger_{\boldsymbol{k}} e^{-i\sum_{\boldsymbol{k}} a^\dagger_{\boldsymbol{k}} a_{\boldsymbol{k}} \omega_L t} = a^\dagger_{\boldsymbol{k}} + i\omega_L t a^\dagger_{\boldsymbol{k}} + \frac{(i\omega_L t)^2}{2!} a^\dagger_{\boldsymbol{k}} + \ldots = a^\dagger_{\boldsymbol{k}} e^{i\omega_L t}, \tag{II.4c}$$

$$e^{i\sum_{\boldsymbol{k}} a^\dagger_{\boldsymbol{k}} a_{\boldsymbol{k}} \omega_L t} a_{\boldsymbol{k}} e^{-i\sum_{\boldsymbol{k}} a^\dagger_{\boldsymbol{k}} a_{\boldsymbol{k}} \omega_L t} = a_{\boldsymbol{k}} - i\omega_L t a_{\boldsymbol{k}} + \frac{(-i\omega_L t)^2}{2!} a_{\boldsymbol{k}} + \ldots = a_{\boldsymbol{k}} e^{-i\omega_L t}, \tag{II.4d}$$

where we have also used commutation relations between the atomic operators S^+, S^-, S_z as derived in App. A.1 and the fundamental commutation relation between creation and annihilation operators: $[a_{\boldsymbol{k}'}, a^\dagger_{\boldsymbol{k}}] = \delta_{\boldsymbol{k}'\boldsymbol{k}}$.

Now we will evaluate Eq. (II.2) gradually using the above relations. For a better overview, we divide it into four parts

$$\tilde{H} = \sum_{i=1}^{4} \tilde{H}_i. \tag{II.5}$$

The first two parts are trivial:

$$\tilde{H}_1 = \sum_{\boldsymbol{k}} \hbar(\omega_{\boldsymbol{k}} - \omega_L) a^\dagger_{\boldsymbol{k}} a_{\boldsymbol{k}} + \hbar(\omega_0 - \omega_L) S_z, \tag{II.6}$$

all involved operators commute (atomic operators commute with creation and annihilation operators, since they do not belong to the same subsystem) and

$$\tilde{H}_2 = \hbar G S_z \cos(\omega_L t) = \frac{\hbar G}{2} S_z (e^{i\omega_L t} + e^{-i\omega_L t}), \tag{II.7}$$

where we have just rewritten the cosine function in its complex form.

For the next parts, we need the relations of Eqs. (II.4):

$$\begin{aligned}
\tilde{H}_3 &= -e^{\frac{i}{\hbar}H_0 t}\hbar\Omega(S^+ + S^-)\cos(\omega_L t)e^{-\frac{i}{\hbar}H_0 t} \\
&= -e^{i\omega_L S_z t}\hbar\Omega(S^+ + S^-)e^{-i\omega_L S_z t}\cos(\omega_L t) \\
&= -\hbar\Omega(S^+ e^{i\omega_L t} + S^- e^{-i\omega_L t})\cos(\omega_L t) \\
&= -\frac{\hbar\Omega}{2}(S^+ e^{i\omega_L t} + S^- e^{-i\omega_L t})(e^{i\omega_L t} + e^{-i\omega_L t}) \\
&= -\frac{\hbar\Omega}{2}(S^+ e^{2i\omega_L t} + S^+ + S^- e^{-2i\omega_L t} + S^-),
\end{aligned} \tag{II.8}$$

and

$$\begin{aligned}
\tilde{H}_4 &= e^{\frac{i}{\hbar}H_0 t} i \sum_{\boldsymbol{k}} (\boldsymbol{g_k} \cdot \boldsymbol{d})(a_{\boldsymbol{k}}^\dagger - a_{\boldsymbol{k}})(S^+ + S^-)e^{-\frac{i}{\hbar}H_0 t} \\
&= i \sum_{\boldsymbol{k}} (\boldsymbol{g_k} \cdot \boldsymbol{d})(a_{\boldsymbol{k}}^\dagger e^{i\omega_L t} - a_{\boldsymbol{k}} e^{-i\omega_L t})(S^+ e^{i\omega_L t} + S^- e^{-i\omega_L t}) \\
&= i \sum_{\boldsymbol{k}} (\boldsymbol{g_k} \cdot \boldsymbol{d})(a_{\boldsymbol{k}}^\dagger S^- - a_{\boldsymbol{k}} S^+) + i \sum_{\boldsymbol{k}} (\boldsymbol{g_k} \cdot \boldsymbol{d})(a_{\boldsymbol{k}}^\dagger S^+ e^{2i\omega_L t} - a_{\boldsymbol{k}} S^- e^{-2i\omega_l t}).
\end{aligned} \tag{II.9}$$

The sum of the different parts gives us the final result

$$\begin{aligned}
\tilde{H} &= \tilde{H}_1 + \tilde{H}_2 + \tilde{H}_3 + \tilde{H}_4 \\
&= \sum_{\boldsymbol{k}} \hbar(\omega_{\boldsymbol{k}} - \omega_L) a_{\boldsymbol{k}}^\dagger a_{\boldsymbol{k}} + \hbar(\omega_0 - \omega_L)S_z - \frac{\hbar\Omega}{2}(S^+ + S^-) \\
&\quad - \frac{\hbar\Omega}{2}(S^+ e^{2i\omega_L t} + S^- e^{-2i\omega_L t}) \\
&\quad + \frac{\hbar G}{2} S_z (e^{i\omega_L t} + e^{-i\omega_L t}) + i \sum_{\boldsymbol{k}} (\boldsymbol{g_k} \cdot \boldsymbol{d})(a_{\boldsymbol{k}}^\dagger S^- - a_{\boldsymbol{k}} S^+) \\
&\quad + i \sum_{\boldsymbol{k}} (\boldsymbol{g_k} \cdot \boldsymbol{d})(a_{\boldsymbol{k}}^\dagger S^+ e^{2i\omega_L t} - a_{\boldsymbol{k}} S^- e^{-2i\omega_L t}).
\end{aligned} \tag{II.10}$$

This means that we have transformed the initial Hamiltonian of Eq. (II.1) to the above Hamiltonian. The structure is quite similar, but has some important differences. The frequency of each field mode in the expression of the energy of the field modes $\sum_{\boldsymbol{k}} \hbar(\omega_{\boldsymbol{k}} - \omega_L) a^\dagger_{\boldsymbol{k}} a_{\boldsymbol{k}}$ is now shifted by ω_L. The same also applies to the frequency in the expression of the energy of the atom $\hbar(\omega_0 - \omega_L) S_z$. The coupling of the transition dipole moment $\boldsymbol{d}$ to the classical driving field is now split into two parts, a time independent part $\hbar\Omega/2(S^+ + S^-)$ and a time dependent part $\hbar\Omega/2(S^+ e^{2i\omega_L t} + S^- e^{-2i\omega_L t})$. The most important term that changed is the one describing the interaction between the quantized environment and the two-level system. The energy-conserving part $i \sum_{\boldsymbol{k}} (\boldsymbol{g}_{\boldsymbol{k}} \cdot \boldsymbol{d})(a^\dagger_{\boldsymbol{k}} S^- - a_{\boldsymbol{k}} S^+)$ now is time-independent and the energy non-conserving part is time-dependent, $i \sum_{\boldsymbol{k}} (\boldsymbol{g}_{\boldsymbol{k}} \cdot \boldsymbol{d})(a^\dagger_{\boldsymbol{k}} S^+ e^{2i\omega_L t} - a_{\boldsymbol{k}} S^- e^{-2i\omega_L t})$. With this form of the Hamiltonian it is now convenient to apply perturbation theory, since we have time-dependent parts that can be seen as a perturbation to the time-independent parts

II.2. Time-dependent perturbation theory

At first we explicitly divide the Hamiltonian in a time-independent part

$$\begin{aligned} H := &\sum_{\boldsymbol{k}} \hbar(\omega_{\boldsymbol{k}} - \omega_L) a^\dagger_{\boldsymbol{k}} a_{\boldsymbol{k}} + \hbar(\omega_0 - \omega_L) S_z - \frac{\hbar\Omega}{2}(S^+ + S^-) \\ &+ i \sum_{\boldsymbol{k}} (\boldsymbol{g}_{\boldsymbol{k}} \cdot \boldsymbol{d})(a^\dagger_{\boldsymbol{k}} S^- - a_{\boldsymbol{k}} S^+) \end{aligned} \tag{II.11}$$

and a time-dependent part

$$\begin{aligned} H' := &\frac{\hbar G}{2} S_z (e^{i\omega_L t} + e^{-i\omega_L t}) - \frac{\hbar\Omega}{2}(S^+ e^{2i\omega_L t} + S^- e^{-2i\omega_L t}) \\ &+ i \sum_{\boldsymbol{k}} (\boldsymbol{g}_{\boldsymbol{k}} \cdot \boldsymbol{d})(a^\dagger_{\boldsymbol{k}} S^+ e^{2i\omega_L t} - a_{\boldsymbol{k}} S^- e^{-2i\omega_L t}), \end{aligned} \tag{II.12}$$

which may be regarded as a perturbation to H (note that $\tilde{H}$ of the previous part has become H again for reasons of easier notation). We may assume that G (see Eq. (I.19)), the Rabi frequency Ω and the coupling $\boldsymbol{g}_{\boldsymbol{k}} \cdot \boldsymbol{d}/\hbar$ are all much smaller than the laser frequency ω_L: $G << \omega_L$, $\Omega << \omega_L$ and $\boldsymbol{g}_{\boldsymbol{k}} \cdot \boldsymbol{d}/\hbar << \omega_L$. This will become clearer afterwards with the explicit values in the numerical calculations. Therefore, we can use second order perturbation theory as shown in App. B.1 to approximate the time-dependent Hamiltonian

$$\begin{aligned} H_{pert} &= -\frac{i}{\hbar} H' \int dt H' \\ &= -\frac{i}{\hbar}(H'_1 + H'_2 + H'_3)(\int dt H'_1 + \int dt H'_2 + \int dt H'_3). \end{aligned} \tag{II.13}$$

In order to have a better overview of the calculations, we split the Hamiltonian H' into three parts and define $H_1' = \hbar G/2\ S_z(e^{i\omega_L t}+e^{-i\omega_L t})$, $H_2' = -\hbar\Omega/2\ (S^+e^{2i\omega_L t}+S^-e^{-2i\omega_L t})$ and $H_3' = i\sum_{\boldsymbol{k}}(\boldsymbol{g_k}\cdot\boldsymbol{d})(a_{\boldsymbol{k}}^\dagger S^+e^{2i\omega_L t} - a_{\boldsymbol{k}}S^-e^{-2i\omega_L t})$. At first we calculate the needed integrals

$$\begin{aligned}\int dtH_1' &= \int dt\frac{\hbar G}{2}S_z(e^{i\omega_L t}+e^{-i\omega_L t})\\ &= \frac{\hbar G}{2i\omega_L}S_z(e^{i\omega_L t}-e^{-i\omega_L t}),\end{aligned} \tag{II.14}$$

$$\begin{aligned}\int dtH_2' &= -\int dt\frac{\hbar\Omega}{2}(S^+e^{2i\omega_L t}+S^-e^{-2i\omega_L t})\\ &= -\frac{\hbar\Omega}{4i\omega_L}(S^+e^{2i\omega_L t}-S^-e^{-2i\omega_L t}),\end{aligned} \tag{II.15}$$

and

$$\begin{aligned}\int dtH_3' &= \int dt\, i\sum_{\boldsymbol{k}}(\boldsymbol{g_k}\cdot\boldsymbol{d})(a_{\boldsymbol{k}}^\dagger S^+e^{2i\omega_L t} - a_{\boldsymbol{k}}S^-e^{-2i\omega_L t})\\ &= \frac{1}{2\omega_L}\sum_{\boldsymbol{k}}(\boldsymbol{g_k}\cdot\boldsymbol{d})(a_{\boldsymbol{k}}^\dagger S^+e^{2i\omega_L t} + a_{\boldsymbol{k}}S^-e^{-2i\omega_L t}).\end{aligned} \tag{II.16}$$

At this point it is very clear that fast oscillating terms are more supressed and therefore contribute less to the Hamiltonian. In the following they are always neglected when compared to slowly oscillating terms. So we see that the unitary transformation of the beginning was useful, since the less important terms of the Hamiltonian became faster oscillating time-dependent terms. Now the calculations of Eq. (II.13) will be performed in various steps. For that purpose we define the nine different parts

$$H_{pert}^{mn} = -\frac{i}{\hbar}H_m'\int dtH_n', \tag{II.17}$$

with $m \in \{1,2,3\}$ and $n \in \{1,2,3\}$. They are calculated one by one. The first part

$$\begin{aligned}H_{pert}^{1,1} &= -\frac{i}{\hbar}\frac{\hbar G}{2}S_z(e^{i\omega_L t}+e^{-i\omega_L t})\frac{\hbar G}{2i\omega_L}S_z(e^{i\omega_L t}-e^{-i\omega_L t})\\ &= -\frac{\hbar G^2}{16\omega_L}(e^{2i\omega_L t}-e^{-2i\omega_L t}),\end{aligned} \tag{II.18}$$

does not depend on any operator and because of the time-dependent phase $e^{2i\omega_L t}$ it may be considered as fast oscillating and will be neglected in the following. The second term

$$\begin{aligned} H_{pert}^{1,2} &= -\frac{i}{\hbar}\frac{\hbar G}{2} S_z(e^{i\omega_L t} + e^{-i\omega_L t})\frac{i\hbar\Omega}{4\omega_L}(S^+ e^{2i\omega_L t} - S^- e^{-2i\omega_L t}) \\ &= \frac{\hbar\Omega G}{16\omega_L}(S^+ e^{i\omega_L t} + S^- e^{-i\omega_L t}), \end{aligned} \tag{II.19}$$

has a similar structure to a term of the time-independent part (II.11) of the Hamiltonian, except for the oscillating phase. That is why it can be neglected too. In the third part

$$\begin{aligned} H_{pert}^{1,3} &= -\frac{i}{\hbar}\frac{\hbar G}{2} S_z(e^{i\omega_L t} + e^{-i\omega_L t})\frac{1}{2\omega_L}\sum_{\boldsymbol{k}}(\boldsymbol{g_k}\cdot\boldsymbol{d})(a_{\boldsymbol{k}}^\dagger S^+ e^{2i\omega_L t} + a_{\boldsymbol{k}} S^- e^{-2i\omega_L t}) \\ &= \frac{G}{4i\omega_L} S_z \sum_{\boldsymbol{k}}(\boldsymbol{g_k}\cdot\boldsymbol{d})(a_{\boldsymbol{k}}^\dagger S^+ e^{3i\omega_L t} + a_{\boldsymbol{k}}^\dagger S^+ e^{i\omega_L t} \\ &+ a_{\boldsymbol{k}} S^- e^{-i\omega_L t} + a_{\boldsymbol{k}} S^- e^{-3i\omega_L t}), \end{aligned}$$

we neglect fast oscillating terms again, remaining with the following expression

$$\begin{aligned} H_{pert}^{1,3} &= \frac{G}{4i\omega_L} S_z \sum_{\boldsymbol{k}}(\boldsymbol{g_k}\cdot\boldsymbol{d})(a_{\boldsymbol{k}}^\dagger S^+ e^{i\omega_L t} + a_{\boldsymbol{k}} S^- e^{-i\omega_L t}) \\ &= \frac{G}{8i\omega_L} \sum_{\boldsymbol{k}}(\boldsymbol{g_k}\cdot\boldsymbol{d})(a_{\boldsymbol{k}}^\dagger S^+ e^{i\omega_L t} + a_{\boldsymbol{k}} S^- e^{-i\omega_L t}). \end{aligned} \tag{II.20}$$

The fourth term

$$\begin{aligned} H_{pert}^{2,1} &= \frac{i}{\hbar}\frac{\hbar\Omega}{2}(S^+ e^{2i\omega_L t} + S^- e^{-2i\omega_L t})\frac{\hbar G}{2i\omega_L} S_z(e^{i\omega_L t} - e^{-i\omega_L t}) \\ &= \frac{\hbar G\Omega}{4\omega_L}(S^- e^{-i\omega_L t} - S^+ e^{i\omega_L t}) S_z \\ &= \frac{\hbar G\Omega}{8\omega_L}(S^- e^{-i\omega_L t} + S^+ e^{i\omega_L t}), \end{aligned} \tag{II.21}$$

is neglected for the same reason as for Eq. (II.19). This correction

$$\begin{aligned} H_{pert}^{2,2} &= -\frac{i}{\hbar}\frac{\hbar\Omega}{2}(S^+e^{2i\omega_L t} + S^-e^{-2i\omega_L t})\frac{\hbar\Omega}{4i\omega_L}(S^+e^{2i\omega_L t} - S^-e^{-2i\omega_L t}) \\ &= -\frac{\hbar\Omega^2}{8\omega_L}(-S^+S^- + S^-S^+) \\ &= \frac{\hbar\Omega^2}{4\omega_L}S_z, \end{aligned} \tag{II.22}$$

of the Hamiltonian shifts the energy $\hbar\omega_0$ of the two-level system by $\hbar\Omega^2/4\omega_L$. Since the Rabi frequency Ω (I.19) depends on the amplitude of the driving field $\boldsymbol{E}_0$, this is an effect of strong-field lasers. It has been discovered by F. Bloch and A. Siegert in 1940 [50] and is therefore called Bloch-Siegert shift. The sixth term

$$\begin{aligned} H_{pert}^{2,3} &= \frac{i}{\hbar}\frac{\hbar\Omega}{2}(S^+e^{2i\omega_L t} + S^-e^{-2i\omega_L t})\frac{1}{2\omega_L}\sum_{\boldsymbol{k}}(\boldsymbol{g_k}\cdot\boldsymbol{d})(a_{\boldsymbol{k}}^\dagger S^+e^{2i\omega_L t} + a_{\boldsymbol{k}}S^-e^{-2i\omega_L t}) \\ &= \frac{i\Omega}{8\omega_L}\sum_{\boldsymbol{k}}(\boldsymbol{g_k}\cdot\boldsymbol{d})(a_{\boldsymbol{k}}^\dagger + a_{\boldsymbol{k}}) + \frac{i\Omega}{4\omega_L}\sum_{\boldsymbol{k}}(\boldsymbol{g_k}\cdot\boldsymbol{d})(a_{\boldsymbol{k}}S_z - a_{\boldsymbol{k}}^\dagger S_z), \end{aligned} \tag{II.23}$$

cancels exactly with the first part of (II.25). The seventh term

$$\begin{aligned} H_{pert}^{3,1} &= -\frac{i}{\hbar}i\sum_{\boldsymbol{k}}(\boldsymbol{g_k}\cdot\boldsymbol{d})(a_{\boldsymbol{k}}^\dagger S^+e^{2i\omega_L t} - a_{\boldsymbol{k}}S^-e^{-2i\omega_L t})\frac{\hbar G}{2i\omega_L}S_z(e^{i\omega_L t} - e^{-i\omega_L t}) \\ &= \frac{G}{4i\omega_L}\sum_{\boldsymbol{k}}(\boldsymbol{g_k}\cdot\boldsymbol{d})(a_{\boldsymbol{k}}^\dagger S^+e^{i\omega_L t} - a_{\boldsymbol{k}}S^-e^{-i\omega_L t}), \end{aligned} \tag{II.24}$$

has the same structure of Eq. (II.20). This means that we may just add them up, as it is done in the end. The eighth term

$$\begin{aligned} H_{pert}^{3,2} &= -\frac{i}{\hbar}i\sum_{\boldsymbol{k}}(\boldsymbol{g_k}\cdot\boldsymbol{d})(a_{\boldsymbol{k}}^\dagger S^+e^{2i\omega_L t} - a_{\boldsymbol{k}}S^-e^{-2i\omega_L t})\frac{\hbar\Omega}{4i\omega_L}(S^-e^{-2i\omega_L t} - S^+e^{2i\omega_L t}) \\ &= \frac{\Omega}{8i\omega_L}\sum_{\boldsymbol{k}}(\boldsymbol{g_k}\cdot\boldsymbol{d})(a_{\boldsymbol{k}}^\dagger + a_{\boldsymbol{k}}) + \frac{\Omega}{4i\omega_L}\sum_{\boldsymbol{k}}(\boldsymbol{g_k}\cdot\boldsymbol{d})(a_{\boldsymbol{k}}^\dagger S_z - a_{\boldsymbol{k}}S_z), \end{aligned} \tag{II.25}$$

cancels exactly with the first part of Eq. (II.23) and the last term

$$\begin{aligned} H^{3,3}_{pert} &= -\frac{i}{\hbar} i \sum_{\boldsymbol{k}} (\boldsymbol{g_k} \cdot \boldsymbol{d})(a^\dagger_{\boldsymbol{k}} S^+ e^{2i\omega_L t} - a_{\boldsymbol{k}} S^- e^{-2i\omega_L t}) \\ &\quad \times \frac{1}{2\omega_L} \sum_{\boldsymbol{k}} (\boldsymbol{g_k} \cdot \boldsymbol{d})(a^\dagger_{\boldsymbol{k}} S^+ e^{2i\omega_L t} + a_{\boldsymbol{k}} S^- e^{-2i\omega_L t}) \\ &= \frac{1}{2\hbar\omega_L} \sum_{\boldsymbol{k},\boldsymbol{k}'} (\boldsymbol{g_k} \cdot \boldsymbol{d})(\boldsymbol{g_{k'}} \cdot \boldsymbol{d})(a^\dagger_{\boldsymbol{k}} S^+ a_{\boldsymbol{k}'} S^- - a_{\boldsymbol{k}} a^\dagger_{\boldsymbol{k}'} S^- S^+), \end{aligned} \tag{II.26}$$

is neglected due to its higher order, which would lead to a negligible correction.

In summary we have neglected the terms in Eqs. (II.18), (II.19), (II.21), and (II.26), for reasons that are explained above. Eq. (II.12) therefore reduces to

$$\begin{aligned} H_{pert} &= \frac{G}{8i\omega_L} \sum_{\boldsymbol{k}} (\boldsymbol{g_k} \cdot \boldsymbol{d})(a^\dagger_{\boldsymbol{k}} S^+ e^{i\omega_L t} - a_{\boldsymbol{k}} S^- e^{-i\omega_L t}) + \frac{\hbar\Omega^2}{4\omega_L} S_z \\ &\quad + \frac{\Omega}{8i\omega_L} \sum_{\boldsymbol{k}} (\boldsymbol{g_k} \cdot \boldsymbol{d})(a_{\boldsymbol{k}} + a^\dagger_{\boldsymbol{k}}) - \frac{\Omega}{4i\omega_L} \sum_{\boldsymbol{k}} (\boldsymbol{g_k} \cdot \boldsymbol{d})(a_{\boldsymbol{k}} S_z - a^\dagger_{\boldsymbol{k}} S_z) \\ &\quad + \frac{G}{4i\omega_L} \sum_{\boldsymbol{k}} (\boldsymbol{g_k} \cdot \boldsymbol{d})(a^\dagger_{\boldsymbol{k}} S^+ e^{i\omega_L t} - a_{\boldsymbol{k}} S^- e^{-i\omega_L t}) \\ &\quad - \frac{\Omega}{8i\omega_L} \sum_{\boldsymbol{k}} (\boldsymbol{g_k} \cdot \boldsymbol{d})(a^\dagger_{\boldsymbol{k}} + a_{\boldsymbol{k}}) + \frac{\Omega}{4i\omega_L} \sum_{\boldsymbol{k}} (\boldsymbol{g_k} \cdot \boldsymbol{d})(a^\dagger_{\boldsymbol{k}} S_z - a_{\boldsymbol{k}} S_z) \\ &= \frac{\hbar\Omega^2}{4\omega_L} S_z + \frac{3G}{8i\omega_L} \sum_{\boldsymbol{k}} (\boldsymbol{g_k} \cdot \boldsymbol{d})(a^\dagger_{\boldsymbol{k}} S^+ e^{i\omega_L t} - a_{\boldsymbol{k}} S^- e^{-i\omega_L t}) \\ &\quad - \frac{\Omega}{2i\omega_L} \sum_{\boldsymbol{k}} (\boldsymbol{g_k} \cdot \boldsymbol{d})(a_{\boldsymbol{k}} - a^\dagger_{\boldsymbol{k}}) S_z. \end{aligned} \tag{II.27}$$

Combining this Hamiltonian with the time-independent part in Eq. (II.11) leads to the total Hamiltonian

$$\begin{aligned} H &= \sum_{\boldsymbol{k}} \hbar(\omega_{\boldsymbol{k}} - \omega_L) a^\dagger_{\boldsymbol{k}} a_{\boldsymbol{k}} + \hbar(\omega_0 - \omega_L + \frac{\Omega^2}{4\omega_L}) S_z - \frac{\hbar\Omega}{2}(S^+ + S^-) \\ &\quad + i \sum_{\boldsymbol{k}} (\boldsymbol{g_k} \cdot \boldsymbol{d})(a^\dagger_{\boldsymbol{k}} S^- - a_{\boldsymbol{k}} S^+) + \frac{3G}{8i\omega_L} \sum_{\boldsymbol{k}} (\boldsymbol{g_k} \cdot \boldsymbol{d})(a^\dagger_{\boldsymbol{k}} S^+ e^{i\omega_L t} - a_{\boldsymbol{k}} S^- e^{-i\omega_L t}) \\ &\quad + \frac{i\Omega}{2\omega_L} \sum_{\boldsymbol{k}} (\boldsymbol{g_k} \cdot \boldsymbol{d})(a_{\boldsymbol{k}} - a^\dagger_{\boldsymbol{k}}) S_z, \end{aligned} \tag{II.28}$$

where in summary we keep only the slowest time-dependent terms and for which we define

$$H_0 = \hbar(\omega_0 - \omega_L + \frac{\Omega^2}{4\omega_L})S_z - \frac{\hbar\Omega}{2}(S^+ + S^-). \tag{II.29}$$

Perturbation theory was the last change carried out on the Hamiltonian, meaning that we now have the final form we will work with. A few changes from the Hamiltonian of Eq. (II.10) can be observed. At first we note that the energy of the atom is shifted by the Bloch-Siegert shift $\hbar\Omega^2/(4\omega_L)$. It has quadratic dependence on the Rabi frequency Ω, which may be controlled by the intensity of the applied driving field. Therefore the Bloch-Siegert shift is an effect that starts playing a role under the action of strong fields. We also observe the term $3G/(8i\omega_L)\sum_{\boldsymbol{k}}(\boldsymbol{g_k}\cdot\boldsymbol{d})(a_{\boldsymbol{k}}^\dagger S^+ e^{i\omega_L t} - a_{\boldsymbol{k}} S^- e^{-i\omega_L t})$ that is proportional to G and is a direct consequence of the broken inversion symmetry of the system. It is responsible for the emission of the THz-photon. The energy of the field modes $\sum_{\boldsymbol{k}}\hbar(\omega_{\boldsymbol{k}} - \omega_L)a_{\boldsymbol{k}}^\dagger a_{\boldsymbol{k}}$, the interaction of the driving field with the transition dipole moment $-\hbar\Omega/2(S^+ + S^-)$ and the energy conserving interactions of the environment with the two-level system $i\sum_{\boldsymbol{k}}(\boldsymbol{g_k}\cdot\boldsymbol{d})(a_{\boldsymbol{k}}^\dagger S^- - a_{\boldsymbol{k}} S^+)$ remain unchanged.

III. Master equation approach

In this chapter we will mainly be concerned in deriving the equations of motion of operators $Q(t)$ belonging to the atomic subsystem only. Up to this point we have set up a Hamiltonian describing the physical phenomena of the considered system and performed several manipulations in order to simplify the derivation of the equations of motion: at first we applied a unitary transformation on the Hamiltonian and after that made use of perturbation theory getting the final form in Eq. (II.28). In order to derive the equations of motion for the atomic operators we work in the Heisenberg picture and use the Heisenberg equation of motion, which may be found in many standard textbooks such as [49],

$$\frac{d}{dt}\langle Q(t)\rangle = \frac{i}{\hbar}\langle [H,Q]\rangle\,, \tag{III.1}$$

where the angle brackets denote the expectation value. By inserting the Hamiltonian of Eq. (II.28) and evaluating the commutators, we readily have an equation of motion for the operators we want to calculate,

$$\begin{aligned}
\frac{d}{dt}\langle Q(t)\rangle &= \frac{i}{\hbar}\langle [H,Q]\rangle \\
&= \frac{i}{\hbar}\langle [H_0,Q]\rangle - \sum_{\boldsymbol{k}} \frac{(\boldsymbol{g_k}\cdot\boldsymbol{d})}{\hbar}\langle [(a^\dagger_{\boldsymbol{k}}S^- - a_{\boldsymbol{k}}S^+),Q]\rangle \\
&\quad + \frac{3G}{8\hbar\omega_L}\sum_{\boldsymbol{k}}(\boldsymbol{g_k}\cdot\boldsymbol{d})\langle [(a^\dagger_{\boldsymbol{k}}S^+e^{i\omega_L t} - a_{\boldsymbol{k}}S^-e^{-i\omega_L t}),Q]\rangle \\
&\quad - \frac{\Omega}{2\hbar\omega_L}\sum_{\boldsymbol{k}}(\boldsymbol{g_k}\cdot\boldsymbol{d})\langle (a_{\boldsymbol{k}} - a^\dagger_{\boldsymbol{k}})[S_z,Q]\rangle \\
&= \frac{i}{\hbar}\langle [H_0,Q]\rangle - \sum_{\boldsymbol{k}} \frac{(\boldsymbol{g_k}\cdot\boldsymbol{d})}{\hbar}\Big(\langle a^\dagger_{\boldsymbol{k}}[S^-,Q]\rangle - \langle [S^+,Q]a_{\boldsymbol{k}}\rangle\Big) \\
&\quad + \frac{3G}{8\hbar\omega_L}\sum_{\boldsymbol{k}}(\boldsymbol{g_k}\cdot\boldsymbol{d})\Big(\langle a^\dagger_{\boldsymbol{k}}[S^+,Q]\rangle e^{i\omega_L t} + \langle [Q,S^-]a_{\boldsymbol{k}}\rangle e^{-i\omega_L t}\Big) \\
&\quad + \frac{\Omega}{2\hbar\omega_L}\sum_{\boldsymbol{k}}(\boldsymbol{g_k}\cdot\boldsymbol{d})\Big(\langle a^\dagger_{\boldsymbol{k}}[S_z,Q]\rangle + \langle [Q,S_z]a_{\boldsymbol{k}}\rangle\Big).
\end{aligned} \tag{III.2}$$

Since the operator $Q(t)$ belongs to the atomic subsystem only, the commutators $[Q, a_{\boldsymbol{k}}]$ and $[Q, a_{\boldsymbol{k}}^\dagger]$ vanish. We assume that the coupling between light and matter is weak, so that we can eliminate the electromagnetic field operators in the Born-Markov approximation that is further explained in App. A.4. The field operators will be expressed as a function of atomic operators. Again we resort to Heisenberg's equation of motion and have to remind ourselves of the fundamental commutation relation for the creation and annihilation operators $[a_{\boldsymbol{k}}, a_{\boldsymbol{k}'}^\dagger] = \delta_{\boldsymbol{k}\boldsymbol{k}'}$,

$$\begin{aligned}
\frac{d}{dt}a_{\boldsymbol{k}}(t) &= \frac{i}{\hbar}[H, a_{\boldsymbol{k}}] \\
&= i\sum_{\boldsymbol{k}}(\omega_{\boldsymbol{k}} - \omega_L)[a_{\boldsymbol{k}}^\dagger a_{\boldsymbol{k}}, a_{\boldsymbol{k}}] - \sum_{\boldsymbol{k}} \frac{(\boldsymbol{g}_{\boldsymbol{k}} \cdot \boldsymbol{d})}{\hbar}[(a_{\boldsymbol{k}}^\dagger S^- - a_{\boldsymbol{k}} S^+), a_{\boldsymbol{k}}] \\
&\quad + \frac{3G}{8\hbar\omega_L}\sum_{\boldsymbol{k}}(\boldsymbol{g}_{\boldsymbol{k}} \cdot \boldsymbol{d})([a_{\boldsymbol{k}}^\dagger S^+, a_{\boldsymbol{k}}]e^{i\omega_L t} - [a_{\boldsymbol{k}} S^-, a_{\boldsymbol{k}}]e^{-i\omega_L t}) \\
&\quad - \frac{\Omega}{2\hbar\omega_L}\sum_{\boldsymbol{k}}(\boldsymbol{g}_{\boldsymbol{k}} \cdot \boldsymbol{d})[(a_{\boldsymbol{k}} - a_{\boldsymbol{k}}^\dagger), a_{\boldsymbol{k}}]S_z \\
&= -i(\omega_{\boldsymbol{k}} - \omega_L)a_{\boldsymbol{k}} + \frac{(\boldsymbol{g}_{\boldsymbol{k}} \cdot \boldsymbol{d})}{\hbar}S^- - \frac{3G}{8\hbar\omega_L}(\boldsymbol{g}_{\boldsymbol{k}} \cdot \boldsymbol{d})S^+ e^{i\omega_L t} \\
&\quad - \frac{\Omega}{2\hbar\omega_L}(\boldsymbol{g}_{\boldsymbol{k}} \cdot \boldsymbol{d})S_z.
\end{aligned} \tag{III.3}$$

The same calculation can be explicitly performed for the creation operator $a_{\boldsymbol{k}}^\dagger$, or one just notices that the equations of $a_{\boldsymbol{k}}$ and $a_{\boldsymbol{k}}^\dagger$ are hermitian conjugate:

$$\begin{aligned}
\frac{d}{dt}a_{\boldsymbol{k}}^\dagger(t) &= \frac{i}{\hbar}[H, a_{\boldsymbol{k}}^\dagger] \\
&= i(\omega_{\boldsymbol{k}} - \omega_L)a_{\boldsymbol{k}}^\dagger + \frac{(\boldsymbol{g}_{\boldsymbol{k}} \cdot \boldsymbol{d})}{\hbar}S^+ - \frac{3G}{8\hbar\omega_L}(\boldsymbol{g}_{\boldsymbol{k}} \cdot \boldsymbol{d})S^- e^{-i\omega_L t} \\
&\quad - \frac{\Omega}{2\hbar\omega_L}(\boldsymbol{g}_{\boldsymbol{k}} \cdot \boldsymbol{d})S_z.
\end{aligned} \tag{III.4}$$

The equations of motion correspond to simple linear inhomogeneous differential equations of first order which can be solved with basic methods. A general solution is derived in App. A.3. The result reads

$$
\begin{aligned}
a_{\boldsymbol{k}}^{\dagger}(t) = a_{\boldsymbol{k}}^{\dagger}(0)e^{i(\omega_{\boldsymbol{k}}-\omega_L)t} + \frac{(\boldsymbol{g}_{\boldsymbol{k}} \cdot \boldsymbol{d})}{\hbar} \int_0^t dt' S^+(t') e^{i(\omega_{\boldsymbol{k}}-\omega_L)(t-t')} \\
- \frac{3G}{8\hbar\omega_L}(\boldsymbol{g}_{\boldsymbol{k}} \cdot \boldsymbol{d}) \int_0^t dt' e^{i(\omega_{\boldsymbol{k}}-\omega_L)(t-t')} S^-(t') e^{-i\omega_L t'} \\
- \frac{\Omega}{2\hbar\omega_L}(\boldsymbol{g}_{\boldsymbol{k}} \cdot \boldsymbol{d}) \int_0^t dt' e^{i(\omega_{\boldsymbol{k}}-\omega_L)(t-t')} S_z(t').
\end{aligned}
\tag{III.5}
$$

The time integrals contain the product of time-dependent atomic operators and exponential functions. In order to perform the integration one has to somehow pull the operators out of the integral. This can be achieved by making use of the Born-Markov approximation explained in App. A.4, where we get

$$
S^+(t') = S^+(t) e^{-i(\Delta + \frac{\Omega^2}{4\omega_L})(t-t')}, \tag{III.6}
$$

$$
S^-(t') = S^-(t) e^{i(\Delta + \frac{\Omega^2}{4\omega_L})(t-t')}, \tag{III.7}
$$

$$
S_z(t') = S_z(t). \tag{III.8}
$$

The three integrals in Eq. (III.5) will be called K_1, K_2 and K_3 and can be performed separately:

$$
\begin{aligned}
K_1 &= \lim_{t\to\infty} \int_0^t dt' e^{i(\omega_{\boldsymbol{k}}-\omega_L-\Delta-\frac{\Omega^2}{4\omega_L})(t-t')} \\
&= \lim_{t\to\infty} \int_0^t dt' e^{i(\omega_{\boldsymbol{k}}-\omega_0-\frac{\Omega^2}{4\omega_L})(t-t')} \\
&= \pi\delta(\omega_{\boldsymbol{k}} - \omega_0 - \frac{\Omega^2}{4\omega_L}) + iP\frac{1}{\omega_{\boldsymbol{k}} - \omega_0 - \frac{\Omega^2}{4\omega_L}},
\end{aligned}
\tag{III.9}
$$

where we have set the limit of the time integration to infinity. This approximation is justified, since for big values of time the integrand becomes fast oscillating and thus negligible. We also used the identity

$$
\int_0^\infty d\tau e^{\pm i\epsilon\tau} = \pi\delta(\epsilon) \pm iP\frac{1}{\epsilon}, \tag{III.10}
$$

where $\delta(\epsilon)$ is Dirac's delta function and P is Cauchy's principal value. They are defined as

$$\delta(x-u) = \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{ik(x-u)}dk \tag{III.11}$$

and

$$P\int_{-a}^{b}\frac{f(\omega)}{\omega} = \lim_{\epsilon\to 0}\left(\int_{-a}^{-\epsilon}\frac{f(\omega)}{\omega}d\omega + \int_{\epsilon}^{b}\frac{f(\omega)}{\omega}d\omega\right). \tag{III.12}$$

A more detailed discussion may be found in [51]. Now we continue to calculate the second and third integral using the same method

$$\begin{aligned}
K_2 &= \lim_{t\to\infty}\int_0^t dt' e^{i(\omega_{\boldsymbol{k}}-\omega_L+\omega_0-\omega_L+\frac{\Omega^2}{4\omega_L})(t-t')}e^{-i\omega_L t'} \\
&= \lim_{t\to\infty}\int_0^t dt' e^{i(\omega_{\boldsymbol{k}}-2\omega_L+\omega_0+\frac{\Omega^2}{4\omega_L})(t-t')}e^{-i\omega_L t'} \\
&= \lim_{\tau\to\infty}\int_0^\tau d\tau e^{i(\omega_{\boldsymbol{k}}-\omega_L+\omega_0+\frac{\Omega^2}{4\omega_L})\tau}e^{-i\omega_L t} \\
&= \pi\delta(\omega_{\boldsymbol{k}}+\omega_0-\omega_L+\frac{\Omega^2}{4\omega_L})e^{-i\omega_L t} \\
&\quad + \frac{iP}{\omega_{\boldsymbol{k}}+\omega_0-\omega_L+\frac{\Omega^2}{4\omega_L}}e^{-i\omega_L t},
\end{aligned} \tag{III.13}$$

$$\begin{aligned}
K_3 &= \lim_{t\to\infty}\int_0^t dt' e^{i(\omega_{\boldsymbol{k}}-\omega_L)(t-t')} \\
&= \pi\delta(\omega_{\boldsymbol{k}}-\omega_L) + \frac{iP}{\omega_{\boldsymbol{k}}-\omega_L}.
\end{aligned} \tag{III.14}$$

We see that all three results contain terms proportional to Dirac's delta function and Cauchy's principal value. In the following calculations the latter is going to be neglected. As very well explained in [46], the term proportional to Cauchy's principal value leads to a very small shift in the energy of the two-level system, contributing to the Lamb shift which we do not consider.

Having now calculated the integrals, the creation operator $a_{\boldsymbol{k}}^\dagger$ acquires the form

$$
\begin{aligned}
a^\dagger_{\boldsymbol{k}}(t) = a^\dagger_{\boldsymbol{k}}(0)e^{i(\omega_{\boldsymbol{k}}-\omega_L)t} &+ \pi\frac{(\boldsymbol{g_k}\cdot\boldsymbol{d})}{\hbar}S^+(t)\delta\left(\omega_{\boldsymbol{k}}-\omega_0-\frac{\Omega^2}{4\omega_L}\right)\\
&- \frac{3G}{8\hbar\omega_L}\pi(\boldsymbol{g_k}\cdot\boldsymbol{d})S^-(t)\delta\left(\omega_{\boldsymbol{k}}+\omega_0-\omega_L+\frac{\Omega^2}{4\omega_L}\right)e^{-i\omega_L t}\\
&- \frac{\Omega}{2\hbar\omega_L}\pi(\boldsymbol{g_k}\cdot\boldsymbol{d})S_z(t)\delta(\omega_{\boldsymbol{k}}-\omega_L).
\end{aligned}
\tag{III.15}
$$

The same may also be done for the annihilation operator $a_{\boldsymbol{k}}$,

$$
\begin{aligned}
a_{\boldsymbol{k}}(t) = a_{\boldsymbol{k}}(0)e^{-i(\omega_{\boldsymbol{k}}-\omega_L)t} &+ \pi\frac{(\boldsymbol{g_k}\cdot\boldsymbol{d})}{\hbar}S^-(t)\delta\left(\omega_{\boldsymbol{k}}-\omega_0-\frac{\Omega^2}{4\omega_L}\right)\\
&- \frac{3G}{8\hbar\omega_L}\pi(\boldsymbol{g_k}\cdot\boldsymbol{d})S^+(t)\delta\left(\omega_{\boldsymbol{k}}+\omega_0-\omega_L+\frac{\Omega^2}{4\omega_L}\right)e^{i\omega_L t}\\
&- \frac{\Omega}{2\hbar\omega_L}\pi(\boldsymbol{g_k}\cdot\boldsymbol{d})S_z(t)\delta(\omega_{\boldsymbol{k}}-\omega_L).
\end{aligned}
\tag{III.16}
$$

Next we define the different decay rates of spontaneous emission γ_R, γ_L and γ_T,

$$\gamma_R = \gamma(\omega_0+\frac{\Omega^2}{4\omega_L}) = \pi\sum_{\boldsymbol{k}}\frac{(\boldsymbol{g_k}\cdot\boldsymbol{d})^2}{\hbar^2}\delta\left(\omega_{\boldsymbol{k}}-\omega_0-\frac{\Omega^2}{4\omega_L}\right) \tag{III.17a}$$

$$\gamma_L = \gamma(\omega_L) = \pi\sum_{\boldsymbol{k}}\frac{(\boldsymbol{g_k}\cdot\boldsymbol{d})^2}{\hbar^2}\delta(\omega_{\boldsymbol{k}}-\omega_L) \tag{III.17b}$$

$$\gamma_T = \gamma(\omega_L-\omega_0-\frac{\Omega^2}{4\omega_L}) = \pi\sum_{\boldsymbol{k}}\frac{(\boldsymbol{g_k}\cdot\boldsymbol{d})^2}{\hbar^2}\delta\left(\omega_{\boldsymbol{k}}-\omega_L+\omega_0+\frac{\Omega^2}{4\omega_L}\right), \tag{III.17c}$$

where we assume that $\gamma_T \neq 0$ only for $\omega_L > \omega_0 + \Omega^2/(4\omega_L)$. This means that without the detuning of the driving laser, the decay rate γ_T vanishes. The subscripts indicate the frequency of the emission; "R" stands for resonance, "L" stands for laser and "T" stands for THz. The three decay rates differ in the argument of the respective delta functions. In each equation we have a sum over all the modes of the electromagnetic field. Since we have different delta functions, the spontaneous emission into the mode of the radiation field will be of different frequency. $\gamma(\omega_0 + \Omega^2/(4\omega_L))$ for example is the decay rate of spontaneous emission of frequency $\omega_0 + \Omega^2/(4\omega_L)$. In App. B.2, the calculation of the decay rates is carried out explicitly.

After having calculated explicit forms of the creation and annihilation operators in Eqs. (III.15), (III.16) we are now in a position to insert those expressions in Eq. (III.2), getting a differential equation that depends only on the atomic operators $\langle S_z\rangle, \langle S^+\rangle, \langle S^-\rangle$,

$$\begin{aligned}
&\frac{d}{dt}\langle Q(t)\rangle - \frac{i}{\hbar}\langle [H_0, Q]\rangle \\
&= \langle \left(-\gamma_R S^+ + \frac{3G}{8\hbar\omega_L}\gamma_T S^- e^{-i\omega_L t} + \frac{\Omega}{2\omega_L}\gamma_L S_z \right) [S^-, Q]\rangle \\
&\quad + \frac{3G}{8\omega_L}\langle \left(\gamma_R S^+ - \frac{3G}{8\hbar\omega_L}\gamma_T S^- e^{-i\omega_L t} - \frac{\Omega}{2\omega_L}\gamma_L S_z \right) [S^+, Q]\rangle e^{i\omega_L t} \\
&\quad + \frac{\Omega}{2\omega_L}\langle \left(\gamma_R S^+ - \frac{3G}{8\hbar\omega_L}\gamma_T S^- e^{-i\omega_L t} - \frac{\Omega}{2\omega_L}\gamma_L S_z \right) [S_z, Q]\rangle + h.c..
\end{aligned} \tag{III.18}$$

Since the contributions of fast oscillating terms are very small, we neglect them, being left with

$$\begin{aligned}
&\frac{d}{dt}\langle Q(t)\rangle - \frac{i}{\hbar}\langle [H_0, Q]\rangle \\
&= -\gamma_R(\langle S^+[S^-, Q]\rangle + \langle [Q, S^+]S^-\rangle) \\
&\quad + \frac{\Omega}{2\omega_L}\gamma_L(\langle S_z[S^-, Q]\rangle + \langle [Q, S^+]S_z\rangle) \\
&\quad - (\frac{3G}{8\omega_L})^2\gamma_T(\langle S^-[S^+, Q]\rangle + \langle [Q, S^-]S^+\rangle) \\
&\quad + \frac{\Omega}{2\omega_L}\gamma_R(\langle S^+[S_z, Q]\rangle + \langle [Q, S_z]S^-\rangle) \\
&\quad - (\frac{\Omega}{2\omega_L})^2\gamma_L(\langle S_z[S_z, Q]\rangle + \langle [Q, S_z]S_z\rangle).
\end{aligned} \tag{III.19}$$

Now we have the final equation of motion after having done all the necessary approximations. $Q(t)$ may now be replaced by our needed operators:

$$\begin{aligned}
\frac{d}{dt}\langle S_z(t)\rangle &= i\frac{\Omega}{2}(\langle S^+\rangle - \langle S^-\rangle) - 2\gamma_R(\frac{1}{2} + \langle S_z\rangle) \\
&\quad - \frac{\Omega}{4\omega_L}\gamma_L(\langle S^-\rangle + \langle S^+\rangle) + 2(\frac{3G}{8\omega_L})^2\gamma_T(\frac{1}{2} - \langle S_z\rangle).
\end{aligned} \tag{III.20}$$

$$\begin{aligned}
\frac{d}{dt}\langle S^+(t)\rangle &= (i\Delta + i\frac{\Omega^2}{4\omega_L})\langle S^+\rangle + i\Omega\langle S_z\rangle - \gamma_R\langle S^+\rangle - \frac{\Omega}{4\omega_L}\gamma_L \\
&\quad - (\frac{3G}{8\omega_L})^2\gamma_T\langle S^+\rangle - \frac{\Omega}{2\omega_L}\gamma_R(\frac{1}{2} + \langle S_z\rangle) - (\frac{\Omega}{2\omega_L})^2\gamma_L\langle S^+\rangle .
\end{aligned} \tag{III.21}$$

$$\frac{d}{dt}\langle S^-(t)\rangle = -(i\Delta + i\frac{\Omega^2}{4\omega_L})\langle S^-\rangle - i\Omega\langle S_z\rangle - \gamma_R\langle S^-\rangle - \frac{\Omega}{4\omega_L}\gamma_L - (\frac{3G}{8\omega_L})^2\gamma_T\langle S^-\rangle - \frac{\Omega}{2\omega_L}\gamma_R(\frac{1}{2} + \langle S_z\rangle) - (\frac{\Omega}{2\omega_L})^2\gamma_L\langle S^-\rangle \,. \quad \text{(III.22)}$$

The three differential equations above represent a set of coupled linear differential equations of first order. The three unknown time-dependent parameters are the atomic operators $\langle S_z\rangle$, $\langle S^+\rangle$ and $\langle S^-\rangle$. All physical interpretation may be derived from those equations. In the next Chapter we choose a numerical approach to solve and interpret them.

IV. Physical interpretation

IV.1. Interpreting the master equation

Our model consists of a two-level system which describes two energy levels of a molecule. In our model we do not only define the transition dipole moment, but also an intrinsic dipole moment for each level with broken inversion symmetry, characteristic for molecules, for example. This means that $|\wp_{11}| \neq |\wp_{22}|$. This two-level system is driven by a slightly off-resonant classical laser. We derived a master equation III.19 for the dynamics of the operators, which will be discussed in the following section, giving us insight into the physical processes that play an important role. Our numerical calculations will be performed with the parameters of gamma globulin macromolecules. We start by writing down again the master equation:

$$\begin{aligned}
&\frac{d}{dt}\langle Q(t)\rangle - \frac{i}{\hbar}\langle [H_0, Q]\rangle \\
&= -\gamma_R(\langle S^+[S^-, Q]\rangle + \langle [Q, S^+]S^-\rangle) \\
&\quad + \frac{\Omega}{2\omega_L}\gamma_L(\langle S_z[S^-, Q]\rangle + \langle [Q, S^+]S_z\rangle) \\
&\quad - (\frac{3G}{8\omega_L})^2\gamma_T(\langle S^-[S^+, Q]\rangle + \langle [Q, S^-]S^+\rangle) \\
&\quad + \frac{\Omega}{2\omega_L}\gamma_R(\langle S^+[S_z, Q]\rangle + \langle [Q, S_z]S^-\rangle) \\
&\quad - (\frac{\Omega}{2\omega_L})^2\gamma_L(\langle S_z[S_z, Q]\rangle + \langle [Q, S_z]S_z\rangle).
\end{aligned} \tag{IV.1}$$

Every term can be assigned to a different physical process and one should remind oneself of the definitions of the decay rates in Eqs. (III.17).

The **first summand** proportional to $\gamma_R = \gamma(\omega_0 + \Omega^2/(4\omega_L))$ can be interpreted as the pure spontaneous emission of a photon of frequency $\omega_0 + \Omega^2/(4\omega_L)$ from the second energy level to the first energy level, taking into consideration the Bloch-Siegert shift $\Omega^2/(4\omega_L)$. The process is illustrated in Fig. IV.1. With the parameters we will use later on, the photon will be in the optical frequency range.

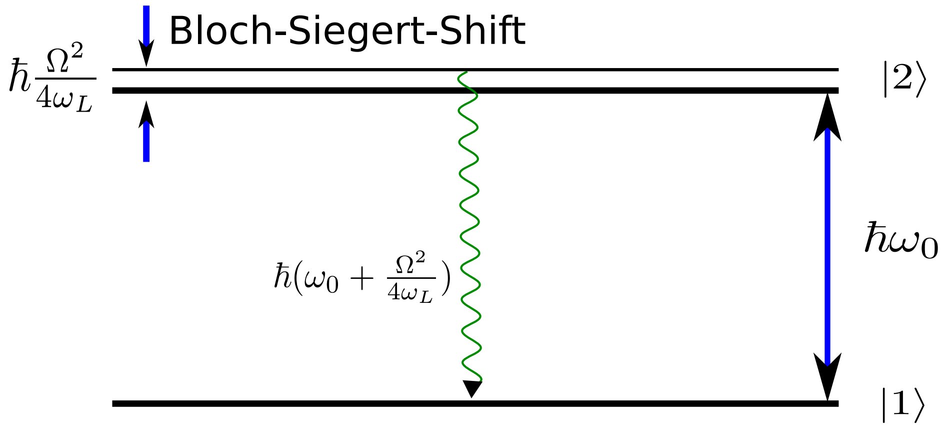

Figure IV.1.: The excited two-level system spontaneously emits a photon of frequency $\omega_0 + \Omega^2/(4\omega_L)$.

The **second summand** proportional to $\gamma_L = \gamma(\omega_L)$ can be assigned to a process of induced spontaneous emission. The driving field with the frequency ω_L shines on the two-level-system that immediately emits a photon of the same frequency. We are dealing with an effect that is taken into account due to perturbation theory. The process is illustrated in Fig. IV.2.

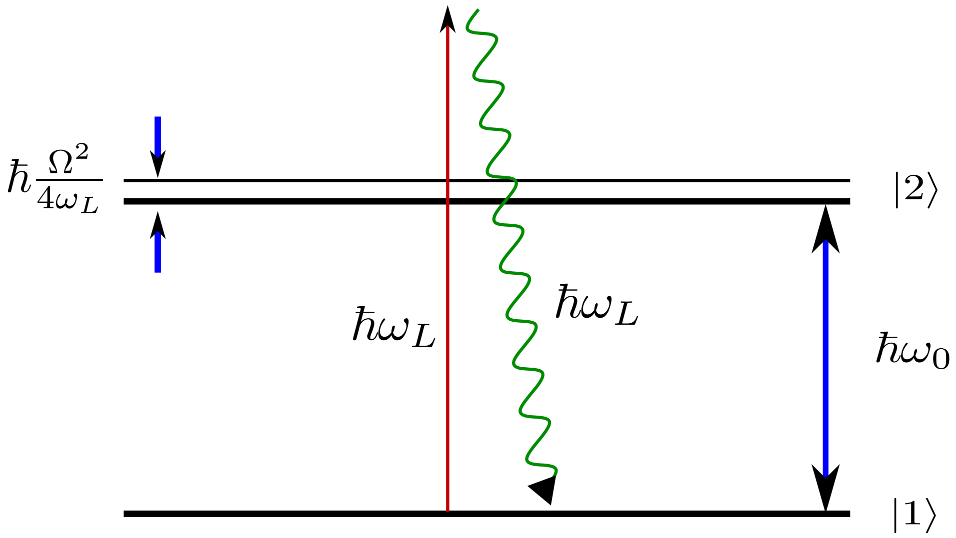

Figure IV.2.: The off-resonant laser of frequency ω_L excites the two-level system and induces the spontaneous emission of a photon of frequency ω_L, falling back into the ground state.

The **third summand** proportional to $\gamma_T = \gamma(\omega_L - \omega_0 - \Omega^2/(4\omega_L))$ can also be assigned to a process of induced spontaneous emission. The off-resonant laser shines on the two-level system followed by the emission of a photon of frequency $\omega_L - \omega_0 - \Omega^2/(4\omega_L)$. This is exactly the frequency with which the laser is detuned from the resonance taking into account the Bloch-Siegert shift. The process is illustrated in Fig. IV.3. We note that the frequency of the photon is determined by the detuning of the laser. Later on, we will choose the detuning such that the photon has THz-frequency.

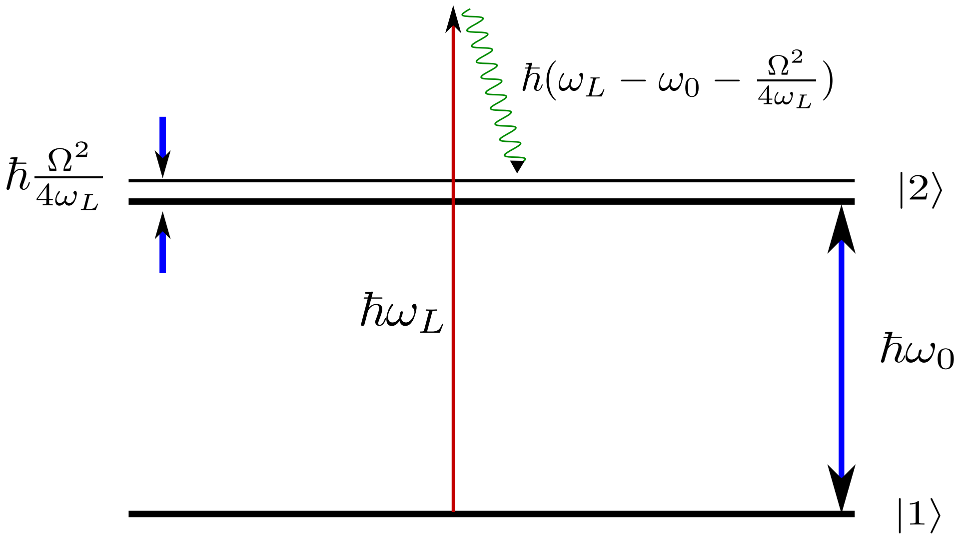

Figure IV.3.: The off-resonant laser of frequency ω_L excites the two-level system and induces the spontaneous emission of a photon of frequency $\omega_L - \omega_0 - \Omega^2/(4\omega_L)$. The system remains excited.

The **fourth summand** proportional to $\gamma_R = \gamma(\omega_0 + \Omega^2/(4\omega_L))$ represents a non-linear process in which the laser shines on the two-level system and induces the emission of a photon of frequency $\omega_0+\Omega^2/(4\omega_L)$. This effect is shown in Fig. IV.4.

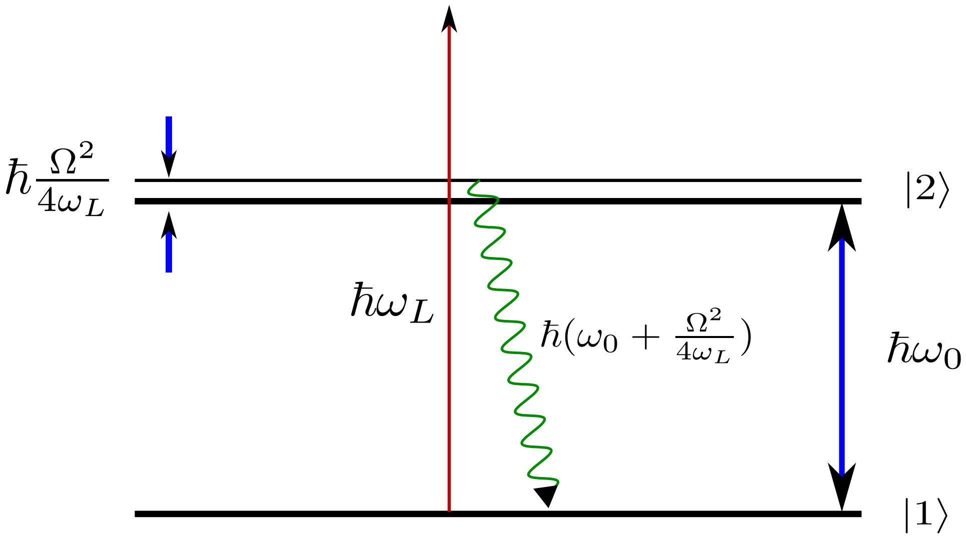

Figure IV.4.: The off-resonant laser of frequency ω_L excites the two-level system that spontaneously emits a photon of frequency $\omega_0+\Omega^2/(4\omega_L)$, falling back into the ground state.

The fifth and **last summand** proportional to $\gamma_L = \gamma(\omega_L)$ describes the dephasing induced by the strong driving field.

For our purposes, the most important terms are those of the first and third summand that are responsible for the emission of the non-classical photon pair and that are going to be further studied in the next parts.

IV.2. Physical realization

Up to this point we have successfully described and partially interpreted our theoretical system by setting up a Hamiltonian and modifying it in such a way that we could use a master equation approach to derive the equations of motion of the atomic operators. Now we are in a position to use real parameters of real physical systems to make predictions of physical phenomena. Two examples of quantum systems with broken inversion symmetry suited for our purposes are gamma globulin macromolecules and quantum dots treated in the following two subsections. Since their parameters are very similar to each other, we plot and discuss the results for gamma globulin only. Similar systems with broken inversion symmetry have recently been investigated in research [39].

IV.2.1. Gamma globulin

Our model may be applied to gamma globulin macromolecules which play a crucial role in the immune system. They may be modeled as two-level systems, which have a transition dipole moment between the two energy levels, as well as intrinsic dipole moments regarding each energy level. The parameters of gamma globulin [52] molecules read $|\omega_2 - \omega_1| \cong 4.8 \times 10^{15}\mathrm{s}^{-1}$, $|\wp_{21}| \cong 1\mathrm{D}$ and $|\wp_{22} - \wp_{11}| \cong 100\mathrm{D}$ (D stands for the unit Debye). Now if we choose the laser frequency to be off-resonant by the detuning $\Delta = 10^{13}\mathrm{s}^{-1}$ with respect to the transition frequency, the emitted photon corresponding to the process in Fig. IV.3 will have THz-frequency.

IV.2.2. Quantum dots

Quantum dots (QD) are 0-dimensional quantum systems having an electron confined in all three space dimensions. They are also known as artifical atoms, reflecting the fact that the energy states are quantized [53]. As in common atoms a driving electromagnetic field can excite a QD, with the difference that here an electron is lifted from the valence band into the conduction band, well-known concepts from solid state physics. The bandgap corresponds approximately to the transition frequency $\hbar\omega_0$ of the QD. Possible realizations of such semiconductor nanostructures are nitride based devices, such as gallium nitride (GaN) for example, exhibiting a hexagonal crystal structure. As a consequence of their non-centrosymmetric form, nitride QDs show a unique feature, namely a very pronounced piezoelectric effect producing static electric fields with strengths of several MV/cm [54–56]. This field separates the conduction-band electrons from the valence-band holes spatially und thus creates a dipole moment of magnitude $|\wp_{22} - \wp_{11}| \cong el$, where l is the height of the QD [57, 58]. A typical QD has a height of several nanometers, for which we can estimate the dipole moment $|\wp_{22} - \wp_{11}| \cong 10\mathrm{D}$. The transition dipole moment may as well be estimated as $\wp_{12} \cong 10\mathrm{D}$ and the bandgap is of about $E_g = 3.24\,\mathrm{eV}$ which corresponds to a transition frequency of $\omega_0 = 4.92 \times 10^{15}\mathrm{s}^{-1}$. For further details see [59].

IV.3. Intensity

In the following section we will calculate the intensity of the specific process that is illustrated in Fig. IV.3, namely the spontaneous emission of a photon with THz-frequency. All the calculations have been performed numerically. The average light intensity at point $\boldsymbol{r}$ at time t is defined as

$$\langle I(\boldsymbol{r},t)\rangle = \langle E^{(-)}(\boldsymbol{r},t)E^{(+)}(\boldsymbol{r},t)\rangle\,. \tag{IV.2}$$

As shown in App. B.4, this can be calculated explicitly in terms of atomic operators

$$\langle I(\boldsymbol{r},t)\rangle \propto \langle S^{-}(\boldsymbol{r},t)S^{+}(\boldsymbol{r},t)\rangle = \frac{1}{2} - \langle S_z\rangle\,. \tag{IV.3}$$

For that purpose the coupled differential equations (III.20, III.21, III.22) need to be solved numerically. The initial conditions for the time-dependent calculations will always be $\langle S_z(0)\rangle = -1/2$ and $\langle S^{\pm}(0)\rangle = 0$, meaning that the molecule is initially in its ground state. At first we calculate the time-dependent solutions with a specific Rabi-frequency of $\Omega = 10^{12}s^{-1}$ and detuning $\Delta = 10^{12}s^{-1}$. All the other parameters may be calculated by inserting the data of the gamma globulin macromolecules. Our result is plotted in Fig. IV.5. We recognize the behavior of a driven and damped two-level system, which at a certain time reaches a steady-state.

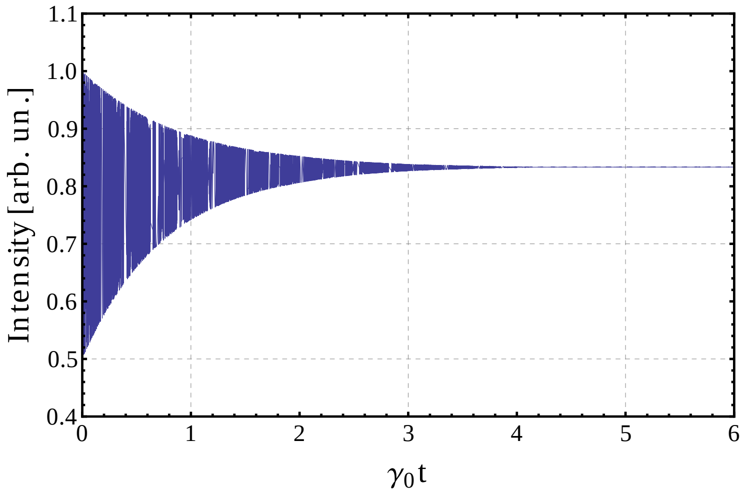

Figure IV.5.: The intensity $\frac{1}{2} - \langle S_z \rangle$ of the THz-emission as a function of time scaled by the decay rate γ_0. We used the following parameters: Rabi-frequency of $\Omega = 10^{12}\,\mathrm{s}^{-1}$ and detuning of $\Delta = 10^{12}\,\mathrm{s}^{-1}$, as well as the data of gamma globulin. One observes the behavior of a damped oscillator, which reaches a steady state as time progresses. We observe damped Rabi oscillations.

Next, we are interested in the behavior of the intensity as a function of the Rabi frequency $\Omega = \wp_{12} \cdot \boldsymbol{E}_0/\hbar$ which can be manipulated by adjusting the intensity of the driving laser field. This time we are not interested in the time evolution, but just in the steady states, which are plotted as a function of the Rabi frequency in Fig. IV.6. Technical details are shown in App. B.5.

At first we observe constant behavior of the intensity with growing Rabi frequency. At an order of magnitude of about $\Omega = 10^{12}\,\mathrm{s}^{-1}$, we observe that the curve starts dropping. To understand this behavior we have to take a look at the population $\langle S_z \rangle$ of the process as a function of the Rabi frequency in Fig. IV.7.

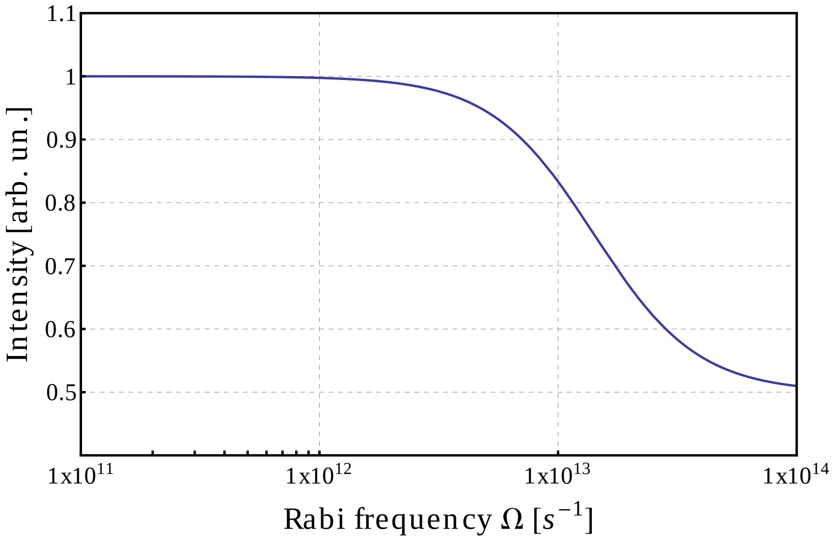

Figure IV.6.: The steady-state solution for the intensity of the THz-emission as a function of the Rabi frequency Ω on a logarithmic scale. Used parameters: Transition frequency $\omega_0 = 5.0 \times 10^{15}\,\mathrm{s}^{-1}$, laser frequency $\omega_L = \omega_0 + 10^{13}\,\mathrm{s}^{-1}$, detuning $\Delta = 10^{13}\,\mathrm{s}^{-1}$ and decay rate $\gamma_0 = 3 \times 10^6\,\mathrm{s}^{-1}$ with respect to ω_0.

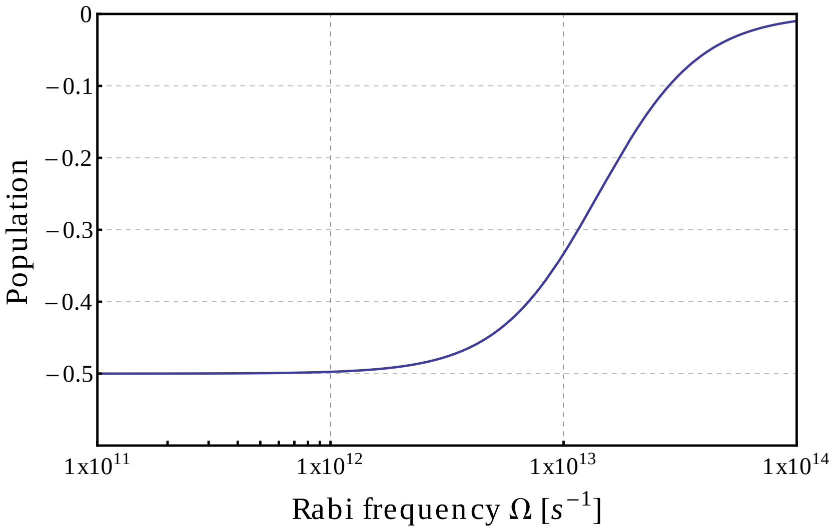

Figure IV.7.: The steady-state solution for the population $\langle S_z \rangle$ as a function of the Rabi frequency Ω on a logarithmic scale. Used parameters: Transition frequency $\omega_0 = 5.0 \times 10^{15}\mathrm{s}^{-1}$, laser frequency $\omega_L = \omega_0 + 10^{13}\,\mathrm{s}^{-1}$, detuning $\Delta = 10^{13}\,\mathrm{s}^{-1}$ and decay rate $\gamma_0 = 3 \times 10^6\,\mathrm{s}^{-1}$ with respect to ω_0.

The population $\langle S_z \rangle$ always takes values between $-0.5 \leqslant \langle S_z \rangle \leqslant 0.5$, which can be seen if one takes a look at the definition of the operator (see Eqs. (A.1)). For one single molecule, a value of -0.5 means that the two-level system is in its ground state. This is exactly the case for low Rabi frequencies. At increasing Rabi frequencies, the probability to find the molecule in the excited state increases. This means that with increasing population in the excited state the intensity of the THz-photon emission of Fig. IV.6 decreases. This is no surprise, since the emission process as illustrated in Fig. IV.5 can only be induced, if the molecule is in its ground state.

In order to assure the validity of our approximations, we have to make sure that $\Omega/\omega_L \ll 1$ as required by our perturbative calculation. Further, we have to keep in mind that the magnitude of the Bloch-Siegert shift $\Omega^2/4\omega_L$ should not exceed that of the difference between the laser frequency ω_L and the two-level system frequency ω_0: $\Omega^2/4\omega_L < \omega_L - \omega_0$. For $\omega_L = 5 \times 10^{15}\text{s}^{-1} - 10^{13}\text{s}^{-1}$, this means that the Rabi frequency should not exceed $\Omega = 10^{14}\text{s}^{-1}$.

IV.4. Non-classical effects in the statistical properties of light

In this section, we follow the theoretical treatment of the excellent review by Rodney Loudon [60] with the same title. When speaking of the classical theory of light, one usually refers to Maxwell's theory of electromagnetism which interprets light as electromagnetic waves. To each space-time point a well-defined vector quantity (the electric field) can be assigned. The quantum theory of light, however, looks quite different. The electric field vector is substituted by an operator which obeys certain commutation relations, and the underlying theory is quantum electrodynamics. The peculiar measurement process of quantum mechanics, which changes the state of the measured system, lies at the heart of the difference of both theories. This difference becomes more pronounced at very low intensities and can be verified by measuring the degree of second-order coherence. In the following we will calculate second order cross correlations of the processes of Figs. IV.1 and IV.3 studied before and show the violation of a Cauchy-Schwarz inequality.

IV.4.1. Classical degree of second order coherence

The classical degree of second-order coherence is defined as

$$G_{12}^{(2)}(\tau) = \frac{\langle I_1(\tau) I_2(0) \rangle}{\langle I_1 \rangle \langle I_2 \rangle} \tag{IV.4}$$

and correlates the intensities I_1 and I_2 of two different beams at different positions with time delay τ. Further we assume that all light beams are stationary and have ergodic properties, so that ensemble averages and time averages are the same. The following set of inequalities is valid in the classical case:

$$G_{12}^{(2)}(\tau) \geq 0, \tag{IV.5a}$$

$$G_{11}^{(2)}(0) G_{22}^{(2)}(0) \geq [G_{12}^{(2)}(\tau)]^2. \tag{IV.5b}$$

The first inequality holds for any pair of positive real numbers and the second one is basically the Cauchy-Schwarz inequality, where τ is the time delay between the detections.

IV.4.2. Quantum degree of second-order coherence

The quantum degree of second-order coherence, for which the above inequalities are not valid anymore, is defined as

$$g_{ij}^{(2)}(\tau) = \frac{\langle E_i^{(-)}(t)E_j^{(-)}(t+\tau)E_j^{(+)}(t+\tau)E_i^{(+)}(t)\rangle}{\langle E_i^{(-)}(t)E_i^{(+)}(t)\rangle\langle E_j^{(-)}(t)E_j^{(+)}(t)\rangle}, \tag{IV.6}$$

and can be interpreted as a measure for the probability for detecting a photon of a given frequency at time t and another one of different frequency at time $t+\tau$. For a brief theoretical justification see App. B.3. The subscripts take the fact into consideration that we might be dealing with a double-beam cross correlation function that measures photons of different beams. The limitations of the classical theory can be proven if this second-order correlation function of a specific system violates the classical inequalities of Eqs. IV.5. In this section we will show that this is the case for one process we study.

We are specifically interested in the correlation between the processes illustrated in Figs. IV.1 and IV.3. At first we will calculate the correlation function $g_{12}^{(2)}(t)$ describing the probability of first detecting a photon from the process of Fig. IV.3 followed by a photon from the other process (a THz-photon followed by an optical photon) and afterwards the correlation function $g_{21}^{(2)}(t)$ of the inverse order of detection (an optical photon followed by a THz-photon). The second-order correlation function also gives us an insight into the statistics of the process. If it has an exact value of $g^{(2)} = 1$, we are dealing with coherent (or Poissonian) light. If the value is $g^{(2)} < 1$, the observed light follows sub-Poissonian statistics, and if $g^{(2)} > 1$, we have super-Poissonian statistics.

The second-order correlation function describing the emission of first a THz-photon and then an optical photon can be written in a more useful way as a function of atomic operators as shown in App. B.4 and further evaluated here. We finally obtain

$$\begin{aligned}
g_{12}^{(2)}(t) &= \frac{\langle S^-(t)S^+(t)S^-(t)S^+(t)\rangle}{\langle S^-(t)S^+(t)\rangle\langle S^+(t)S^-(t)\rangle} \\
&= \frac{\langle S^-(t)(\frac{1}{2}+S_z(t))S^+(t)\rangle}{\langle S^-(t)S^+(t)\rangle\langle S^+(t)S^-(t)\rangle} \\
&= \frac{\frac{1}{2}\langle S^-(t)S^+(t)\rangle + \frac{1}{2}\langle S^-(t)S^+(t)\rangle}{\langle S^-(t)S^+(t)\rangle\langle S^+(t)S^-(t)\rangle} \\
&= \frac{1}{\frac{1}{2}+\langle S_z(t)\rangle},
\end{aligned} \tag{IV.7}$$

where we have used the relations of Eqs. (A.6). The time-dependent solution for our gamma globulin molecules are shown in Fig. IV.8 and the steady-state solution as a function of the Rabi frequency Ω is shown in Figs. IV.9 and IV.10, where the plots have different frequency ranges.

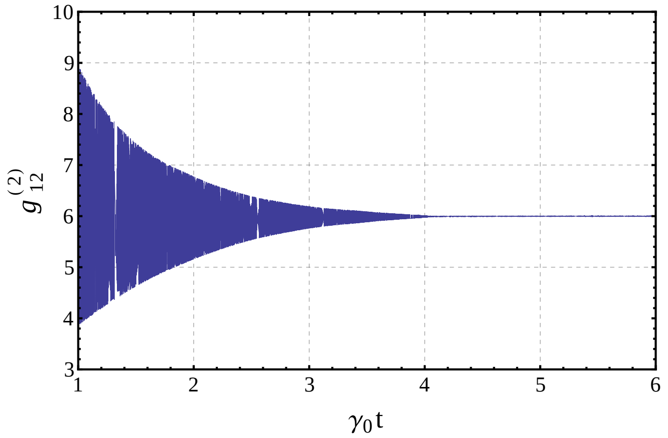

Figure IV.8.: The second-order intensity-intensity correlation function $g_{12}^{(2)}$ describing the probability of first the THz-emission followed by an optical emission as a function of time scaled by the decay rate. The used parameters are those of the gamma globulin molecule and we choose a detuning of $\Delta = 10^{12}\,s^{-1}$ and a Rabi frequency of $\Omega = 10^{12}\,\mathrm{s}^{-1}$.

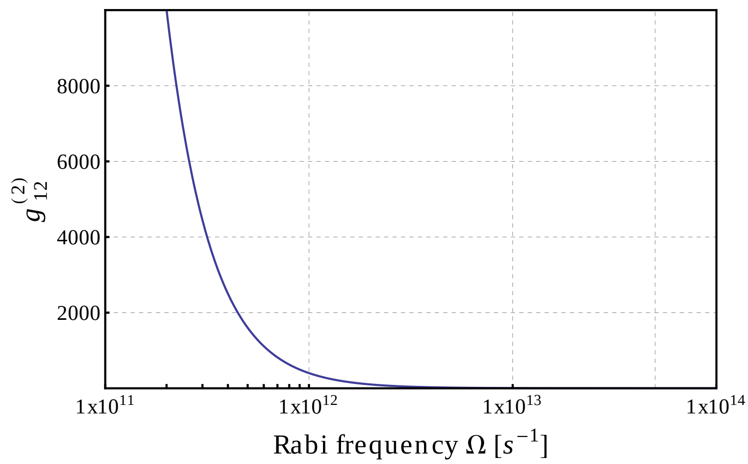

Figure IV.9.: The steady-state solution for the second-order intensity-intensity correlation function $g_{12}^{(2)}$ describing the probability of first the THz-emission followed by an optical emission as a function of the Rabi frequency Ω on a logarithmic scale. Used parameters: Transition frequency $\omega_0 = 5.0 \times 10^{15}\,\mathrm{s}^{-1}$, laser frequency $\omega_L = \omega_0 + 10^{13}\,\mathrm{s}^{-1}$, detuning $\Delta = 10^{13}\,\mathrm{s}^{-1}$ and decay rate $\gamma_0 = 3 \times 10^6\,\mathrm{s}^{-1}$ with respect to ω_0.

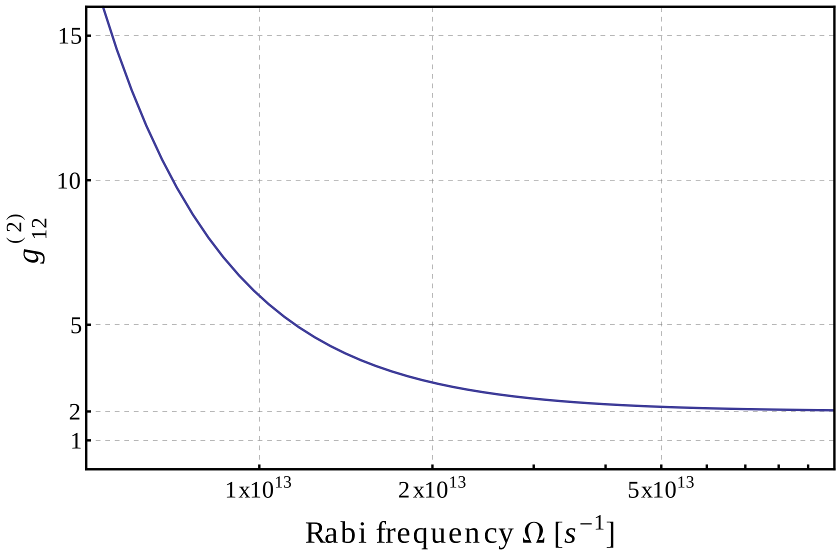

Figure IV.10.: The steady-state solution for the second-order intensity-intensity correlation function $g_{12}^{(2)}$ describing the probability of first the THz-emission followed by an optical emission as a function of the Rabi frequency Ω on a logarithmic scale. Used parameters: Transition frequency $\omega_0 = 5.0 \times 10^{15}\,\mathrm{s}^{-1}$, laser frequency $\omega_L = \omega_0 + 10^{13}\,\mathrm{s}^{-1}$, detuning $\Delta = 10^{13}\,\mathrm{s}^{-1}$ and decay rate $\gamma_0 = 3 \times 10^6\,\mathrm{s}^{-1}$ with respect to ω_0.

The steady-state solutions of Figs. IV.9 and IV.10 reveal the dependence of the intensity-intensity correlation function on the Rabi frequency Ω. We observe a very strongly decreasing curve. While the first figure covers a very broad spectrum of frequencies, the second one is limited to higher frequencies. We can see the very strong correlation between the two emitted photons. The emission of a THz-photon is preceded by an excitation of the atom which then spontaneously emits an optical photon. This explains the strong correlation, because this way the emission of a THz-photon is immediately followed by the emission of an optical photon. Since the Rabi frequency (I.19) depends on the intensity of the driving laser, we can easily tune the correlation of the emitted photon pair. We also note that $g_{12}^{(2)} > 1$, meaning that the photons obey the super-Poissonian statistics. Thus we may speak of a tunable source of strongly correlated photon pairs.

The same may also be evaluated for the correlation function describing the probability for detecting at first an optical photon and then a THz-photon. The time-dependent solution for our gamma globulin molecules are shown in Fig. IV.11 and the steady-state solution as a function of the Rabi frequency Ω is shown in Fig. IV.12. The respective correlation function is

$$\begin{aligned} g_{21}^{(2)}(t) &= \frac{\langle S^+(t)S^-(t)S^+(t)S^-(t)\rangle}{\langle S^+(t)S^-(t)\rangle\langle S^-(t)S^+(t)\rangle} \\ &= \frac{1}{\frac{1}{2} - \langle S_z(t)\rangle}, \end{aligned} \tag{IV.8}$$

whose derivation can again be found in App. B.4.

In contrast to the previous results we observe very low correlation with low Rabi frequencies Ω. As the frequency grows, the correlation also slightly grows, but never exceeds the value of $g^{(2)} = 2$. We may interpret the weak correlation as a consequence of the strong population of the excited state. This means that the probability for the emission of a THz-photon is very low, since it needs the atom to be in the ground state. The correlation function also reveals that the emitted light is almost coherent ($g_{21} \approx 1$).

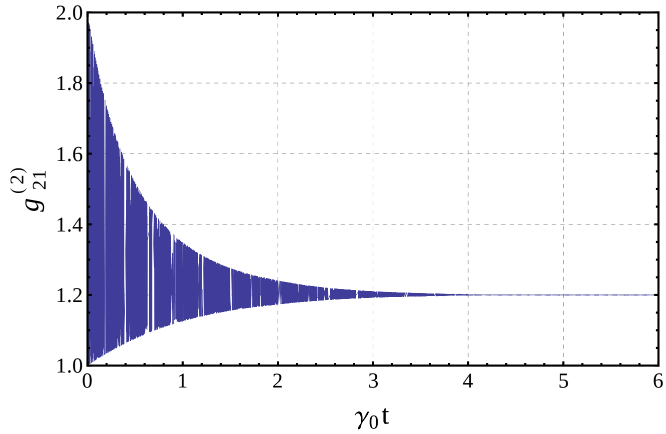

Figure IV.11.: The second-order intensity-intensity correlation function $g_{21}^{(2)}$ describing the probability of first the optical emission followed by the THz-emission as a function of time scaled by the decay rate. The used parameters are those of the gamma globulin molecule and we choose a detuning of $\Delta = 10^{12}\ s^{-1}$ and a Rabi frequency of $\Omega = 10^{12}\ s^{-1}$.

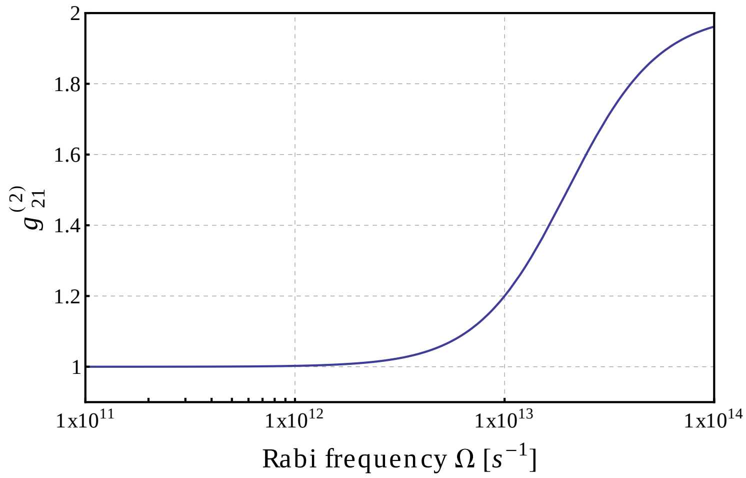

Figure IV.12.: The steady-state solution for the second-order intensity-intensity correlation function $g_{21}^{(2)}$ describing the probability of first the optical emission followed by the THz-emission as a function of the Rabi frequency Ω on a logarithmic scale. Used parameters: Transition frequency $\omega_0 = 5.0 \times 10^{15}\,\mathrm{s}^{-1}$, laser frequency $\omega_L = \omega_0 + 10^{13}\,\mathrm{s}^{-1}$, detuning $\Delta = 10^{13}\,\mathrm{s}^{-1}$ and decay rate $\gamma_0 = 3 \times 10^6\,\mathrm{s}^{-1}$ with respect to ω_0.

IV.4.3. Violation of a Cauchy-Schwarz inequality

As implied before in Sec. IV.4.1, we observe the violation of a Cauchy-Schwarz inequality

$$g_{11}^{(2)}(0)g_{22}^{(2)}(0) \geq [g_{12}^{(2)}(\tau)]^2. \tag{IV.9}$$

To prove the violation we calculate the different parts by using the relations of App. B.4. We start by calculating the correlation function of the optical photons,

$$\begin{aligned} g_{11}^{(2)} &= \frac{\langle E_1^{(-)}(t)E_1^{(-)}(t)E_1^{(+)}(t)E_1^{(+)}(t)\rangle}{\langle E_1^{(-)}(t)E_1^{(+)}(t)\rangle\langle E_1^{(-)}(t)E_1^{(+)}(t)\rangle} \\ &= \frac{\langle S^+(t)S^+(t)S^-(t)S^-(t)\rangle}{\langle S^+(t)S^-(t)\rangle\langle S^+(t)S^-(t)\rangle} = 0, \end{aligned} \tag{IV.10}$$

and repeat the same procedure for the THz-photons

$$\begin{aligned} g_{22}^{(2)} &= \frac{\langle E_2^{(-)}(t)E_2^{(-)}(t)E_2^{(+)}(t)E_2^{(+)}(t)\rangle}{\langle E_2^{(-)}(t)E_2^{(+)}(t)\rangle\langle E_2^{(-)}(t)E_2^{(+)}(t)\rangle} \\ &= \frac{\langle S^-(t)S^-(t)S^+(t)S^+(t)\rangle}{\langle S^-(t)S^+(t)\rangle\langle S^-(t)S^+(t)\rangle} = 0. \end{aligned} \tag{IV.11}$$

We thus find that the left part of Eq. (IV.9) vanishes and we are left to check if the cross-correlation $g_{12}^{(2)}(\tau)$ is different from zero. From the Figs. IV.10 and IV.9) we may read that $g_{12}^{(2)}(0) > 0$ and $g_{21}^{(2)}(0) > 0$. So we see that the Cauchy-Schwarz inequality is violated and that we are dealing with non-classical pairs of photons.

Summary and outlook

In the preceding work we investigated the behavior of a two-level quantum system with broken inversion symmetry that is strongly driven by an off-resonant laser. We have found a strong tunable correlation between the emission of a THz-photon that is just followed by the emission of an optical photon. The process is summarized in Fig. (IV.13).

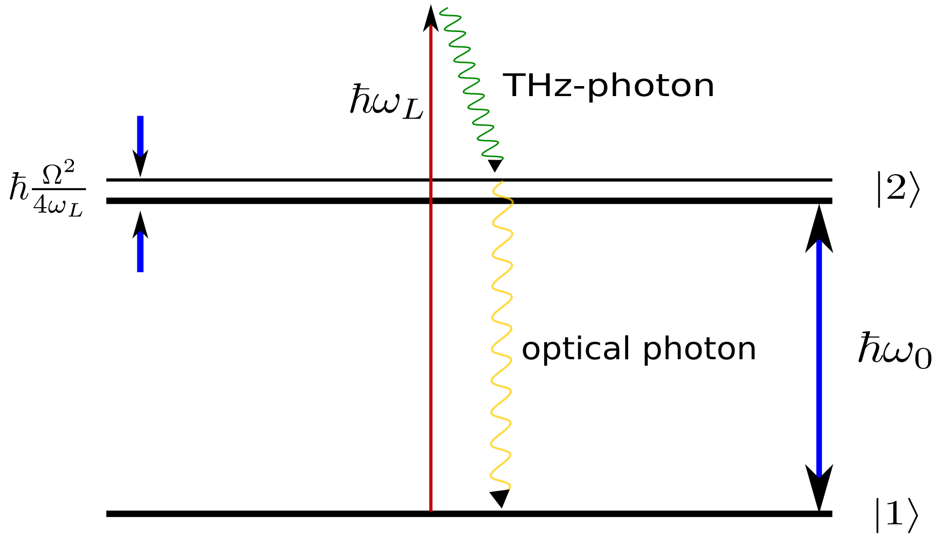

Figure IV.13.: The emission of the non-classical photon pair. The off-resonant laser of frequency ω_L excites the two-level system and induces the emission of a THz-photon that is followed by the spontaneous emission of an optical photon.

At first we have set up a Hamiltonian accounting for all the physical interactions of the system based on the semiclassical and fully quantum mechanical theory of atom-field interactions. Next we have manipulated this Hamiltonian with a unitary transformation and perturbation theory in such a way that we could evaluate it with a master equation approach, obtaining equations of motion for the atomic operators. Then we have defined different damping processes in the dynamics of the system (see Figs. IV.1, IV.2, IV.3, IV.4). The several additional processes of spontaneous emission are a direct consequence of the laser that off-resonantly drives the two-level system. Using the parameters of gamma globulin macromolecules we have calculated the intensity of the spontaneous emission of Fig. IV.3 (spontaneous emission of a THz-photon) and the intensity-intensity correlation functions between the THz-photons and the optical photons as depicted in Fig. IV.13. Gamma globulin macromolecules and quantum dots are possible physical systems for the experimental realization of our model, since they exhibit broken inversion symmetry that is a necessary condition for the emission of the THz-photon.

We have found a strong tunable correlation between the emission of a THz-photon and the subsequent emission of an optical photon. By measuring the correlation of the photon pair we have also demonstrated the violation of a Cauchy-Schwarz inequality showing its non-classical character and moreover we have seen that it follows super-Poissonian photon counting statistics. Strongly correlated non-classical pairs of photons are of great interest in the emerging field of quantum computation and quantum information. Specially non-classical photon pairs of different frequencies are needed to connect the nodes of a quantum network that have different characteristic frequencies. With the intensity of the laser we are able to tune the strength of the correlation of the photon pair and with the degree of detuning we are able to control the exact frequency of the THz-photon. This means that we propose a source for non-classical photon pairs of tunable frequency (at least one photon is tunable) and of tunable correlation.

Another application that should not be neglected is the ability to use our system as a tunable source for radiation in the THz-regime, i.e. a regime in the electromagnetic spectrum that has not yet been well studied and which still misses efficient and reliable sources.

In the preceding work we have derived equations that analyze the interaction of a single two-level system with light. In a next step it would be interesting to analyze the additional effects of a multi-particle system. In a recent paper [36] a strongly and off-resonantly driven dilute cloud of atoms without broken inversion symmetry has been studied. In a similar approach one could also take into account terms considering the broken inversion symmetry. We expect, for example, that the intensity of our THz-radiation increases, turning the system more interesting for a THz-radiation source.

A. Appendix

In order not to disturb the fluency of the present work, we have postponed some mathematical details and technical derivations. We will begin by defining the atomic transition operators and prove some important relations that are used throughout the whole work. Then we are going to show how to do an unitary transformation of the Hamiltonian, which is used in Sec. II.1. After that we will give a general solution for a linear inhomogeneous differential equation of first order, which is needed in Ch. III to solve the equations of motion of the creation and annihilation operators, $a_{\boldsymbol{k}}^\dagger$ and $a_{\boldsymbol{k}}$, respectively.

A.1. Atom transition operators

For a detailed treatment see the first chapter of [61]. At first we define our needed atomic operators for a two-level-system, with energy levels $|1\rangle$ and $|2\rangle$ with a transition frequency of ω_0:

$$S^+ = |2\rangle\langle 1|, \tag{A.1a}$$
$$S^- = |1\rangle\langle 2|, \tag{A.1b}$$
$$S_z = \frac{1}{2}(|2\rangle\langle 2| - |1\rangle\langle 1|). \tag{A.1c}$$

Supposing an ONS (orthonormal system) we can prove the following important commutation relations

$$[S_z, S^+] = S^+, \tag{A.2a}$$
$$[S_z, S^-] = -S^-, \tag{A.2b}$$
$$[S^+, S^-] = 2S_z. \tag{A.2c}$$

Using the definitions of Eqs. A.1, the proofs for Eqs. A.2 turn out to be very simple

$$\begin{aligned}[S_z, S^+] &= S_z S^+ - S^+ S_z \\ &= \frac{1}{2}(|2\rangle\langle 2| - |1\rangle\langle 1|)\,|2\rangle\langle 1| - |2\rangle\langle 1|\,\frac{1}{2}(|2\rangle\langle 2| - |1\rangle\langle 1|) \\ &= \frac{1}{2}\,|2\rangle\langle 1| + \frac{1}{2}\,|2\rangle\langle 1| \\ &= S^+,\end{aligned} \tag{A.3}$$

$$\begin{aligned}[S_z, S^-] &= S_z S^- - S^- S_z \\ &= \frac{1}{2}(|2\rangle\langle 2| - |1\rangle\langle 1|)\,|1\rangle\langle 2| - |1\rangle\langle 2|\,\frac{1}{2}(|2\rangle\langle 2| - |1\rangle\langle 1|) \\ &= -\frac{1}{2}\,|1\rangle\langle 2| - \frac{1}{2}\,|1\rangle\langle 2| \\ &= -S^-,\end{aligned} \tag{A.4}$$

and

$$\begin{aligned}[S^+, S^-] &= S^+ S^- - S^- S^+ \\ &= |2\rangle\langle 1|\,|1\rangle\langle 2| - |1\rangle\langle 2|\,|2\rangle\langle 1| \\ &= |2\rangle\langle 2| - |1\rangle\langle 1| \\ &= 2S_z.\end{aligned} \tag{A.5}$$

Next we also have to consider the following important relations,

$$S^+ S^- = \frac{1}{2} + S_z, \tag{A.6a}$$

$$S^- S^+ = \frac{1}{2} - S_z, \tag{A.6b}$$

$$S_z S^- = -\frac{1}{2} S^-, \tag{A.6c}$$

$$S_z S^+ = \frac{1}{2} S^+, \tag{A.6d}$$

$$S^+ S_z = -\frac{1}{2} S^+, \tag{A.6e}$$

$$S^- S_z = \frac{1}{2} S^-. \tag{A.6f}$$

Again we prove Eqs. A.6 by using the definitions of Eqs. A.1 and the completeness relation I.4:

$$
\begin{aligned}
S^+S^- &= |2\rangle\langle 1|\,|1\rangle\langle 2| \\
&= |2\rangle\langle 2| \\
&= \frac{1}{2}|2\rangle\langle 2| + \frac{1}{2}|2\rangle\langle 2| + \frac{1}{2}|1\rangle\langle 1| - \frac{1}{2}|1\rangle\langle 1| \\
&= \frac{1}{2}|2\rangle\langle 2| - \frac{1}{2}|1\rangle\langle 1| + \frac{1}{2} \\
&= \frac{1}{2} + S_z,
\end{aligned}
\tag{A.7}
$$

$$
\begin{aligned}
S^-S^+ &= |1\rangle\langle 2|\,|2\rangle\langle 1| \\
&= |1\rangle\langle 1| \\
&= \frac{1}{2}|1\rangle\langle 1| + \frac{1}{2}|1\rangle\langle 1| + \frac{1}{2}|2\rangle\langle 2| - \frac{1}{2}|2\rangle\langle 2| \\
&= \frac{1}{2}|1\rangle\langle 1| - \frac{1}{2}|2\rangle\langle 2| + \frac{1}{2} \\
&= \frac{1}{2} - S_z,
\end{aligned}
\tag{A.8}
$$

$$
S_zS^- = \frac{1}{2}(|2\rangle\langle 2| - |1\rangle\langle 1|)\,|1\rangle\langle 2| = -\frac{1}{2}|1\rangle\langle 2| = -\frac{1}{2}S^-, \tag{A.9}
$$

$$
S_zS^+ = \frac{1}{2}(|2\rangle\langle 2| - |1\rangle\langle 1|)\,|2\rangle\langle 1| = \frac{1}{2}|2\rangle\langle 1| = \frac{1}{2}S^+, \tag{A.10}
$$

$$
S^-S_z = |1\rangle\langle 2|\,\frac{1}{2}(|2\rangle\langle 2| - |1\rangle\langle 1|) = \frac{1}{2}|1\rangle\langle 2| = \frac{1}{2}S^-, \tag{A.11}
$$

and

$$
S^+S_z = |2\rangle\langle 1|\,\frac{1}{2}(|2\rangle\langle 2| - |1\rangle\langle 1|) = -\frac{1}{2}|2\rangle\langle 1| = -\frac{1}{2}S^+. \tag{A.12}
$$

Those simple relations are used throughout the whole thesis.

A.2. Rotating frame representation

A similar derivation may be found in various standard textbooks such as [49] in the derivation part of the interaction picture. Let H be an arbitrary Hamiltonian with respective quantum mechanical states $|\psi(t)\rangle$ satisfying the Schrödinger equation

$$i\hbar\frac{\partial}{\partial t}|\psi(t)\rangle = H|\psi(t)\rangle\,. \tag{A.13}$$

We can perform the following unitary transformation

$$|\psi(t)\rangle = e^{-\frac{i}{\hbar}H_0t}|\tilde{\psi}(t)\rangle\,, \tag{A.14}$$

which can be inserted in Eq. (A.13)

$$i\hbar(e^{-\frac{i}{\hbar}H_0t}\frac{\partial}{\partial t}|\tilde{\psi}(t)\rangle - \frac{i}{\hbar}H_0e^{-\frac{i}{\hbar}H_0t}|\tilde{\psi}(t)\rangle) = He^{-\frac{i}{\hbar}H_0t}|\tilde{\psi}(t)\rangle\,. \tag{A.15}$$

This leads to

$$i\hbar e^{-\frac{i}{\hbar}H_0t}\frac{\partial}{\partial t}|\tilde{\psi}(t)\rangle = (H-H_0)e^{-\frac{i}{\hbar}H_0t}|\tilde{\psi}(t)\rangle\,, \tag{A.16}$$

so that in the end one has again a Schrödinger equation:

$$i\hbar\frac{\partial}{\partial t}|\tilde{\psi}\rangle = H_{rot}|\tilde{\psi}\rangle\,, \tag{A.17}$$

with

$$H_{rot} = e^{\frac{i}{\hbar}H_0t}(H-H_0)e^{-\frac{i}{\hbar}H_0t}. \tag{A.18}$$

A.3. Solving a linear inhomogeneous differential equation of first order

We consider the following differential equation

$$\frac{d}{dt}f(t) = i(\omega_k - \omega_L)f(t) + g(t), \tag{A.19}$$

where $f(t)$ and $g(t)$ are sufficiently smooth functions. The homogeneous solution (for $g(t) = 0$) reads

$$f_{hom}(t) = f(0)e^{i(\omega_k - \omega_L)t}. \tag{A.20}$$

For the inhomogeneous part we make the following Ansatz

$$f_{inh}(t) = z(t)e^{i(\omega_k - \omega_L)t}, \tag{A.21}$$

where z(t) is an arbitrary function of time. This Ansatz may be inserted in Eq. (A.19)

$$\begin{aligned} \dot{z}(t)e^{i(\omega_k-\omega_L)t} + z(t)i(\omega_k - \omega_L)e^{i(\omega_k-\omega_L)t} &= i(\omega_k - \omega_L)z(t)e^{i(\omega_k-\omega_L)t} + g(t) \\ \Rightarrow \dot{z}(t)e^{i(\omega_k-\omega_L)t} &= g(t) \\ \Rightarrow \dot{z}(t) &= e^{-i(\omega_k-\omega_L)t}g(t) \\ \Rightarrow z(t) &= \int_0^t dt' e^{-i(\omega_k-\omega_L)t'}g(t'). \end{aligned} \tag{A.22}$$

Therefore the inhomogeneous solution reads

$$f_{inh}(t) = \int_0^t dt' e^{i(\omega_k-\omega_L)(t-t')}g(t'). \tag{A.23}$$

Since we are dealing with a linear equation, the solution is the sum of the homogeneous and the inhomogeneous part

$$f(t) = f_{hom}(t) + f_{inh}(t) = f(0)e^{i(\omega_k-\omega_L)t} + \int_0^t dt' e^{i(\omega_k-\omega_L)(t-t')}g(t'). \tag{A.24}$$

Similar helpful derivations may be found in [62].

A.4. Born-Markov approximation

Now we proceed to the so called Born-Markov approximation used in Eq. (III.6). In simple words, the Born approximation takes account of the weak matter-field coupling, so that an emitted photon does not react back on the atom. The Markovian approximation usually denotes the short memory approximation, where one just takes into account the most recent value of the considered operator. By pulling the operators out of the integral in Eq. (III.5), it is exactly the "history" of the operator that is not taken into account anymore. In our approximation the Hamiltonian of Eq. (II.28) will be approximated by

$$H = \hbar(\Delta + \frac{\Omega^2}{4\omega_L})S_z. \tag{A.25}$$

This way the equation of motion of $\langle S^+(t)\rangle$ may be approximated by

$$\begin{aligned} \frac{d}{dt}\langle S^+(t)\rangle &= \frac{i}{\hbar}\langle[\hbar(\Delta + \frac{\Omega^2}{4\omega_L})S_z, S^+]\rangle \\ &= i(\Delta + \frac{\Omega^2}{4\omega_L})\langle S^+\rangle . \end{aligned} \tag{A.26}$$

By means of the standard method of separation of variables for solving ordinary and partial differential equations this leads to

$$\langle S^+(t)\rangle = \langle S^+(0)\rangle\, e^{i(\Delta+\frac{\Omega^2}{4\omega_L})t}, \tag{A.27}$$

which may be solved for $\langle S^+(0)\rangle$

$$\langle S^+(0)\rangle = \langle S^+(t)\rangle\, e^{-i(\Delta+\frac{\Omega^2}{4\omega_L})t}. \tag{A.28}$$

Eq. (A.27) may also be written in terms of the time t' giving us the final desired form

$$\begin{aligned} \langle S^+(t')\rangle &= \langle S^+(0)\rangle\, e^{i(\Delta+\frac{\Omega^2}{4\omega_L})t'} \\ &= \langle S^+(t)\rangle\, e^{i(\Delta+\frac{\Omega^2}{4\omega_L})(t'-t)}. \end{aligned} \tag{A.29}$$

The Born-Markov approximation is an approximation for weak couplings of the two-level system and the quantized environment. This may be seen explicitly in the fact that the master equation III.19 only contains terms of second order in the coupling $\boldsymbol{g_k} \cdot \boldsymbol{d}/\hbar$.

B. Appendix

B.1. Perturbation theory

We follow the treatment of James [63] and Tan *et al.* [64]. We start with the Schrödinger equation in the interaction picture

$$i\hbar\frac{d}{dt}\left|\psi(t)\right\rangle = H(t)\left|\psi(t)\right\rangle, \tag{B.1}$$

which can be integrated formally

$$\left|\psi(t)\right\rangle = \left|\psi(0)\right\rangle - \frac{i}{\hbar}\int_0^t dt' H(t')\left|\psi(t')\right\rangle, \tag{B.2}$$

and substituted back into the initial Schrödinger equation B.1

$$i\hbar\frac{d}{dt}\left|\psi(t)\right\rangle = H(t)\left|\psi(0)\right\rangle - \frac{i}{\hbar}H(t)\int_0^t dt' H(t')\left|\psi(t')\right\rangle. \tag{B.3}$$

The first term on the right-hand side of the equation can be neglected and the wave function can be pulled out of the integral (Markovian approximation). We are left with

$$i\hbar\frac{d}{dt}\left|\psi(t)\right\rangle = H_{eff}\left|\psi(t)\right\rangle, \tag{B.4}$$

where

$$H_{eff} = -\frac{i}{\hbar}H(t)\int dt' H(t'). \tag{B.5}$$

B.2. Single-atom decay rate

In this section we explicitly calculate the decay rate of spontaneous emission. It has been done before and is usually done in the Weisskopf-Wigner approximation [65]. First, we make the assumption that the vacuum modes are very close to each other in frequency space, so that we can write the sum as an integral

$$\sum_k \to \frac{V}{(2\pi)^3} \int_0^\infty dk k^2 \int_0^{2\pi} d\varphi \int_0^\pi d\theta \sin\theta. \tag{B.6}$$

It follows for the decay rate of eqs. (III.17) that

$$\begin{aligned}\gamma &= \pi \sum_{\boldsymbol{k}} \frac{(\boldsymbol{g_k} \cdot \boldsymbol{d})^2}{\hbar^2} \delta(\omega_{\boldsymbol{k}} - \omega_0) \\ &= \pi \frac{V}{(2\pi)^3} \int_0^\infty dk k^2 \int_0^{2\pi} d\varphi \int_0^\pi d\theta \sin\theta \frac{(\boldsymbol{g_k} \cdot \boldsymbol{d})^2}{\hbar^2} \delta(\omega_{\boldsymbol{k}} - \omega_0).\end{aligned} \tag{B.7}$$

Using the dispersion relation for photons $E = pc = \hbar kc = \hbar\omega$ and inserting an explicit value for the coupling constant $\boldsymbol{g_k} \cdot \boldsymbol{d}$, which was defined in the end of chapter I and in Eq. (I.10), we get

$$\begin{aligned}\gamma &= \frac{V}{(2\pi)^3 c^3} \int_0^\infty d\omega \omega^2 \int_0^{2\pi} d\varphi \int_0^\pi d\theta \sin\theta \, \frac{2d^2 \pi \hbar\omega}{V\hbar^2} \sum_\lambda (\boldsymbol{e}_\lambda \cdot \boldsymbol{n})^2 \pi \delta(\omega - \omega_0) \\ &= \frac{2d^2}{\hbar(2\pi)^2 c^3} \int_0^\infty d\omega \omega^3 \int_0^{2\pi} d\varphi \int_0^\pi d\theta \sin\theta \, \sum_\lambda (\boldsymbol{e}_\lambda \cdot \boldsymbol{n})^2 \pi \delta(\omega - \omega_0),\end{aligned} \tag{B.8}$$

where we take the sum over λ, the two possible polarizations of the photon. Before continuing we make some geometrical reasoning regarding the polarization getting the expression

$$\sum_\lambda (\boldsymbol{e}_\lambda \cdot \boldsymbol{n})^2 = (\boldsymbol{e}_x \cdot \boldsymbol{n})^2 + (\boldsymbol{e}_y \cdot \boldsymbol{n})^2 = 1 - (\boldsymbol{e}_z \cdot \boldsymbol{n})^2 = 1 - \cos^2\theta, \tag{B.9}$$

which we can substitute in the above calculation leaving us with the final result

$$
\begin{aligned}
\gamma &= \frac{d^2}{\hbar(2\pi)^2 c^3} \int_0^\infty d\omega\omega^3 \int_0^{2\pi} d\varphi \int_0^\pi d\theta \sin\theta \, (1-\cos^2\theta)\pi\delta(\omega-\omega_0) \\
&= \frac{\omega_0^3 d^2}{2\hbar c^3} \int_0^\pi d\theta \sin\theta \, (1-\cos^2\theta) = \frac{\omega_0^3 d^2}{2\hbar c^3} \int_{-1}^1 dx(1-x^2) \\
&= \frac{\omega_0^3 d^2}{2\hbar c^3} \frac{4}{3} = \frac{2\omega_0^3 d^2}{3\hbar c^3} .
\end{aligned} \tag{B.10}
$$

This means that the decay rate of spontaneous emission has a cubic dependence on the transition frequency ω_0 and a quadratic dependence on the transition dipole moment $\boldsymbol{d}$ only.

B.3. Correlation functions and optical coherence

The following treatment is based on a series of papers published by Roy J. Glauber in the sixties [66, 67]. At first we define mathematically an ideal photon detector by imagining a system of negligible size that is able to absorb a photon by photoionization, for example. One may describe the field by defining an initial state $|i\rangle$ and a final state $|f\rangle$ for the absorption process. The matrix element for absorbing a photon from the field at $\boldsymbol{r}$ between times t and $t+dt$ therefore is

$$
\langle f|E^{(+)}(\boldsymbol{r},t)|i\rangle \, , \tag{B.11}
$$

where $E^{(+)}$ is the positive part of the electric field operator. Since the final state is never measured the transition probability is proportional to

$$
\begin{aligned}
w_1(\boldsymbol{r},t) &= \sum_f |\, \langle f|E^{(+)}(\boldsymbol{r},t)|i\rangle \,|^2 \\
&= \sum_f \langle i|E^{(-)}(\boldsymbol{r},t)|f\rangle \, \langle f|E^{(+)}(\boldsymbol{r},t)|i\rangle \\
&= \langle i|E^{(-)}(\boldsymbol{r},t)E^{(+)}(\boldsymbol{r},t)|i\rangle \, ,
\end{aligned} \tag{B.12}
$$

where we have used the completeness relation. Since we are never able to know exactly the initial state, we have to sum over all possibilities with probabilities P_i

$$
\begin{aligned}
w_1(\boldsymbol{r},t) &= \sum_i P_i \, \langle i|E^{(-)}(\boldsymbol{r},t)E^{(+)}(\boldsymbol{r},t)|i\rangle \\
&= Tr[\rho E^{(-)}(\boldsymbol{r},t)E^{(+)}(\boldsymbol{r},t)],
\end{aligned} \tag{B.13}
$$

where ρ is the density operator for the field defined as

$$\rho = \sum_i P_i |i\rangle \langle i| , \tag{B.14}$$

and where we have defined the trace of an operator A

$$Tr[A] = \sum_i \langle i|A|i\rangle . \tag{B.15}$$

Now we are able to define the first-order normalized quantum mechanical correlation function of the field

$$\begin{aligned} G^{(1)}(\boldsymbol{r}_1, \boldsymbol{r}_2; t_1, t_2) &= Tr[\rho E^{(-)}(\boldsymbol{r}_1, t_1) E^{(+)}(\boldsymbol{r}_2, t_2)] \\ &= \langle E^{(-)}(\boldsymbol{r}_1, t_1) E^{(+)}(\boldsymbol{r}_2, t_2)\rangle, \end{aligned} \tag{B.16}$$

and by analogy the second-order quantum mechanical correlation function

$$G^{(2)}(\boldsymbol{r}_1, \boldsymbol{r}_2, \boldsymbol{r}_3, \boldsymbol{r}_4; t_1, t_2, t_3, t_4) = \langle E^{(-)}(\boldsymbol{r}_1, t_1) E^{(-)}(\boldsymbol{r}_2, t_2) E^{(+)}(\boldsymbol{r}_3, t_3) E^{(+)}(\boldsymbol{r}_4, t_4)\rangle. \tag{B.17}$$

As a matter of convenience one can also define the normalized correlation functions

$$g^{(1)}(\boldsymbol{r}_1, \boldsymbol{r}_2; t_1, t_2) = \frac{\langle E^{(-)}(\boldsymbol{r}_1, t_1) E^{(+)}(\boldsymbol{r}_2, t_2)\rangle}{\sqrt{\langle E^{(-)}(\boldsymbol{r}_1, t_1) E^{(+)}(\boldsymbol{r}_1, t_1)\rangle \langle E^{(-)}(\boldsymbol{r}_2, t_2) E^{(+)}(\boldsymbol{r}_2, t_2)\rangle}}, \tag{B.18}$$

$$g^{(2)}(\boldsymbol{r}_1, \boldsymbol{r}_2; t_1, t_2) = \frac{\langle E^{(-)}(\boldsymbol{r}_1, t_1) E^{(-)}(\boldsymbol{r}_2, t_2) E^{(+)}(\boldsymbol{r}_2, t_2) E^{(+)}(\boldsymbol{r}_1, t_1)\rangle}{\langle E^{(-)}(\boldsymbol{r}_1, t_1) E^{(+)}(\boldsymbol{r}_1, t_1)\rangle \langle E^{(-)}(\boldsymbol{r}_2, t_2) E^{(+)}(\boldsymbol{r}_2, t_2)\rangle}. \tag{B.19}$$

B.4. Intensity and correlation as a function of atomic operators

The average light intensity at point $\boldsymbol{r}$ and time t is defined as

$$\langle I(\boldsymbol{r},t)\rangle = \langle \boldsymbol{E}^{-}(\boldsymbol{r},t)\boldsymbol{E}^{+}(\boldsymbol{r},t)\rangle \,. \tag{B.20}$$

In order to explicitly calculate the intensity of the THz-emission one needs to regard the explicit form of the creation and annihilation operators Eqs. (III.15 and III.16). The terms important for the THz-emission are listed below:

$$a^{\dagger}_{\boldsymbol{k}}(t) \sim -\frac{3G}{8\hbar\omega_L}(\boldsymbol{g_k}\cdot\boldsymbol{d})S^{-}(t)\pi\delta(\omega_{\boldsymbol{k}}+\omega_0-\omega_L+\frac{\Omega^2}{4\omega_L})e^{-i\omega_L t}, \tag{B.21}$$

and

$$a_{\boldsymbol{k}}(t) \sim -\frac{3G}{8\hbar\omega_L}(\boldsymbol{g_k}\cdot\boldsymbol{d})S^{+}(t)\pi\delta(\omega_{\boldsymbol{k}}+\omega_0-\omega_L+\frac{\Omega^2}{4\omega_L})e^{i\omega_L t}. \tag{B.22}$$

Thus the positive part of the electric field operator responsible for the THz-emission has the following form

$$\begin{aligned}\boldsymbol{E}^{+}_{THz}(\boldsymbol{r},t) &= i\sum_{\boldsymbol{k}}\hat{\epsilon}_{\boldsymbol{k}}\varepsilon_{\boldsymbol{k}}a_{\boldsymbol{k}}e^{-i\omega_{\boldsymbol{k}}t+i\boldsymbol{k}\cdot\boldsymbol{r}}\\ &= -\sum_{\boldsymbol{k}}\hat{\epsilon}_{\boldsymbol{k}}\varepsilon_{\boldsymbol{k}}\frac{3G}{8\hbar\omega_L}(\boldsymbol{g_k}\cdot\boldsymbol{d})S^{+}(t)\pi\delta(\omega_{\boldsymbol{k}}+\omega_0-\omega_L+\frac{\Omega^2}{4\omega_L})\\ &\quad\times e^{i(\omega_L-\omega_{\boldsymbol{k}})t+i\boldsymbol{k}\cdot\boldsymbol{r}},\end{aligned} \tag{B.23}$$

and the negative part is just the hermitian conjugate

$$\begin{aligned}\boldsymbol{E}^{-}_{THz}(\boldsymbol{r},t) &= -i\sum_{\boldsymbol{k}}\hat{\epsilon}_{\boldsymbol{k}}\varepsilon_{\boldsymbol{k}}a^{\dagger}_{\boldsymbol{k}}e^{i\omega_{\boldsymbol{k}}t-i\boldsymbol{k}\cdot\boldsymbol{r}}\\ &= \sum_{\boldsymbol{k}}\hat{\epsilon}_{\boldsymbol{k}}\varepsilon_{\boldsymbol{k}}\frac{3G}{8\hbar\omega_L}(\boldsymbol{g_k}\cdot\boldsymbol{d})S^{-}(t)\pi\delta(\omega_{\boldsymbol{k}}+\omega_0-\omega_L+\frac{\Omega^2}{4\omega_L})\\ &\quad\times e^{-i(\omega_L-\omega_{\boldsymbol{k}})t-i\boldsymbol{k}\cdot\boldsymbol{r}}.\end{aligned} \tag{B.24}$$

So we may say that

$$\begin{aligned} \boldsymbol{E}_{THz}^{+}(\boldsymbol{r},t) \propto S^{+}(t), \\ \boldsymbol{E}_{THz}^{-}(\boldsymbol{r},t) \propto S^{-}(t), \end{aligned} \tag{B.25}$$

getting a simple expression for the intensity of the THz-emission

$$\langle I(\boldsymbol{r},t)\rangle \propto \langle S^{-}(t)S^{+}(t)\rangle \,. \tag{B.26}$$

This expression is in contradiction with the normal definition of intensity found in general textbooks such as [12] that have the atomic operators $S^{-}(t)$, $S^{+}(t)$ in the inverse order. The physical explanation may be given by looking at Fig. IV.3. We see that in order to emit a THz-photon, the atom has to be excited from the ground state into the excited state, contradicting the normal process, in which a photon is emitted, just when the two-level system is already excited.

The same may be done for the second order correlation function

$$g_{ij}^{(2)}(\tau) = \frac{\langle E_i^{(-)}(t)E_j^{(-)}(t+\tau)E_j^{(+)}(t+\tau)E_i^{(+)}(t)\rangle}{\langle E_i^{(-)}(t)E_i^{(+)}(t)\rangle\langle E_j^{(-)}(t)E_j^{(+)}(t)\rangle}, \tag{B.27}$$

with the difference that the electric field operator is not just in the numerator, but also in the denominator of the expression. This means that we may not only derive a proportionality, but an exact equality of $g_{ij}^{(2)}$ as a function of atomic operators, since the proportionality factors just cancel. Therefore we have for the correlation function describing the probability for the emission of a THz-photon followed by an optical one

$$g_{12}^{(2)}(0) = \frac{\langle S^{-}(t)S^{+}(t)S^{-}(t)S^{+}(t)\rangle}{\langle S^{-}(t)S^{+}(t)\rangle\langle S^{+}(t)S^{-}(t)\rangle}, \tag{B.28}$$

and for the function describing the probability for the emission of an optical photon followed by a THz-photon

$$g_{21}^{(2)}(0) = \frac{\langle S^{+}(t)S^{-}(t)S^{+}(t)S^{-}(t)\rangle}{\langle S^{+}(t)S^{-}(t)\rangle\langle S^{-}(t)S^{+}(t)\rangle}. \tag{B.29}$$

B.5. Decay rates as a function of the Rabi frequency

As calculated in the appendix B.2 the expression for the decay rate is the following

$$\gamma_0 = \gamma(\omega_0) = \frac{2}{3}\frac{d^2\omega_0^3}{c^3\hbar}. \tag{B.30}$$

In order to calculate the frequency dependent decay rates as a function of γ_0 and the Rabi frequency Ω we do the following transformations

$$\begin{aligned} \gamma(\omega_L) = \frac{2}{3}\frac{d^2\omega_L^3}{c^3\hbar} &= \frac{2}{3}\frac{d^2\omega_0^3}{c^3\hbar}(1+\frac{\Omega^2}{4\omega_L\omega_0}+\frac{\Delta}{\omega_0})^3 \\ &= \gamma_0\left(1+\frac{\Omega^2}{4\omega_L\omega_0}+\frac{\Delta}{\omega_0}\right)^3, \end{aligned} \tag{B.31}$$

$$\begin{aligned} \gamma(\omega_0+\frac{\Omega^2}{4\omega_L\omega_0}) &= \frac{2}{3}\frac{d^2\omega_0^3}{c^3\hbar}(1+\frac{\Omega^2}{4\omega_L\omega_0})^3 \\ &= \gamma_0(1+\frac{\Omega^2}{4\omega_L\omega_0})^3, \end{aligned} \tag{B.32}$$

and

$$\begin{aligned} \gamma(\omega_L-\omega_0-\frac{\Omega^2}{4\omega_L}) &= \frac{2}{3}\frac{d^2\omega_0^3}{c^3\hbar}(\frac{\omega_L}{\omega_0}-1-\frac{\Omega^2}{4\omega_L\omega_0})^3 \\ &= \gamma_0(\frac{\Delta}{\omega_0})^3. \end{aligned} \tag{B.33}$$

Those expressions are used in the numerical calculations of the stationary functions depending on the Rabi frequencies.

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Ich versichere, dass ich diese Arbeit selbstständig verfasst habe und keine anderen als die angegebenen Quellen und Hilfsmittel benutzt habe.

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