Density Matrices and the Quantum Monte-Carlo Method in Quantum Optics

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Abstract: In this set of lecture notes we present a wave function approach to study the evolution of a small system coupled to a large reservoir.

First, we give a summary of the standard treatment of such systems based on density matrices. A practical point of view is taken in this part, emphasizing equations that allow efficient numerical solution.

We then introduce the Quantum Monte-Carlo approach, which is a method in which an ensemble of wave-functions is evolved with a random element present in the evolution of every individual state vector. We prove that this method is equivalent to the standard approach for a wide class of relaxation problems.

For systems with a number of states N much larger than unity, the Quantum Monte-Carlo approach can be less "expensive" in terms of calculation time than the Master Equation treatment. We present some examples of numerical applications of the method, and we also consider the additional insight that may be gained from the existence of alternative stochastic wave-function schemes.

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Contents:

1 Introduction to this course

A quantum system which is not fully isolated from its surroundings must be described by quantities which are more general than just a wavefunction, and its dynamics shows features beyond those contained in the usual solutions of Schrödinger's equation. It is the purpose of these lectures to present a review of the tools that may be applied to a system which is weakly coupled to a reservoir into which it dissipates energy. An atom decaying by spontaneous emission due to the coupling to the surrounding quantized electromagnetic field, or a field mode which is damped due to absorption of photons by atoms in a medium, are examples of such "open systems" in quantum optics. Damping in quantum optics problems is special because, unlike in other fields of physics, it may occur on the same time scale as coherent processes (e.g. laser excitation). Thus it neither restricts the system dynamics to the final state of the decay process, nor can it be accounted for by a simple perturbative correction to the result of the coherent evolution.

First, the standard treatment of dissipation in quantum optics is reviewed. The density matrix is introduced, and the structure of the master equation, yielding its time evolution, is discussed together with another important theoretical result, the quantum regression theorem. I try to make a slightly different presentation than what is found in many text books, serving hopefully as a supplement to the understanding for the more experienced reader, and as a short cut to the subject for the beginner in this field.

Recently, a novel treatment of such systems was introduced. It is possible to apply wave functions rather than density matrices, provided a stochastic element is included in the evolution of the wave functions. This new insight was obtained more or less simultaneously in a number of groups although the motivation and reasoning applied differed a lot.

Together with Yvan Castin and Jean Dalibard at ENS in Paris I had reached a rather pessimistic view on the feasibility of numerical calculations on laser cooling in the general case where the external degrees of freedom have to be described quantum mechanically [1]. The problem is the important role of spontaneous emission for the process; one has to apply density matrices with a number of elements equal to the square of the number of state vectors describing the system. For laser cooling this easily brings the number of equations to be solved up to $10^8 - 10^{12}$!

What a substantial reduction if one could do the calculations with wavefunctions rather than with a density matrix. Without blaming this on my colleagues, I was so fortunate to be misled by the misconception that any single atom, being only one single quantum system, should be described by a *wave function*, and the density matrix should be reserved for ensembles of systems. This view point is not consistent with the definition of the density matrix in these notes (see e.g. the wave function discussed

above Eq.(7)), but it was, for me, an important driving force, and, as these lectures will demonstrate, a wave function formulation *can* be formulated. It is clear that the proper time evolution has to be stochastic in order to represent the density matrix evolution from pure into mixed states. Jean Dalibard brought in some of his experience from the delay function treatment of the quantum jump problem [2], and we constructed the Monte Carlo Wave Function method (MCWF) [3, 4, 5]. In Section 3 of these notes we shall present this method in detail and we shall discuss the physical interpretation of the crucial stochastic elements in the evolution: The association of a wave function to each individual system can be made meaningful by a reference to the theory of measurements in quantum mechanics.

A new misconception which, however, did not last long, was that these indeterministic wave functions could provide a short cut to the calculation of correlation functions. This is unfortunately not so, the fluctuations in operator averages due to the random character of the wave function evolution may represent some, but certainly not all the "quantum noise" which is so important in many quantum optics problems. In Section 4 we discuss the application of the quantum regression theorem together with the wave function simulations to obtain the correct noise correlations. We also discuss the existence of different simulation schemes, some involving jumps, some being in the form of continuous stochastic differential equations, and all representing the same system equally well.

In the context of non classical field generation, Carmichael [6] has proposed an approach named "quantum trajectories", inspired by the theory of photoelectron counting [7] and quite similar to the spirit of [3] . Based on the continuous quantum theory of measurement [8], Zoller and co-workers [9] have developed a Monte-Carlo simulation of the atomic master equation for spontaneous emission. Within the framework of quantum jump theory, Hegerfeldt and Wilser have considered a quantum mechanical model for describing a single radiating atom which could also be the starting point for a Monte-Carlo evolution with atomic wave functions [10]. Finally the relation to a general stochastic formulation of quantum mechanics, considered by a number of authors, was pointed out to us by Gisin [11]. Gisin and Percival have since then applied their "state diffusion model" to a number of quantum optics problems [12].

We have referred to our approach as the Monte Carlo Wave Function method (MCWF). We hesitated calling it "Quantum Monte Carlo" because this name is already associated with another method in theoretical physics, transforming Schrödinger's equation into a thermodynamics type problem solvable by classical Monte Carlo simulations. Our procedure does not elliminate the quantum nature of the problem at any point. In several applications, however, the method has now been referred to as the Quantum Monte Carlo method, and I shall use this name throughout.

It has been a pleasure for me to work with Yvan Castin, Jean Dalibard and Kirstine Berg-Sørensen on the development and application of the MCWF method. Alterna-

tive viewpoints and applications, presented by other groups, have been, and still are, important sources of inspiration to us, and I have enjoyed the enthusiastic atmosphere in our discussions with the above mentioned authors, and many others, so far.

These lecture notes have improved substantially from the initial version to the present result. This is, in particular, due to a careful reading of the manuscript by Ejvind Bonderup, and I am very grateful for his numerous suggestions to the presentation.

2 Standard treatment of dissipation in quantum optics: Density Matrices, the Master Equation, the Quantum Regression Theorem.

2.1 The Density Matrix as a Catalogue

Consider a quantum system, S. Using a basis set $\{|i\rangle\}$ we can write the wavefunction for the system,

$$|\psi\rangle = \sum_i c_i |i\rangle \tag{1}$$

and we can cast Schrödinger's equation for the wavefunction

$$i\hbar \frac{d}{dt}|\psi\rangle = H|\psi\rangle \tag{2}$$

into a set of linear equations coupling the amplitudes c_i.

In quantum mechanics, physical quantities are represented by linear operators, acting in the space of wavefunctions, and the theory predicts the average outcome of experiments measuring the corresponding physical property. A linear operator, A, is specified completely by its action on all basis states,

$$A|j\rangle = \sum_i A_{ij}|i\rangle. \tag{3}$$

Hence, if $\{|i\rangle\}$ is an orthonormal set, we can write the operator as

$$A = \sum_{ij} A_{ij}|i\rangle\langle j|. \tag{4}$$

The outer, diadic, product, $|i\rangle\langle j|$, of two basis functions is defined as the operator in the system Hilbert space which maps a state vector $|\psi\rangle$ into a vector proportional to $|i\rangle$ with amplitude given by the overlap of $|\psi\rangle$ with the j'th basis function, $(|i\rangle\langle j|)|\psi\rangle = \langle j|\psi\rangle \cdot |i\rangle$. It is convenient to apply this element of Dirac notation in the quantum optics problems considered in these lectures.

Any operator can be written in the form (4), hence all information about the system is contained in the set of expectation values $\big\langle(|i\rangle\langle j|)\big\rangle$. For a system described by a finite number N of basis functions, this implies that a matrix of $N \times N$ elements gives access to all possible system observables. Now, we already have a set of only N variables, the amplitudes c_i, which provides the same information, so why consider an extension of this set ?

One answer to this question lies in the observation that if we have for example a big ensemble of systems, or when our system is part of a much larger quantum mechanical system, with a total number of basis states that may easily exceed N^2, any observable referring to the S-component of the system, is still determined by the $N \times N$ elements

mentioned. In this perspective N^2 may be a small number. It is therefore useful to introduce a *catalogue* containing these numbers. We call this catalogue the ***density matrix*** (restricted to the system S), and we define

$$\rho_{ij} = \Big\langle (|j\rangle\langle i|) \Big\rangle. \tag{5}$$

Note how, for a pure state (1), $\rho_{ij} = c_i c_j^*$, and for an ensemble of systems with different wavefunctions, $|\psi^{(m)}\rangle = \sum_i c_i^{(m)} |i\rangle$, the ensemble and quantum expectation value in (5) becomes

$$\rho_{ij} = \frac{1}{m} \sum_m c_i^{(m)} c_j^{(m)*} \equiv \overline{c_i c_j^*}. \tag{6}$$

We have introduced the bar $\overline{\cdot}$ to indicate the ensemble average. The statistical definition, (6), of the density matrix may be found in a number of quantum mechanics text books, but is is not necessarily a good picture of the situation in quantum optics. Here, we are more often facing a system coupled to some other variables, so that the total wavefunction may be represented by product states, $|i\rangle \otimes |\alpha\rangle$, where $|i\rangle$ denotes one of the basis states of system S, and where the states $|\alpha\rangle$ span the remaining components. For a single system with such a wavefunction, $|\Psi\rangle = \sum_{i\alpha} c_{i\alpha} |i\rangle \otimes |\alpha\rangle$, we have the density matrix elements,

$$\rho_{ij} = \sum_\alpha c_{i\alpha} c_{j\alpha}^*, \tag{7}$$

and it may be more economical to deal with the N^2 elements of ρ than the, maybe, larger number of amplitudes $c_{i\alpha}$. Provided, of course, that we can get access to these values - the subject of the next subsection.

A few remarks about the density matrix: The diagonal element ρ_{ii} yields the population of the state $|i\rangle$. If the system is given by a pure state as in (1) with non-vanishing populations of two or more basis states, the state is in a *coherent superposition*, and the non-vanishing density matrix element $\rho_{ij} = c_i c_j^*$ is called the *coherence*, or the $(ij)-$coherence. In the more general case described by (6) and (7) we note that the (ij)-coherence may be zero, while both populations of the states $|i\rangle$ and $|j\rangle$ are finite; ρ is then said to represent an *incoherent* mixture of the states involved. The density matrix has unit trace (normalization), $Tr(\rho) = \sum_i \rho_{ii} = 1$, and it is a hermitian matrix, $\rho_{ij} = \rho_{ji}^*$, which is easily seen from any of the equations (5,6,7).

All of ρ's eigenvalues are positive, and having unit sum, they can readily be interpreted as the populations of the eigenstates of the density matrix. In this basis there are no coherences, and in most discussions of the density matrix these eigenstates are taken as a preferred basis and each individual system of the ensemble is claimed to be in one of these states, so that the fractions of an ensemble populating each state reflect the populations in ρ. In this way the density matrix unites classical probabilities and usual wave functions. The diagonalization of ρ may be an efficient way of synthesizing a density matrix, i.e., preparing an ensemble with certain average properties, but for the cases we are interested in there is no reason to prefer this basis. The

eigenstates of ρ have completely arbitrary relations to for example the "true" states $|\psi^{(m)}\rangle$ in a real ensemble (6), or to the states conditioned on properties outside S such as $|\psi_{|\alpha}\rangle \propto \sum_i c_{i\alpha}|i\rangle$. The Quantum Monte Carlo method replaces ρ by an ensemble of wavefunctions, and various aspects of the corresponding theory will elucidate this point further.

It is natural to associate to the matrix $\{\rho_{ij}\}$ an operator, ρ, the *density operator*, which in analogy to (4) can be written,

$$\rho = \sum_{ij} \rho_{ij}|i\rangle\langle j|. \tag{8}$$

A pure state $|\psi\rangle$ has the density operator, $\rho = |\psi\rangle\langle\psi|$. The ensemble (6) leads to $\rho = m^{-1}\sum_m |\psi^{(m)}\rangle\langle\psi^{(m)}|$, and for a system which is part of a larger problem, the density operator is obtained as a *partial trace*, $\rho = \sum_\alpha \langle\alpha|\Psi\rangle\langle\Psi|\alpha\rangle$, where $|\Psi\rangle$ is of the type leading to (7). With this symbol it is possible to apply a compact notation for matrix elements and expectation values, e.g.

$$\langle A\rangle = Tr(\rho A). \tag{9}$$

We find from Schrödinger's equation that the density matrix evolves according to

$$\frac{d\rho}{dt} = \frac{1}{i\hbar}[H,\rho]. \tag{10}$$

The time dependence of a matrix element ρ_{ij} is found by taking the $\langle i|\ \cdot\ |j\rangle$ matrix element of both sides of Eq.(10) with ρ on the form of Eq.(8).

2.2 The Master Equation

The fact that $\rho(t)$ can be calculated with not too much effort is crucial for a number of quantum optics problems.

2.2.1 On the need for a new equation

The equation (10) may be applied for example to collision problems where the evolution is unitary, given by the Hamiltonian H, but where the initial state is represented as a mixture. In this case (10) organizes the theory: angular momentum properties, impact parameter and velocity distributions could be formulated in terms of density matrix theory, but as long as the statistics is only in the initial conditions and not in the dynamics of the process, such a formulation is, if not impractical, then at least not necessary. In practical applications it is more convenient to solve the Schrödinger equation (2) for each of the relevant initial wavefunctions, and then to perform an incoherent average at the end.

A different situation appears for an ensemble of identical atoms, which are all individually described by the Hamiltonian H, but which in addition may undergo elastic collisions, either with one another or with a background gas in the experiment. The Hilbert space of wavefunctions describing all constituents of this large system may be enormous, and we may consider instead the state vector of a single atom in the ensemble, and include a random element in its evolution. Consider for example a two-level atom with a wavefunction $|\psi\rangle = c_g|g\rangle + c_e|e\rangle$. During a collision the atomic energy levels may be perturbed, and as a consequence of the collision the amplitudes are multiplied by extra phase factors, $e^{i\chi_g}$, $e^{i\chi_e}$. The products of amplitudes $c_i c_j^*$ are unchanged for $i = j$, and they experience a complex rotation of $\pm(\chi_g - \chi_e)$ for $i \neq j$. We obtain therefore, with some assumptions about the statistics of the collisions, see e.g. Ref.[13], a modification of the density matrix elements, defined as in (6), and (10) is replaced by

$$\frac{d}{dt}\rho = \frac{1}{i\hbar}[H_S, \rho] + \mathcal{L}_{\rm relax}[\rho]. \tag{11}$$

$\mathcal{L}_{\rm relax}[\rho]$ describes the decay of the mean value $\overline{c_g c_e^*}$ and its complex conjugate due to the randomness of the collisions. Below we shall indicate how the relaxation operator $\mathcal{L}_{\rm relax}$ may be derived from the full system+surroundings Hamiltonian.

The density matrix elements, defined as in (7), may experience a similar damping if the system + its surroundings initially populates a state $|\Psi\rangle = |\psi\rangle \otimes |\alpha_0\rangle$ and then, because of the coupling, gradually evolves into an entangled state populating different $|\alpha\rangle$'s. The best example is probably that of a two-level atom coupled to the quantized electromagnetic field. Due to the coupling, see (35) below, an initial atomic superposition state with no photons present, will evolve into a state populating also different one-photon states $|1_\lambda\rangle$:

$$(c_g|g\rangle + c_e|e\rangle) \otimes |0\rangle \rightarrow \quad (c'_g|g\rangle + c'_e|e\rangle) \otimes |0\rangle + \textstyle\sum_\lambda c_\lambda |g\rangle \otimes |1_\lambda\rangle \tag{12}$$

By going to second order in the coupling, Wigner and Weisskopf showed, that a discrete state $|e\rangle$ coupled to a broad continuum acquires both a shift (here: Lamb shift) and becomes unstable, i.e. c_e decays in time [1].

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[1] Let W_λ denote the coupling matrix element between the discrete level $|i\rangle = |e\rangle \otimes |0\rangle$ and the continuum states $|\lambda\rangle = |g\rangle \otimes |1_\lambda\rangle$. We write the wave function as
$|\Psi(t)\rangle = b(t)e^{-iE_i t/\hbar}|i\rangle + \sum_\lambda b_\lambda(t)e^{-iE_\lambda t/\hbar}|\lambda\rangle$.
The Schrödinger equation for the amplitudes b_λ
$i\hbar\frac{d}{dt}b_\lambda(t) = e^{i(E_\lambda - E_i)t/\hbar}W_\lambda b(t)$
is integrated, and the result is inserted in the equation for $b(t)$:
$\frac{d}{dt}b(t) = \frac{-1}{\hbar^2}\sum_\lambda \int_0^t dt' e^{i(E_\lambda - E_i)(t'-t)/\hbar}|W_\lambda|^2 b(t')$.
Now, if $|W_\lambda|^2$ is essentially constant over a broad energy range $\hbar\Delta$, and if $b(t)$ can be assumed to vary on a long time scale τ, so that we can choose $\Delta^{-1} \ll t \ll \tau$, we may take $|W_\lambda|^2$ and $b(t')$ out of the t'-integral above. The integral of the remaining exponential equals $\hbar[\pi\delta(E_\lambda - E_i) + i\mathcal{P}(\frac{1}{E_\lambda - E_i})]$. The subsequent sum (integral) over the energies E_λ (with the appropriate density of states) then gives the factors in the closed equation for $b(t)$,
$\frac{d}{dt}b(t) = (-\frac{\Gamma}{2} + i\frac{\delta E}{\hbar})b(t)$,
where $\Gamma \propto |W_\lambda|^2_{(E_\lambda = E_i)}$. The energy shift δE (the principal value part of the integral) is included in the energy E_i in practical applications.

Correspondingly the squared norm of the last term in (12) will grow linearly with time for short times, and not quadratically which is the more common quantum mechanical evolution. The evolution of the density matrix elements is described by the master equation (11), where the Lamb shift may be absorbed in H_S, and where the relaxation terms may be written

$$\dot{\rho}_{ee} = -\Gamma\rho_{ee} \quad \dot{\rho}_{eg} = -\frac{\Gamma}{2}\rho_{eg} \quad \dot{\rho}_{ge} = -\frac{\Gamma}{2}\rho_{ge} \quad \dot{\rho}_{gg} = \Gamma\rho_{ee}. \tag{13}$$

2.2.2 Elements of a general derivation of a master equation

Consider first a closed system. The Schrödinger picture, in which wavefunctions are evolved in time according to (2) is equivalent to the Heisenberg picture, where the state vector is kept fixed but operators are evolved in time according to

$$\frac{d}{dt}A = \frac{1}{i\hbar}[A, H]. \tag{14}$$

This is verified by checking that expectation values of operators evaluated in the two pictures agree, $\langle\psi(t)|A|\psi(t)\rangle = \langle\psi|A(t)|\psi\rangle$.

The Heisenberg equation of evolution (14) also applies when the operator A is one of the dyads $(|i\rangle\langle j|)$. In the Heisenberg picture also these operators are time dependent, loosened from their connection, at $t = 0$, to the set of basis states. The operator $(|i\rangle\langle j|)$ therefore obeys (14), and since the commutator $[|i\rangle\langle j|, H]$ can be expressed as a linear combination of the $(|i'\rangle\langle j'|)$'s we obtain a set of linear equations for these operators which may be solved. We shall come back to this set of equations shortly. According to (5), the mean values of these operators (and it is much easier to deal with numbers than operators in numerical applications) are the elements of the density matrix (8). The Schrödinger and Heisenberg pictures are equivalent, and Eqs.(10) and (14) give the same results for $\rho_{ij}(t)$ and $\left\langle(|j\rangle\langle i|)(t)\right\rangle$.

We shall now indicate how to obtain a set of equations for the elements of ρ, in which certain terms represent the effect of the interaction with a larger system. This is clearly not possible in all situations, and without being too specific, we restrict ourselves to the situation of a system S interacting with a "reservoir", R, illustrated in Fig. 1.

Two features characterize the coupling between these two components: it is weak, hence it can be treated by perturbation theory, the reservoir is big and "broad band", hence its state is assumed unchanged by the interaction with the much smaller system S, and it has no "memory". The precise derivation of the theory can be found in one of the text book references [13, 14, 15, 16, 17, 18], here we shall merely indicate the mechanisms and the origin of the terms appearing.

The combined system+reservoir is described by a Hamiltonian

$$H = H_S + H_R + H_{SR}, \tag{15}$$

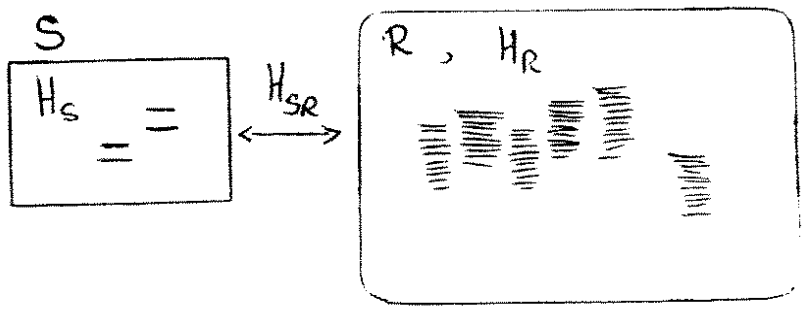

Figure 1: *Schematic representation of a "small" system, S, with few states, coupled to a "big" reservoir, R.*

and we assume at this point that any constant or time dependent perturbation of the system, as for example a laser field exciting some atomic transition, is included in the system Hamiltonian H_S. H_R describes the free evolution of the reservoir, and the coupling between the system and reservoir is H_{SR}.

If we assume that $[|i\rangle\langle j|, H_{SR}] = \sum_\lambda c_{ij,\lambda} R_\lambda$, and $[R_\lambda, H_{SR}] = \sum_{i'j'} \tilde{c}_{\lambda,i'j'} |i'\rangle\langle j'|$, where the operators R_λ act only on the reservoir, we have the Heisenberg equations of evolution[2]

$$\frac{d}{dt}|i\rangle\langle j| = \frac{1}{i\hbar}[|i\rangle\langle j|, H_S + H_R] + \frac{1}{i\hbar}\sum_\lambda c_{ij,\lambda} R_\lambda \tag{16}$$

$$\frac{d}{dt}R_\lambda = \frac{1}{i\hbar}[R_\lambda, H_S + H_R] + \frac{1}{i\hbar}\sum_{i'j'} \tilde{c}_{\lambda,i'j'} |i'\rangle\langle j'|. \tag{17}$$

This evolution is due to the total Hamiltonian. Both H_S, H_R and H_{SR} are therefore time dependent, and the $(|i\rangle\langle j|)$'s pick up "reservoir character" just like the R_λ's pick up "system character". In Section 2 we consider a two-level atom coupled to the quantized radiation field, and here the field operators acquire terms proportional to the atomic dipole - the classical radiated field is proportional to the dipole. At time zero where the Schrödinger and Heisenberg picture agree, the commutator $[|i\rangle\langle j|, H_S]$ is a certain combination of the $(|i'\rangle\langle j'|)$'s, but then this combination remains the same, and Eqs.(16,17) can be applied at all times. This relies on the identity: $[(|i\rangle\langle j|)(t), H_S(t)] = [U(t)(|i\rangle\langle j|)U^\dagger(t), U(t)H_S U^\dagger(t)] = U(t)[(|i\rangle\langle j|), H_S]U^\dagger(t)$, where $U(t)$ is the unitary time evolution operator due to the total Hamiltonian (15). I considered only one of the terms; note that the H_R-term in (16) and the H_S-term in (17) vanish identically.

We now formally integrate Eq.(17) and insert the result into Eq.(16) to make three kinds of terms appear: system dyads due to the commutator with H_S, reservoir oper-

[2] Actually the commutator $[|i\rangle\langle j|, H_{SR}]$ may well be a sum of reservoir operators, ***multiplying certain system operators***, this has no consequence for the arguments in the derivation of the master equation.

ators R_λ^0 evolving in time due to H_R, only, and, to second order in the H_{SR} coupling strength, a new set of system dyads.

The reservoir-assumption is applied to ensure two non-trivial features: The weak coupling to the small system does not change the R_λ's significantly, so that at any time we may integrate Eq.(17) from the undisturbed value R_λ^0, assuming for simplicity that all relevant $\langle R_\lambda \rangle$ expectation values vanish in the state of the reservoir. The term proportional to $\tilde{c}_\lambda$ is an integral over t' containing a factor $\exp(i\omega_\lambda(t'-t))$ in the integrand, where ω_λ is the frequency at which R_λ evolves. The reservoir is "broad band", i.e., it is described by operators R_λ evolving at very different frequencies ω_λ. The sum over λ in (16) therefore effectively introduces a delta function in time which picks out the system operators $|i'\rangle\langle j'|$ at time t. Note the similarity between this reasoning and the Wigner Weisskopf derivation sketched in our previous footnote.

Taking finally expectation values on both sides of (16), we get a closed set of equations for the density matrix elements. For a more detailed discussion of these points the reader may consult one of the textbooks listed in the reference section.

The above discussion is meant to elucidate how the mean values of the $|i\rangle\langle j|$'s, i.e. the density matrix elements, obey a closed set of equations. This is the master equation, and we note that it can be written as in (11). $\mathcal{L}_{\mathbf{relax}}[\rho]$ is linear in the density matrix elements, but more than that: in order to preserve the interpretation of ρ as a density matrix, i.e. $Tr(\rho) = 1$, $\langle\psi|\rho|\psi\rangle \geq 0$ for any $|\psi\rangle$, we must limit $\mathcal{L}_{\mathbf{relax}}[\rho]$ to be of so-called Lindblad form [19]:

$$\mathcal{L}_{\mathbf{relax}}[\rho] = -\frac{1}{2}\sum_i (C_i^+ C_i \rho + \rho C_i^+ C_i) + \sum_i C_i \rho C_i^+, \tag{18}$$

where the sum is over one, a few, or sometimes many different system operators C_i (C_i^+ is the adjoint of C_i, and both C_i and C_i^+ have dimensions of $\sqrt{1/time}$). [3]

If we take a basis for the system Hilbert space we get a matrix representation of the density operator. We are now in the Schrödinger picture and the density matrix elements ρ_{ij} obey a set of coupled first order differential equations:

$$\frac{d}{dt}\rho_{ij} = \sum_{i'j'} M_{ij,i'j'}\rho_{i'j'}. \tag{19}$$

We can therefore arrange the elements ρ_{ij} in a vector, $\vec{\rho}$, and implement the problem on a computer as a usual set of linear equations with a *matrix* M, $\dot{\vec{\rho}} = M\vec{\rho}$. Like H_S, M may be explicitly time dependent. For many problems, however, H_S is either constant or, in a proper frame, the equations of motion may be presented in a form with constant coefficients. Parts of the following discussions are only applicable in this case.

[3] If you consider the equation (14), integrated to second order in the interaction picture with respect to $H_S + H_R$, terms like $[[A, H_{SR}(t')], H_{SR}(t'')]$ appear naturally. Assuming that H_{SR} consists of products of system and reservoir operators, the double commutator leads to terms like in (18), where the reservoir parts have been reduced to numerical factors, absorbed in the C_i operators.

If one has determined the eigenvalues $\{\eta\}$ of the matrix M, and the associated eigenvectors, $M\vec{u}^{\eta} = \eta\vec{u}^{\eta}$, the time evolution is simply given by

$$\vec{\rho}(t) = \sum_{\eta} c_{\eta} \cdot e^{\eta t}\vec{u}^{\eta}, \tag{20}$$

where the c_{η}'s are coefficients determined from the values of the $\rho_{ij}(0)$. Note that M is not hermitian; this is not required to establish an equation like (20), but in order to determine the "coordinates" c_{η}, special means involving left and right eigenvectors must be applied, see, e.g. Ref.[20]. [4]

For many problems, one is interested in stationary expectation values. The matrix M is not invertible, the rows providing the evolution of populations ρ_{ii} are linearly dependent because of the constraint, $Tr(\rho) = \sum_i \rho_{ii} = 1$. This automatically ensures the existence of at least one stationary state, which may be found by matrix inversion. In practice one has to replace, say, the $i_0 i_0$'th row of M by the normalization condition (zero's for $i' \neq j'$ and some constant c for $i' = j'$) and apply the inverse of the resulting matrix to a vector with the value c in the $i_0 i_0$'th position and zero elsewhere. For a problem of reasonable size, this provides a method that may be applied for any density matrix, and which is easy to implement in a computer program using library matrix inversion routines as those found in e.g. Numerical Recipes [20]. We shall give an example of this procedure below.

2.3 The Quantum Regression Theorem

2.3.1 Motivation, Correlation Functions and Spectra

Omitting constants describing solid angles, detector efficiencies, and coupling strengths, the *Wiener-Khintchine theorem* gives for the spectrum emitted by a system coupled to the quantized electromagnetic field:

$$\begin{aligned} F(\omega) &\propto |E^{+}(\omega)|^2 \\ &\propto Re \int_0^{\infty} \langle \hat{E}^{-}(t)\hat{E}^{+}(t+\tau)\rangle \exp(i\omega\tau)d\tau \\ &\propto Re \int_0^{\infty} \langle \hat{d}^{+}(t)\hat{d}^{-}(t+\tau)\rangle \exp(i\omega\tau)d\tau, \end{aligned} \tag{21}$$

where $\hat{E}^{\pm}(t)$ are the positive/negative frequency parts of the quantized electric field, $E^{\pm}(\omega)$ their Fourier transforms, and $\hat{d}^{\pm}$ are the raising/lowering parts of the dipole operator for the system. The averages $\langle\cdot\rangle$ in Eq.(21) are assumed to be taken in the steady state of the system, i.e. $\langle \hat{d}^{+}(t)\hat{d}^{-}(t+\tau)\rangle$ is a function of τ, only. The formula involves the expectation value of a product of operators evaluated at two different times,

[4] In case of degenerate eigenvalues one further gets time dependent factors $\propto t^n e^{\eta t}$, where n is smaller than the multiplicity of the eigenvalue η. A pair of solutions $e^{\eta t}$, $te^{\eta t}$, with $\eta < 0$ is met for example in the case of critical damping of the classical harmonic oscillator.

not the product of expectation values. Such an operator product makes sense in the Heisenberg picture of quantum mechanics in which the operators are time-dependent and the wavefunction and the density matrix are constant, and it is the purpose of this section to review the method for calculating this kind of expectation values.

2.3.2 The Quantum Regression Theorem

We have introduced the density matrix as a catalogue of expectation values of a certain complete set of operators. The master equation, in the form (19), provides the time evolution of this catalogue, but it is not allowed simply to replace ρ_{ij} by $|j\rangle\langle i|$ in this set of equations in order to obtain the Heisenberg picture equations for all diadic products within the system S. In contrast to Eq.(14), the damping terms are now present, and in addition operator terms with vanishing mean values must be added on the RHS,

$$\frac{d}{dt}(|j\rangle\langle i|)(t) = \sum_{i'j'} M_{ij,i'j'}(|j'\rangle\langle i'|)(t) + F_{ji}(t). \tag{22}$$

Physically these terms are due to the interaction Hamiltonian and they appear as a sum of reservoir operators, cf. Eq.(16). They are assumed to have vanishing mean values, hence they do not appear in the density matrix equation (the expectation value of Eq.(22)), but they must be retained in the operator equations. Fortunately they will not play any role in the practical calculations.

To calculate two-time averages of the type $\langle A(t)B(t+\tau)\rangle$, we expand the operator $B(t+\tau) = \sum_{ji} B_{ji}(|j\rangle\langle i|)(t+\tau)$, and for A fixed we then need only the *catalogue of two-time expectation values*

$$\rho_{A,ij}(t+\tau) \equiv \Big\langle A(t)(|j\rangle\langle i|)(t+\tau)\Big\rangle. \tag{23}$$

The two-time expectation value of interest can then be written as

$$\begin{aligned}\langle A(t)B(t+\tau)\rangle &= \sum_{ij}\Big\langle A(t)B_{ji}(|j\rangle\langle i|)(t+\tau)\Big\rangle \\ &= \sum_{ij}\rho_{A,ij}(t+\tau)B_{ji} = Tr(\rho_A(t+\tau)B),\end{aligned} \tag{24}$$

where we treat $\rho_A(t+\tau)$ as a matrix, or an operator, the same way as we do with $\rho(t)$ in the Schrödinger picture.

Due to (22), we have a set of coupled equations,

$$\begin{aligned}\frac{d}{d\tau}\Big\langle A(t)(|j\rangle\langle i|)(t+\tau)\Big\rangle &= \Big\langle A(t)\frac{d}{d\tau}(|j\rangle\langle i|)(t+\tau)\Big\rangle \\ = \Big\langle A(t)\sum_{i'j'} M_{ij,i'j'}(|j'\rangle\langle i'|)(t+\tau)\Big\rangle &+ \Big\langle A(t)F_{ji}(t+\tau)\Big\rangle \\ = \sum_{i'j'} M_{ij,i'j'}\Big\langle A(t)|j'\rangle\langle i'|(t+\tau)\Big\rangle,\ (\tau \geq 0).\end{aligned} \tag{25}$$

The terms $\langle A(t)F_{ji}(t+\tau)$ vanish because $A(t)$ is a system operator and F is a reservoir operator with vanishing expectation value (see comments below), thus

$$\frac{d}{d\tau}\rho_{A,ij}(t+\tau) = \sum_{i'j'} M_{ij,i'j'}\rho_{A,i'j'}(t+\tau),\ (\tau \geq 0). \tag{26}$$

Eq.(26) shows that two-time expectation values can be determined from the knowledge of the matrix M alone, without an analysis of the noise terms in Eq.(22). This is the Quantum Regression Theorem [21] in a formulation which is very useful for numerical applications. The procedure may be applied also when a constant operator multiplies $(|j\rangle\langle i|)(t+\tau)$ from the right, and it readily generalizes to the case of multi-time correlation functions $\langle A(t_1)B(t_2)C(t_3)...\rangle$.

The set of equations (26) is the same as the Master Equation for the density matrix elements with a few exceptions: the initial values, for $\tau = 0$, are obtained as the expectation values $\left\langle A(t)(|j\rangle\langle i|)(t)\right\rangle = Tr(\rho A(|j\rangle\langle i|)) = \sum_k \rho_{ik}\langle k|A|j\rangle$, determined in many applications with the steady state Schrödinger picture density matrix. Since $\rho(t)$ and $\rho_A(t)$ fulfil the same equations of motion, the corresponding steady state matrices are proportional. The matrix $\rho_A(t)$ is observed to have trace equal to $\langle A(t)\rangle$, and since the trace is preserved during time evolution with the Master Equation, we deduce that

$$\tau \to \infty: \quad \rho_A(t+\tau) \to \langle A(t)\rangle \rho^{st}, \tag{27}$$

where ρ^{st} is the steady state solution to the Master Equation. This, in particular, implies the decorrelation of two-time averages at large time separations,

$$\tau \to \infty: \quad \langle A(t)B(t+\tau)\rangle \to \langle A(t)\rangle\langle B\rangle^{st}. \tag{28}$$

Warning: properties such as $\rho_{ij} = \rho^*_{ji}$, ρ_{ii} real, which may be useful in the numerical implementation of the master equation, do not apply to the elements of ρ_A. Apart from this the calculation of two-time expectation values is equivalent to the calculation of one-time averages, cf. Eq.(24).

Two comments should be made on the appearance of the F_{ji}-terms in (22). These "quantum noise" terms are necessary in order to preserve relations between products of operators such as commutators. In the product of two time-evolved operators the F_{ji}-terms appear twice at equal times and finite contributions may be obtained. This also implies that the disappearance of the F_{ji}-terms in Eq.(25), allowing us to refrain from going into a closer analysis of their values, is valid only for $\tau \geq 0$. In case of $\tau < 0$ the noise term $F_{ij}(t+\tau)$ may already have occured once in the time evolution of the operator $A(t)$ and the contribution from $A(t)F_{ji}(t+\tau)$ in Eq.(25) is now finite. For a more precise and very illustrative discussion of this, see e.g. Sec. $C_{IV}2$ of Ref.[15].

2.3.3 The QRT applied to integrals

From the values $\rho_{A,ij}(t+\tau)$ we can construct the two-time averages of interest, e.g. the one appearing in Eq.(21). Instead of considering the complete time dependent solution,

and then the Fourier transform we can obtain equations for the quantity of interest directly. It is, at this point, useful to introduce the difference between the operator A and its expectation value. The two-time average corresponding to $\langle A(t)\rangle\langle B(t+\tau)\rangle$ is just the product of two one-time averages. It is in the calculation of the second part, $\Big\langle (A(t)-\langle A(t)\rangle)B(t+\tau)\Big\rangle$ that we apply the Quantum Regression Theorem, and below we shall assume that $\rho_A(t+\tau)$ represents an operator product, where $\langle A\rangle = 0$.

Writing $\vec{\rho}_A$ for the vector of elements $\rho_{A,ij}$ and M for the matrix with elements $M_{ij,i'j'}$, we have from Eq.(26), $d\vec{\rho}_A/d\tau = M\vec{\rho}_A$, and therefore the identity

$$M\int_0^\infty \vec{\rho}_A(t+\tau)d\tau = \int_0^\infty \frac{d}{d\tau}\vec{\rho}_A(t+\tau)d\tau = \vec{\rho}_A(\infty) - \vec{\rho}_A(t) = -\vec{\rho}_A(t). \qquad (29)$$

As shown in Eq.(27) the term $\vec{\rho}_A(\infty)$ vanishes since $\langle A\rangle = 0$, and the integral of $\vec{\rho}_A(t+\tau)$ follows by a simple matrix inversion. [5]

To determine a Fourier transform as the one in Eq.(21) we note that

$$\begin{aligned}(M+i\Delta)\int_0^\infty \vec{\rho}_A(t+\tau)e^{i\Delta\tau}d\tau \\ = \int_0^\infty (e^{i\Delta\tau}\frac{d}{d\tau}\vec{\rho}_A(t+\tau) + i\Delta\vec{\rho}_A(t+\tau)e^{i\Delta\tau})d\tau \\ = -\vec{\rho}_A(t). \qquad (30)\end{aligned}$$

This set of equations is readily solved for the integral on the LHS like the one above.

If the two-time averages are multiplied by some low power of τ, as e.g. in the calculation of the friction coefficient in semiclassical laser cooling [22], this can also be treated in a manner similar to the examples above. Consider the equation, obtained by partial integration

$$M\int_0^\infty \tau\vec{\rho}_A(t+\tau)d\tau = \int_0^\infty \tau\frac{d}{d\tau}\vec{\rho}_A(t+\tau)d\tau = [\tau\vec{\rho}_A(t+\tau)]_0^\infty - \int_0^\infty \vec{\rho}_A(t+\tau)d\tau. \quad (31)$$

The two-time average converges exponentially towards zero for large τ, hence only the last term on the RHS is non-zero. This term was determined in Eq.(29) above, thus a new inversion procedure of the same matrix yields the integral on the LHS. For an application of these successive steps, see for example the discussion of friction and diffusion in laser cooling in Ref.[23].

2.3.4 The spectrum as a one-time average (I)

When motivating in Section 2.3.1 the need for two-time averages, we noted that these averages give *meaning* in the Heisenberg picture of quantum mechanics. This meaning

[5] As before the matrix M is not invertible, and we replace the (i_0i_0)'th row in M by numbers 0 and c, so that the c's multiply "population" elements $\rho_{A,i'i'}$, $M_{i_0i_0,i'j'} \to c\cdot\delta_{i'j'}$. We then get in the (i_0i_0)'th position of the product: $c\sum_{i'i'}\int_0^\infty \rho_{A,i'i'}(t+\tau) = c\sum_{i'i'}\int_0^\infty \langle A(t)(|i'\rangle\langle i'|)(t+\tau)\rangle = 0$, since the sum $\sum_{i'i'}(|i'\rangle\langle i'|)(t+\tau)$ equals the identity operator at all times, and the expectation value of A vanishes. Thus, in the inversion, we replace the (i_0i_0)'th element of the RHS vector by zero.

is purely formal; with the exception of Hanbury-Brown and Twiss type experiments where the time difference τ is due to a delay of the observation of the latter operator, a correlation function like the one in (21) does not give much physical meaning.

In practice e.g. the spectrum of fluorescence is obtained by shining the light into a frequency tunable Fabry-Perot resonator, and then to watch the intensity built up, that is *a one-time average !* At this point mathematics comes along, and since the rate of change of the field amplitude in the cavity is given by the coupling to the source, the value of the field at a time t can be written as an integral of the source dipole over all previous times t'. The rate of change of the intensity at time t therefore becomes a two-time average involving the dipole at time t and at all previous times [24]. [6]

Instead of the above approach we can look for the spectrum as a one-time average directly. Let the system S denote both the atom and a single mode of the quantized field at frequency ω. The field mode is described as an harmonic oscillator, so we have the extended set of density matrix elements

$$\rho_{ij}^{nm} \equiv (\langle n| \otimes \langle i|)|\rho|(|j\rangle \otimes |m\rangle), \quad n,m = 0,1,.. \ . \tag{32}$$

The coupling, proportional to the small solid angle of the detector, is very weak (otherwise the cavity field might act back on the atom, and we obtain a cavity QED problem), so we may truncate the set of equations at $n,m = 1$. Now, the value of the spectrum at frequency ω is the average of the field number operator, i.e. $\sum_i \rho_{ii}^{11}$, a one-time average calculated in the stationary state of the system. With increasing n or m the elements of ρ decrease by a factor proportional to the small coupling strength, and we may in the master equation neglect contributions from $\rho^{01/10}$ in the evolution of ρ^{00}, and of ρ^{11} in the evolution of $\rho^{01/10}$, but not the other way. This implies that we may solve the problem stepwise: an inversion of M in Eq.(19) gives ρ^{00}, and a subsequent inversion of $M \pm i\Delta$ gives $\rho^{01/10}$ ($\Delta = \omega - \omega_L$ appears due to the cavity field hamiltonian $H_F = \hbar(\omega_L + \Delta)a^+a$). One verifies that the trace of ρ^{11} can be expressed in terms of expectation values of the raising and lowering parts of the dipole operator taken with the density matrices $\rho^{10/01}$: the algebraic steps in this calculation are exactly the same as the ones applied in the QRT calculation.

2.4 Example: the laser excited two-level atom

2.4.1 Atom coupled to the quantized electromagnetic field

Let S represent a two-level atom (states $|g\rangle, |e\rangle$ with energy separation $\hbar\omega_A$) which is coupled, with Rabi frequency Ω, to a classical laser field with frequency $\omega = \omega_A + \delta$,

[6] Since the field inside the cavity evolves at the cavity resonance frequency ω this quantity also enters as an exponential factor in the integral of the source dipole as in (21), and the decay of the field amplitude due to the finite photon lifetime, γ_F^{-1} in the cavity, leads to an extra factor $e^{-\gamma_F \tau}$. This gives the detector its finite bandwidth, γ_F, and it can be accounted for by a subsequent convolution with a Lorentzian.

so that the Hamiltonian in the rotating wave approximation and in a frame rotating at the laser frequency reads:

$$H_S = \frac{1}{2}\hbar\delta(|g\rangle\langle g| - |e\rangle\langle e|) - \frac{1}{2}\hbar\Omega(|e\rangle\langle g| + |g\rangle\langle e|). \tag{33}$$

The reservoir R represents the quantized electromagnetic field,

$$H_R = \sum_\lambda \hbar\omega_\lambda(a_\lambda^\dagger a_\lambda + 1/2) \tag{34}$$

and we have the electric dipole coupling of the system and the reservoir

$$H_{SR} = \sum_\lambda \sqrt{\frac{\hbar\omega_\lambda}{\epsilon_0 V}}\ \vec{d}\cdot\vec{e}_\lambda(a_\lambda + a_\lambda^\dagger), \tag{35}$$

where ω_λ is the frequency of the mode λ, the different modes covering a broad range around the atomic transition frequency. V is the quantization volume, $\vec{e}_\lambda$ is the unit polarization vector of the mode λ and a_λ, $a_\lambda^\dagger$ are the corresponding annihilation and creation operators. $\vec{d}$ is the atomic dipole operator, $\vec{d} = \vec{D}(|e\rangle\langle g| + |g\rangle\langle e|)$, where $\vec{D}$ is a constant vector.

2.4.2 Master Equation

One verifies that the steps discussed in Eqs.(15) through (18) in general terms can be carried out in this explicit case. As a result of the calculation one also identifies the coefficients in the $\mathcal{L}_{\text{relax}}[\rho]$-terms. For the two-level atom coupled to the quantized field in its vacuum state there is only a single operator (and its conjugate) appearing in the Lindblad form (18), $C_i = \sqrt{\Gamma}|g\rangle\langle e|$:

$$\mathcal{L}_{\text{relax}}[\rho] = -\frac{\Gamma}{2}\{|e\rangle\langle e|\rho + \rho|e\rangle\langle e|\} + \Gamma|g\rangle\langle e|\rho|e\rangle\langle g|. \tag{36}$$

As in the case of Eq.(10), equations for the specific elements are obtained by taking the $\langle i|\ \cdot\ |j\rangle$ matrix elements on both sides of this expression. The anti-commutator causes the decay of the excited state population with a rate $2\cdot\Gamma/2$, and of the coherences, ρ_{eg}, ρ_{ge} with a rate $\Gamma/2$ in agreement with Eq.(13). The last term in Eq.(36) represents the feeding of the ground state. With the 2 by 2 density matrix represented as a vector with four elements $\vec{\rho} = (\rho_{gg}, \rho_{ge}, \rho_{eg}, \rho_{ee})^{tr}$, the matrix M reads

$$M = \begin{pmatrix} 0 & -i\Omega/2 & i\Omega/2 & \Gamma \\ -i\Omega/2 & -i\delta - \Gamma/2 & 0 & i\Omega/2 \\ i\Omega/2 & 0 & i\delta - \Gamma/2 & -i\Omega/2 \\ 0 & i\Omega/2 & -i\Omega/2 & -\Gamma \end{pmatrix}. \tag{37}$$

This matrix provides all information about the time evolution of the system.

The eigenvalues of M read: 0, $-\gamma$, $\pm i\Omega_R - \gamma'$, where γ, γ' are on the order of Γ, and where $\Omega_R \simeq \sqrt{\delta^2 + \Omega^2}$ ($\Omega_R = 0$ for Ω small compared to Γ). As expressed in Eq.(20),

the non-vanishing eigenvalues of M determine the transient evolution towards steady state, see also [16, 25], and they also determine the positions and widths of spectral features.

To determine the stationary operator ρ (in the rotating frame), we need to solve Eq.(19) with zero's on the LHS. The first and the last row in the matrix $\boldsymbol{M}$, corresponding to the ground and excited state populations, are linearly dependent and we replace the first row of M in Eq.(37) by (Γ 0 0 Γ), and the LHS vector is replaced by the column vector $(\Gamma, 0, 0, 0)^{tr}$. The two-level atom problem is of course not more complicated than it can be solved analytically, and the results for a slightly more general problem are presented:

The inverse of the matrix

$$M'^{-1}_{\Delta} = \begin{pmatrix} \Gamma & 0 & 0 & \Gamma \\ -i\Omega/2 & -i\delta - \Gamma/2 + i\Delta & 0 & i\Omega/2 \\ i\Omega/2 & 0 & i\delta - \Gamma/2 + i\Delta & -i\Omega/2 \\ 0 & i\Omega/2 & -i\Omega/2 & -\Gamma + i\Delta \end{pmatrix} \tag{38}$$

can be written as a polynomial in the extra variable Δ, introduced for later convenience,

$$M'^{-1}_{\Delta} = D^{-1}_{\Delta}(N_0 + \Delta N_1 + \Delta^2 N_2 + \Delta^3 N_3), \tag{39}$$

where the determinant reads,

$$D_{\Delta} = \delta^2\Gamma^2 + \Gamma^4/4 + \Gamma^2\Omega^2/2 \; - \; 2\Delta^2\Gamma^2 - i\Delta\Gamma(\delta^2 - \Delta^2 + \Omega^2 + 5\Gamma^2/4) \tag{40}$$

and where we have the four matrices

$$N_0 = \Gamma \begin{pmatrix} \delta^2 + \Omega^2/4 + \Gamma^2/4 & \delta\Omega/2 + i\Omega\Gamma/4 & \delta\Omega/2 - i\Omega\Gamma/4 & \delta^2 + \Gamma^2/4 \\ -\delta\Omega/2 - i\Gamma\Omega/4 & -\Gamma^2/2 - \Omega^2/2 + i\Gamma\delta & -\Omega^2/2 & -\delta\Omega - i\Omega\Gamma/2 \\ -\delta\Omega/2 + i\Gamma\Omega/4 & -\Omega^2/2 & -\Gamma^2/2 - \Omega^2/2 - i\Gamma\delta & -\delta\Omega + i\Omega\Gamma/2 \\ \Omega^2/4 & -\delta\Omega/2 - i\Omega\Gamma/4 & -\delta\Omega/2 + i\Omega\Gamma/4 & -\delta^2 - \Gamma^2/4 \end{pmatrix},$$

$$N_1 = \begin{pmatrix} -i(\delta^2 + 5\Gamma^2/4 + \Omega^2/2) & \Omega\Gamma/2 & -\Omega\Gamma/2 & -i\Gamma^2 \\ -3\Omega\Gamma/4 + i\delta\Omega/2 & \Gamma\delta + 3i\Gamma^2/2 & 0 & -\Gamma\Omega \\ 3\Omega\Gamma/4 + i\delta\Omega/2 & 0 & -\Gamma\delta + 3i\Gamma^2/2 & \Gamma\Omega \\ -i\Omega^2/2 & -\Omega\Gamma/2 & \Omega\Gamma/2 & i\Gamma^2 \end{pmatrix},$$

$$N_2 = \begin{pmatrix} -2\Gamma & 0 & 0 & -\Gamma \\ i\Omega/2 & \Gamma & 0 & 0 \\ -i\Omega/2 & 0 & \Gamma & 0 \\ 0 & 0 & 0 & \Gamma \end{pmatrix}, \quad N_3 = \begin{pmatrix} i & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}. \tag{41}$$

The steady state solution to the Master Equation is given by the elements in the first column of the matrix N_0 multiplied by $\Gamma/D_{\Delta=0}$, and from these values one can obtain e.g. the absorption spectrum, and the polarization of a medium of two-level atoms.

2.4.3 Spectrum of fluorescence

According to (21) we need to calculate

$$F(\omega) = Re \int_0^\infty \Big\langle (|e\rangle\langle g|)(t)(|g\rangle\langle e|)(t+\tau) \exp(-i\omega_L \tau) \Big\rangle \exp(i\omega\tau) d\tau, \qquad (42)$$

where the exponential factor at frequency ω_L takes care of the transformation from the rotating frame with the time independent equations of motion for the $|i\rangle\langle j|(t)$ to the original frame with the dipole oscillating at the laser frequency, $d^+(t) = |e\rangle\langle g|(t)e^{i\omega_L t}$.

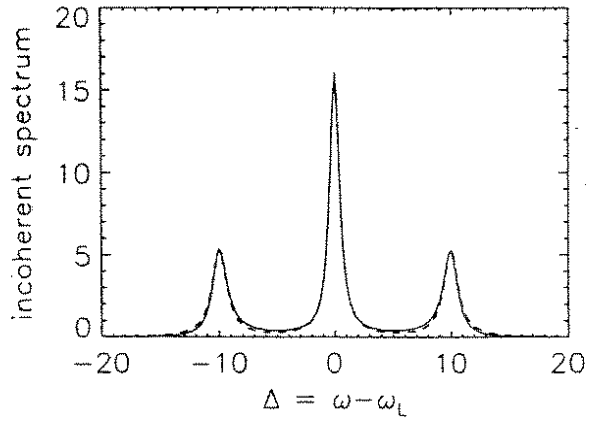

Figure 2: *Spectrum of fluorescence emitted by a laser excited two-level atom. The laser field is on resonance, $\delta = 0$ and $\Omega = 10\Gamma$. The solid curve is the exact result, and the dashed curve represents the approximation (46), (arbitrary units).*

We now decompose the expectation value in (42) into two terms, one yielding

$$F_{coh}(\omega) = Re \int_0^\infty \Big\langle (|e\rangle\langle g|)(t) \Big\rangle \Big\langle (|g\rangle\langle e|)(t+\tau) \Big\rangle \exp(i(\omega - \omega_L)\tau) d\tau = |\rho_{ge}|^2 \pi \delta(\omega - \omega_L). \qquad (43)$$

This is the coherent part of the spectrum; the steady state average dipole oscillating with the driving field emits monochromatic radiation at this frequency. We are now left with the incoherent term

$$F_{inc}(\omega) = Re \int_0^\infty \Big\langle (|e\rangle\langle g|)(t) - \rho_{ge})(|g\rangle\langle e|)(t+\tau) \Big\rangle \exp(i(\omega - \omega_L)\tau) d\tau, \qquad (44)$$

which is of the type considered above, with $A(t) = (|e\rangle\langle g|)(t) - \rho_{ge}$, and what we need is the real part of the (eg)-component of the integral in (30). The initial values, at time t, of the elements of $\vec{\rho}_A$ are determined from the steady state expectation values:

$$\begin{pmatrix} \rho_{A,gg}(t) \\ \rho_{A,ge}(t) \\ \rho_{A,eg}(t) \\ \rho_{A,ee}(t) \end{pmatrix} = \begin{pmatrix} \Big\langle (|e\rangle\langle g| - \rho_{ge})|g\rangle\langle g| \Big\rangle \\ \Big\langle (|e\rangle\langle g| - \rho_{ge})|e\rangle\langle g| \Big\rangle \\ \Big\langle (|e\rangle\langle g| - \rho_{ge})|g\rangle\langle e| \Big\rangle \\ \Big\langle (|e\rangle\langle g| - \rho_{ge})|e\rangle\langle e| \Big\rangle \end{pmatrix} = \begin{pmatrix} \rho_{ge} - \rho_{ge}\rho_{gg} \\ -\rho_{ge}\rho_{ge} \\ \rho_{ee} - \rho_{ge}\rho_{eg} \\ -\rho_{ge}\rho_{ee} \end{pmatrix}. \qquad (45)$$

Here, all steady state density matrix elements are obtained from the first column of the matrix N_0 in Eq.(41). The incoherent part of the spectrum at the frequency $w \equiv \omega_L + \Delta$ we obtain by letting the matrix M'^{-1} act on $-\vec{\rho}_A(t)$, given above (but with the first component replaced by zero), and by extracting the real part of the third element of the resulting vector. The analytical expression is quite complicated. For $\delta = 0$ and $\Omega \gg \Gamma$ it reduces to [26]

$$F(\omega_L + \Delta) \propto \frac{3}{(\Delta - \Omega)^2 + (3\Gamma/4)^2} + \frac{4}{\Delta^2 + \Gamma^2/4} + \frac{3}{(\Delta + \Omega)^2 + (3\Gamma/4)^2}, \qquad (46)$$

showing three Lorentzians located at the laser frequency ω_L, and at $\omega_L \pm \Omega$, see figure. This so-called Mollow-triplet [27] was predicted and observed a long time ago, but, still, theoretical and experimental analyses are carried out to examine various features of this spectrum [28, 29].