

Chapter 2

Resonance fluorescence

The light emission of a system that is excited by light is usually called “fluorescence”.¹ The case of a two-level atom driven by a laser provides a very simple, yet fundamental system in quantum optics. The fluorescence has an emission spectrum showing characteristic deviations from the radiation of a classical dipole. These deviations demonstrate that both the atom and the field are genuinely quantum-mechanical systems. Resonance fluorescence is therefore a phenomenon that became a cornerstone of quantum optics.

Introduction

The spectrum of *resonance fluorescence* describes the light scattered by a two-level system that is driven by a laser. The spectrum is sketched in Fig.2.1 and shows for a sufficiently large laser intensity three bands whose width in frequency is of the order of the atomic decay rate γ . In addition, there is an “elastic line” at the laser frequency whose width is only limited by the frequency width of the laser (in an ideal theory, a monochromatic field). The Mollow triplet (Fig.2.1 below) is an example of “inelastic” or “nonlinear scattering” because the light frequency is changed. We shall see later in this chapter how this effect can be interpreted. It is characteristic for the closed two-state Hilbert space of the two-level system; it would not

¹Depending on the order of magnitude of the radiative decay time, one also uses the names “luminescence” or “phosphorescence”.

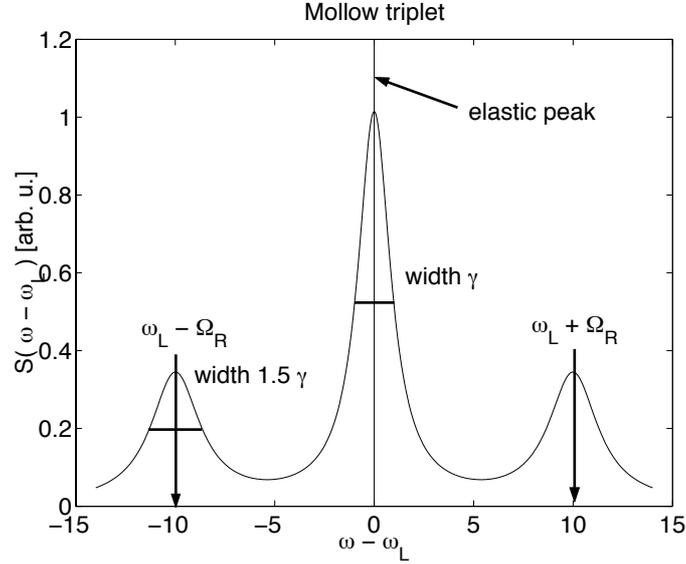


Figure 2.1: The emission spectrum of a driven two-level atom (the Mollow triplet). Parameters: detuning $\Delta = 0$, Rabi frequency $\Omega = 10\gamma$, zero temperature. The ratio between the peak heights is 1 : 3 : 1. The peak surfaces are in the ratio 1 : 2 : 1.

appear for a harmonic oscillator, be it quantized or damped, as long as it is linearly driven.

2.1 Spectrum and dipole correlation

The relevant correlation function for resonance fluorescence involves the electric field at the detector position. Now, since this field solves an inhomogeneous Maxwell equation with the atomic dipole operator (actually the current operator) as a source term, the field contains a component proportional to the dipole itself: in the frame rotating at the laser frequency, the current density is

$$\mathbf{j}(\mathbf{x}, t) = e^{-i\omega_L t} \mathbf{d}_{ge} \delta(\mathbf{x} - \mathbf{x}_A) \left(-i\omega_L + \frac{d}{dt} \right) \sigma(t) + \text{h.c.} \quad (2.1)$$

where in practice, the term $i\omega_L$ is much larger than the time derivative (remember the time scales in the master equation for the atom). The field

radiated by this oscillating dipole is (in the far field approximation)

$$\mathbf{E}(\mathbf{x}, t) = -\frac{\mathbf{d} - \hat{\mathbf{R}}(\hat{\mathbf{R}} \cdot \mathbf{d})}{4\pi\epsilon_0 c^2} e^{-i\omega_L(t-R/c)} \left(-i\omega_L + \frac{d}{dt}\right)^2 \frac{\sigma(t - R/c)}{R} + \text{h.c.} \quad (2.2)$$

where $R = |\mathbf{x} - \mathbf{x}_A|$ is the distance between atom and detector. This equation is the same as in classical electrodynamics. In fact it is just the far field of a Hertzian dipole: it is proportional to $1/R$, contains the projection of the dipole perpendicular to the observation direction, involves the acceleration of the dipole and, finally, depends only on the “retarded” dipole, a time interval R/c prior to observation.

For our purposes, we can assume that the atomic dipole oscillates approximately at the frequency ω_L of the driving laser, assuming that the time scales appearing from d/dt in Eq.(2.2) are much slower. We thus have

$$\left(-i\omega_L + \frac{d}{dt}\right)^2 \sigma(t - x/c) \approx -\omega_L^2 \sigma(t - x/c).$$

R. Glauber developed a theory of photodetection showing that observed signal involve so-called *normally ordered* correlation functions of the electric field (at the position of the detector). We split the electric field operator in Eq.(2.2) in a positive frequency part, $\mathbf{E}^+(t)$, the first term proportional to $e^{-i\omega_L t}$, and the negative frequent part $\mathbf{E}^-(t)$ which is the hermitean conjugate of the former. According to Glauber, we need the following normally ordered autocorrelation function of the electric field,

$$\lim_{t \rightarrow \infty} \langle \mathbf{E}^-(\mathbf{x}, t + \tau) \cdot \mathbf{E}^+(\mathbf{x}, t) \rangle \propto C_\sigma(\tau) = \lim_{t \rightarrow \infty} \langle \sigma^\dagger(t + \tau) \sigma(t) \rangle e^{i\omega_L \tau} \quad (2.3)$$

The proportionality factor is simply ignored in many treatments. Note that in the stationary limit $t \rightarrow \infty$, the delay time R/c drops out from the autocorrelation function.

The spectrum of the resonance fluorescence is thus related to the normally ordered dipole autocorrelation, and we define

$$S_d(\omega - \omega_L) = \lim_{t \rightarrow \infty} \int d\tau \langle \sigma^\dagger(t + \tau) \sigma(t) \rangle e^{-i(\omega - \omega_L)\tau} \quad (2.4)$$

The exponential $e^{i\omega_L \tau}$ appears due to the transformation frame into the frame rotating at the laser frequency ω_L . Due to this factor, the main weight of the spectrum is located near the laser frequency.

We shall suppose that the atom has reached a stationary state determined by the laser field and its radiative decay. The correlation function then only depends on the time difference τ . Using the adjoint, it can be shown that Eq.(2.4) becomes

$$S(\omega - \omega_L) = 2 \operatorname{Re} \int_0^{+\infty} d\tau e^{-i(\omega - \omega_L)\tau} \langle \sigma^\dagger(t + \tau) \sigma(t) \rangle,$$

so that we only have to deal with the case $\tau > 0$. This is the spectrum that we shall calculate in the following. Note that it only depends on the difference frequency $\omega - \omega_L$.

Example: free atom, stable. For a free atom, ignoring spontaneous decay, we have $\sigma(t) = \sigma e^{i\Delta t}$ in the rotating frame with $\Delta = \omega_L - \omega_A$ the detuning (Paris convention). Then a result similar to Eq.(3.14) emerges:

$$S_\sigma(\omega) = \langle \sigma^\dagger \sigma \rangle \delta(\omega - \omega_A) \quad (2.5)$$

where the prefactor is just the probability of finding the atom in the excited state. Hence, an atom in the ground state does not radiate (as could have been expected, consistent with the theory of photodetection). The spectrum (2.5) is of course a too crude approximation because it ignores the natural linewidth of the atomic transition.

Intensity correlations

Before starting the calculation of the dipole spectrum, we mention *intensity correlations* as another relevant correlation function. In the context of resonance fluorescence, they give an experimental check that one deals with a *single* emitter, instead of large number of molecules. The last case applies to natural light, for example, and has been studied by Hanbury Brown & Twiss (1956). Following these experiments, a number of subtle issues on multi-photon interference have been discussed and this contributed to the birth of quantum optics.

The observable in this context is the joint photocount rate of observing a photon at time $t + \tau$ after a first one at time t . According to Glauber's theory of photodetection, this rate is given by the autocorrelation function of the intensity

$$C_I(\tau) = \langle : I(t + \tau) I(t) : \rangle = \langle E^\dagger(t) E^\dagger(t + \tau) E(t + \tau) E(t) \rangle \quad (2.6)$$

where the symbols $:\cdots:$ requires the field operators in normal (and time) order, as indicated by the second expression. The limit $t \rightarrow \infty$ is implicit here. For a single two-level system, we therefore need the correlation function

$$G_2(\tau) = \langle \sigma^\dagger(t) \sigma^\dagger(t + \tau) \sigma(t + \tau) \sigma(t) \rangle \quad (2.7)$$

Let us take the time t as starting point for the density operator $\rho(t)$ and assume for simplicity that the Heisenberg operators like $\sigma^\dagger(t + \tau) \sigma(t + \tau)$ can be evolved with a unitary operator $U(\tau)$. (This is certainly only approximately true for an open system, see below.) Then we have $\sigma(t) = |g\rangle\langle e|$ as the Schrödinger and Heisenberg pictures coincide at this time. The expectation value can be written explicitly

$$\begin{aligned} G_2(\tau) &= \text{tr} \left[|e\rangle\langle g| U^\dagger(\tau) |e\rangle\langle e| U(\tau) |g\rangle\langle e| \rho(t) \right] \\ &= |\langle e| U(\tau) |g\rangle|^2 \rho_{ee}(t) \end{aligned} \quad (2.8)$$

Here, we see that the intensity correlations are proportional to the occupation $\rho_{ee}(t) = p_e(t)$ of the excited state, on the one hand (second factor), and to the probability $|\langle e| U(\tau) |g\rangle|^2$ of finding the atom in the excited state a time τ after it has been “prepared” in the ground state, on the other. Note that this quantity must be zero for $\tau \rightarrow 0$: it takes some time for the laser field to pump the atom into the excited state again. This example illustrates that the “standard interpretation” of correlation functions (see below) can be dangerous in the quantum context because a measurement always perturbs the system. In our case, we can say that the detection of the first photon at time t projects the atom onto the ground state. There it must be because the photon has been released. For the second photon to be emitted, the atom must again be brought to the excited state. This effect is called “anti-bunching” and it is in fact a genuine quantum-mechanical feature of a light-emitting two-level system.

2.2 Quantum regression formula

For the time being, we have worked with Heisenberg operators. Since the atom is an open system, the average in the dipole correlation function can

be understood as

$$\begin{aligned}\langle \sigma^\dagger(t + \tau)\sigma(t) \rangle &= \text{tr}_{SB} \left[U^\dagger(\tau)\sigma^\dagger(t)U(\tau)\sigma(t)P_{SB}(t) \right] \\ &= \text{tr}_S \left[\sigma^\dagger(t) \text{tr}_B \left(U(\tau)\sigma(t)P_{SB}(t)U^\dagger(\tau) \right) \right]\end{aligned}\quad (2.9)$$

where $U(\tau)$ is the complete unitary time evolution of system (atom) and bath (field) and $P_{SB}(t)$ the atom+field density operator at time t .

The expression involving the field trace is a reduced atomic operator that resembles the reduced atomic density matrix. Indeed, the latter can be written

$$\rho(t + \tau) = \text{tr}_B \left(U(\tau)P_{SB}(t)U^\dagger(\tau) \right) \quad (2.10)$$

with the same unitary operator. We now make the hypothesis that at the late time t , the atom+field system density matrix factorizes (as we assumed at time t in the derivation of the master equation),

$$P_{SB}(t) \approx \rho_{\text{st}} \otimes \rho_B \quad (2.11)$$

where ρ_{st} is the stationary density matrix for the atom and ρ_B an equilibrium state for the bath (field). Recall that this is actually an approximation based on the Markov assumption: correlations between atom and field decay rapidly.

Comparing (2.9) and (2.10), we observe that both the density matrix and the atomic operator

$$\varrho_\sigma(\tau) \equiv \text{tr}_B \left(U(\tau)\sigma(t)P_{SB}(t)U^\dagger(\tau) \right) \quad (2.12)$$

are determined by the same Nakajima-Zwanziger map (Sec.??): blow up at time t to a density operator on the system+bath space, evolve with the complete time evolution and take the trace. We can therefore *use the same master equation* that we derived for ρ_A to compute the time-dependence of $\varrho_\sigma(\tau)$. This statement is the “quantum regression formula”.² The only difference is the initial state that involves an additional spin operator

$$\varrho_\sigma(0) \equiv \sigma\rho_{\text{st}}. \quad (2.13)$$

²We shall use the word formula and not “theorem” because there are people who insist that it is based on an approximation and hence not a real theorem.

We have chosen to use the Schrödinger picture at (the anyway arbitrary) time t . According to the quantum regression theorem, the equation of motion for the ϱ_σ operator is

$$\frac{d}{d\tau}\varrho_\sigma = \frac{1}{i\hbar} [H_A, \varrho_\sigma] + \mathcal{L}[\varrho_\sigma]. \quad (2.14)$$

where we have split the generator of the master equation into a Hamiltonian and a Lindblad ‘dissipator’.

2.3 Eigenvalues of the master equation

The explicit solution of (2.14) involves complicated algebraic manipulations, and we shall give only a sketch of the most important techniques and results.

One of the main ideas is to write $\varrho_\sigma(\tau)$ as a sum of eigenfunctions of the master equation. Each of these functions evolves in time with an exponential $e^{-\lambda t}$. Each eigenvalue λ (for positive real part) gives a contribution to the spectrum that is a Lorentzian peak:

$$\int_0^{+\infty} d\tau e^{-i(\omega-\omega_L)\tau} e^{-\lambda t} = \frac{-i}{\omega - \omega_L - i\lambda}$$

The real part of lambda thus gives the width of the corresponding peak, while $\text{Im } \lambda$ gives the frequency shift with respect to the laser frequency. Since the stationary density matrix is reached at long times, we can conclude that all eigenvalues of the master equation must have positive real part (the density matrix cannot explode exponentially).

We can immediately state that $\lambda = 0$ is one eigenvalue of the master equation. This is because both Hamiltonian and Liouvillian conserve the trace of the density matrix (as they should if we want to maintain a probability interpretation of ρ). It is easy to see that the corresponding eigenfunction is the stationary density matrix ρ_{st} (simply because its time derivative vanishes by construction). For the ϱ_σ operator, we thus also find an eigenvalue $\lambda = 0$. This corresponds to a peak in the emission spectrum centred at the laser frequency with zero width — hence a δ function in frequency. The atomic fluorescence thus contains a spectral contribution at precisely the frequency of the laser — the “elastically scattered light”. This

contribution also occurs for a classical dipole when it oscillates in phase with the external field (once the initial transients have died out) and represents the “classical” part of the fluorescence spectrum (atom = classical dipole, photons = classical field).

For the ϱ_σ operator, the eigenfunction corresponding to the elastic emission is also proportional to the stationary density matrix. We can fix the proportionality factor by computing the trace (that must be equal to the initial trace):

$$\varrho_\sigma^{(\text{st})} = \rho_{\text{st}} \text{tr} \varrho_\sigma(0) = \rho_{\text{st}} \text{tr} (\sigma \rho_{\text{st}}) = \rho_{\text{st}} \rho_{eg}^{(\text{st})}$$

The elastic spectrum is thus proportional to the square of the off-diagonal density matrix element:

$$\begin{aligned} S(\omega - \omega_L) &= 2 \text{Re} \text{tr} \left(\sigma^\dagger \rho_{\text{st}} \right) \rho_{eg}^{(\text{st})} \frac{-i}{\omega - \omega_L - i0} \\ &= 2 \text{Im} \frac{|\rho_{eg}^{(\text{st})}|^2}{\omega - \omega_L - i0} \\ &= 2\pi \delta(\omega - \omega_L) |\rho_{eg}^{(\text{st})}|^2. \end{aligned}$$

This spectrum is radiated by the “average” dipole that, in the stationary state, oscillates at the same frequency as the external field. We recall that the average dipole is

$$\langle \mathbf{d} \rangle \propto \langle \sigma(t) + \sigma^\dagger(t) \rangle = \rho_{eg}^{(\text{st})} e^{-i\omega_L t} + \text{c.c.}$$

Herbert Walther, one of the founders of quantum optics in Germany, used to say that the elastic peak in the fluorescence spectrum is a proof that ‘light is a classical wave’ – he probably meant that this can be explained by a classical, monochromatic dipole moment that oscillates at exactly the laser frequency.

What about the other eigenvalues of the master equation? If we expand the ϱ_σ operator in terms of the Pauli matrices (and a term proportional to the identity matrix), the master equation reduces to the Bloch equations with the 3×3 matrix (for zero temperature)

$$\begin{pmatrix} -\gamma/2 & \Delta & 0 \\ -\Delta & -\gamma/2 & -\Omega \\ 0 & \Omega & -\gamma \end{pmatrix}. \quad (2.15)$$

The eigenvalues of this matrix are the solutions of the cubic equation

$$\left(\frac{1}{2}\gamma - \lambda\right)^2 (\gamma - \lambda) + \Omega^2 \left(\frac{1}{2}\gamma - \lambda\right) + \Delta^2 (\gamma - \lambda) = 0 \quad (2.16)$$

Since it is cubic, this equation must have at least one real root, say λ_3 . Since its imaginary part is zero, we thus find another spectral component centred at the laser frequency. This one has a finite width, however. Putting $\Omega = 0$ (extremely weak driving), we find $\lambda_3 = \gamma$, so that this peak has a width given by the decay rate – this explains the fact that the spontaneous decay rate γ_e is identical to the “linewidth” of the atomic transition (at least for the present model; other decoherence mechanisms may broaden the linewidth even without changing the spontaneous decay rate).

The other two roots $\lambda_{1,2}$ are complex and conjugates of each other (because the polynomial has real coefficients). In the limit $\Omega, \Delta \gg \gamma$, we find that $|\lambda_{1,2}| \gg \gamma$, and keeping only the leading terms, we get

$$\begin{aligned} \lambda_{1,2} &\approx \pm i\Omega_R + \frac{3}{4}\gamma \left(1 - \frac{\Delta^2}{3\Omega_R^2}\right) \\ \lambda_3 &\approx \frac{1}{2}\gamma \left(1 + \frac{\Delta^2}{\Omega_R^2}\right) \end{aligned}$$

where $\Omega_R = \sqrt{\Omega^2 + \Delta^2}$ is the generalized Rabi frequency. In this “strong driving” limit, the spectrum thus contains two additional side peaks, displaced by the generalized Rabi frequency from the laser frequency. This spectral structure is called the “Mollow triplet”. It is shown schematically in Fig. 2.1. The heights of the peaks can be obtained from a more detailed analysis, and is discussed in the exercises.

Example. A more complete picture for the emission spectrum is shown in Fig.2.2. This calculation is made for an inhomogeneous spectral density for the photon bath. Some approximations must be done to get a master equation that can be solved with Laplace techniques. These approximations fail, however, to preserve the positivity of the spectrum for some parameter range. More details on this problem can be found in the paper Boedeker & Henkel (2012).

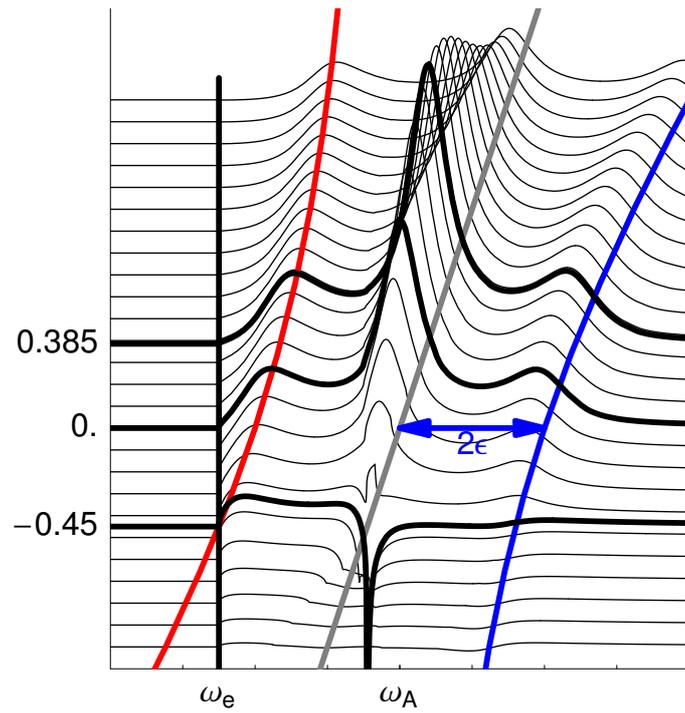


Figure 2.2: Family of spectra for different detunings $\Delta = \omega_L - \omega_A$, marked by the numbers on the left (in units of the Rabi frequency $\Omega = 2\epsilon$). The calculation is made for a photon bath with an inhomogeneous spectral density: there are no available modes for $\omega \leq \omega_e$ (thick vertical line). The red, gray, and blue lines trace the spectrum at the positions of three peaks of the Mollow spectrum.

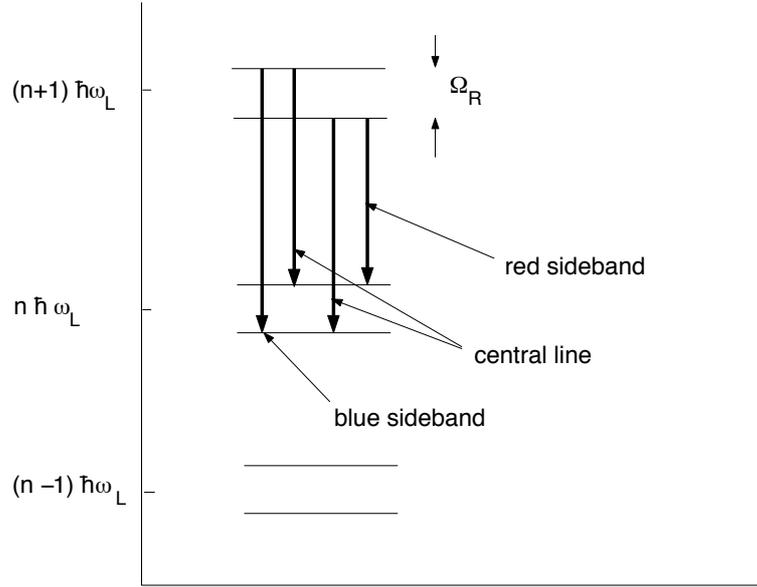


Figure 2.3: Interpretation of the Mollow triplet in terms of transitions between dressed states.

2.4 Interpretation of the Mollow triplet

The peaks in the Mollow spectrum can be interpreted in terms of transitions between the dressed states of the Jaynes Cummings Paul model, following the analysis of Cohen-Tannoudji (1977); Dalibard & Cohen-Tannoudji (1985). Let us focus on zero detuning for simplicity. We have seen previously that the dressed states have energies

$$E_{n\pm} \approx \text{const.} + n\hbar\omega_L \pm \frac{\hbar}{2}\Omega_R$$

where we have neglected the dependence of the Rabi frequency on the photon number (this is a good approximation for a classical laser field with many photons). The emission at the laser frequency comes from transitions

$$|n, +\rangle \rightarrow |n-1, +\rangle \quad \text{and} \quad |n, -\rangle \rightarrow |n-1, -\rangle,$$

as shown schematically in Fig. 2.3. Since both dressed states $|n\pm\rangle$ contain the excited state, they can indeed decay to lower states, thus converting one laser photon into a fluorescence photon.

The transitions on the sidebands $\omega_L \pm \Omega_R$ occur when the atom changes from a state $|n+\rangle$ to $|n-1, -\rangle$ or the other way round. Here, the fluorescence photon is shifted because of the splitting between the dressed states. For the linewidths of the transitions, one has to come back to a calculation like the one we sketched before.

As an alternative interpretation, one can look at the Rabi oscillations the atom performs in a laser field: because the atom flops at the Rabi frequency Ω_R between the ground and excited state, its emission is modulated (amplitude modulation). Therefore, the emission spectrum contains, in addition to the carrier at the laser frequency (as expected from the stationary state) sidebands at $\omega_L \pm \Omega_R$.

2.5 Calculation of Spontaneous Decay

2.5.1 Idea

Many systems in quantum optics are “open” and follow equations of motion that go beyond the Schrödinger equation. This applies to systems for which the Hamiltonian is not completely known or systems in contact with an “environment”. The equations of motion can be formulated as so-called master equations (similar to the Schrödinger picture, dynamics for the system density operator) or as so-called Langevin equations (similar to the Heisenberg picture, dynamics of the system operators). These equations are the quantum analogue of kinetic theories that describe the dynamics of a system including different dissipative effects. They provide, among others, the approach of the system towards thermal equilibrium, but also non-equilibrium states that are driven by external forces, the typical example being a laser field.

In quantum optics, the “system” can be an atom, a collection of atoms, or a field mode in a cavity. The system is “open” because it interacts with the “environment” (rest of the world), that is modelled as a continuum of quantized field modes. An atom decays irreversibly by emitting a photon into previously empty vacuum modes. This phenomenon of “spontaneous emission” does not allow for a description in terms of a Hamiltonian. In fact, the state of the atom does not remain pure, and entropy increases

because the photon can be emitted into any direction of space. A cavity mode decays because photons escape through the cavity mirrors, effectively becoming excitations of the “modes outside the cavity”. Alternatively, the field energy can be absorbed by the material making up the mirrors.

We focus first on the derivation of a master equation for a two-level atom. We shall find from the general theory two results: (1) the rate of spontaneous emission of an electronically excited state and (2) Glauber’s formula for the signal of a photodetectors in terms of normally ordered correlation functions of the electric field operator.

Time scales

The derivation builds on the following hierarchy of time scales:

$$\frac{2\pi}{\omega_A} \leq \tau_c \ll \frac{2\pi}{\Omega} \sim \frac{1}{\gamma} \quad (2.17)$$

where the shortest time scale is the optical period of the light field that is near-resonant with the atomic transition frequency ω_A . This is typically smaller than a few fs (femtoseconds) (in the visible range).

The “correlation time” τ_c of the electromagnetic field describes qualitatively the time interval over which the quantum fluctuations of the electromagnetic field (that couple significantly to the atom) are “smooth”. Two electric field measurements are not significantly “similar” if they are taken at intervals larger than τ_c . We give an estimate below, it turns out to be a few times larger than the optical period.

The third time scale is the Rabi period that scales with the inverse of the Rabi frequency of a laser. This depends of course on the laser power (and the atomic transition dipole), but typical values are in the 1–10 ns (nanoseconds) range. Note that this is much longer than the optical period and the field correlation time.

The last time scale is the lifetime of the excited state of the atom, inversely proportional to the spontaneous decay rate γ . This is typically in the 1–10 ns range as well, depending on the atom.

We thus have a situation similar to “Brownian motion”: the atom is “shaken around” by the vacuum field fluctuations that vary much faster than the typical dynamics for the atomic state (Rabi oscillations, spontaneous decay). The master equation takes advantage of this separation of

time scales to find an equation of motion that can be applied on “slow time scales”. As a result, one gets a single equation that describes both, the “Hamiltonian” effects of a laser (Rabi oscillations) and the “dissipative / friction” effects due to vacuum fluctuations.

Hamiltonian

Two-level atom, field, and electric dipole interaction:

$$H = \frac{\hbar\omega_A}{2}\sigma_3 + \sum_k \hbar\omega_k a_k^\dagger a_k + \sum_k \hbar \left(g_k^* a_k^\dagger \sigma + g_k \sigma^\dagger a_k \right) \quad (2.18)$$

neglect the zero-point energy of the field. Coupling constant

$$\hbar g_k = -\mathbf{d}_{\text{eg}} \cdot \mathbf{f}_k(\mathbf{x}_A) (\hbar\omega_k / 2\varepsilon_0)^{1/2} \quad (2.19)$$

with the mode function $\mathbf{f}_k(\mathbf{x})$ evaluated at the position of the atom. For a single-mode field, this coupling constant is sometimes called the ‘vacuum Rabi frequency’.

In the following, we also use the (re-scaled) electric field operator

$$E(t) = \sum_k g_k a_k(t) + \text{h.c.} \quad (2.20)$$

Field correlation time

To get an estimate of the field correlation time, we compute the two-time correlation (or coherence) function

$$C(\tau) = \langle E(t + \tau) E(t) \rangle \quad (2.21)$$

If we use the coupling constants g_k to write the mode expansion of the quantized field (this is equivalent to re-scaling the electric field so that it has units of frequency), we get

$$\begin{aligned} \langle E(t + \tau) E(t) \rangle &= \sum_k |g_k|^2 \langle a_k(t + \tau) a_k^\dagger(t) \rangle \\ &= \sum_k |g_k|^2 e^{-i\omega_k \tau} \end{aligned} \quad (2.22)$$

An explicit calculation using the mode amplitudes in free space leads to an integral of the form

$$C(\tau) \sim \int_0^{\infty} d\omega \omega^3 e^{-i\omega_k \tau} \quad (2.23)$$

which looks like a third derivative of a δ -function in τ . Well, not exactly since the integration starts at $\omega = 0$. Generalizing to finite temperature, the Fourier transform of the correlation function (2.23) becomes

$$C(\omega) \sim \omega^3 \bar{n}(\omega) = \frac{\omega^3}{\exp(\hbar\omega/k_B T) - 1} \quad (2.24)$$

The sign convention of the Fourier transform is chosen here such that $\omega > 0$ corresponds to photon numbers $a^\dagger a$ ('normal order'), while at $\omega < 0$, the anti-normal order aa^\dagger is picked. See Fig.2.4: the spectrum is proportional to $-\omega^3$ for negative frequencies, $-\omega \gg k_B T/\hbar$. Near zero frequency, it has a quadratic behaviour, at positive frequencies, a maximum near $\omega = 2k_B T/\hbar$ (Wien displacement law) and an exponential decay for $\omega \gg k_B T/\hbar$.

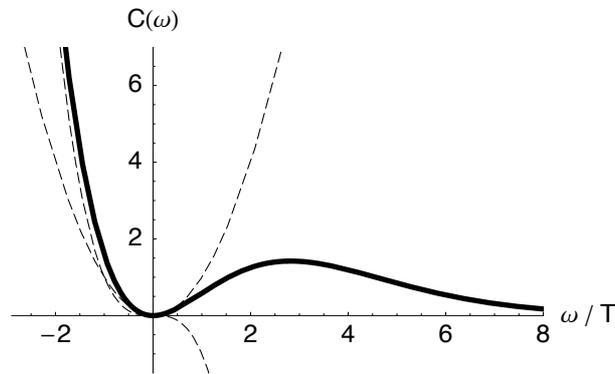


Figure 2.4: Spectrum $C(\omega)$, Eq.(2.24), of the radiation field (normal order) at finite temperature. The frequency is plotted in units of $k_B T/\hbar$. The dashed lines are ω^2 and $-\omega^3$.

Actually, we forgot one thing in this estimate: the Hamiltonian we start with is only valid for field modes k near-resonant with the atomic transition, $\omega_k \sim \omega_A$. It thus seems reasonable to restrict the frequency integral in Eq.(2.23) to an interval around ω_A , with a width $\Delta\omega$ that is typically smaller than ω_A .

Now, from the properties of the Fourier transformation, we know that the correlation function $C(\tau)$ given by the integral (2.23) has a “width in time” given by $\Delta\tau \Delta\omega \sim 1$. Since this width is precisely the definition of the correlation time, we have

$$\frac{1}{\omega_A} \leq \frac{1}{\Delta\omega} \sim \tau_c. \quad (2.25)$$

Typical value: a few 10 fs.

2.5.2 Evolution of the atomic operators

We shall work in the Heisenberg picture. Note that operators describing different degrees of freedom (field, atom) commute at equal times.

Atomic dipole

“eliminate” the field operators by solving their equation of motion:

$$\frac{da_k}{dt} = \frac{i}{\hbar} [H, a_k] = -i\omega_k a_k - ig_k^* \sigma \quad (2.26)$$

$$a_k(t) = a_k(0) e^{-i\omega_k t} - ig_k^* \int_0^t dt' \sigma(t') e^{-i\omega_k(t-t')} \quad (2.27)$$

the “particular solution” contains the “past” of the atomic dipole operator $\sigma(t')$. Insert this into the equation for σ . Let’s look at this equation first:

$$\begin{aligned} \frac{d\sigma}{dt} &= \frac{i}{\hbar} [H, \sigma] = -i\omega_A \sigma + i \sum_k g_k [\sigma^\dagger, \sigma] a_k \\ &= -i\omega_A \sigma + i \sum_k g_k \sigma_3 a_k \end{aligned} \quad (2.28)$$

We see here that the two-level atom leads to nonlinear equations of motion: the operator product $\sigma_3 a_k$ appears. For the moment, these operators commute (at equal times). But we now want to insert the solution (2.27) for $a_k(t)$, and the two terms that appear here do not commute separately with σ_3 . For this reason, we take now a specific operator order (so-called “normal order”) where the annihilation operators (a_k or σ) act first. This is the order already used in Eq.(2.28), and we thus get

$$\frac{d\sigma}{dt} = -i\omega_A \sigma + i\sigma_3 \mathcal{E}_0(t) + \int_0^t dt' \sum_k |g_k|^2 e^{-i\omega_k(t-t')} \sigma_3(t) \sigma(t') \quad (2.29)$$

where we used the abbreviation

$$\mathcal{E}_0(t) = \sum_k g_k a_k(0) e^{-i\omega_k t} \quad (2.30)$$

for the freely evolving electric field operator (positive frequency component only, and re-scaled by the atomic transition dipole). In the integral over t' , we see that the correlation function $C(t - t')$ appears, and using $\tau = t - t'$ as integration variable, we have

$$\frac{d\sigma}{dt} = -i\omega_A \sigma + i\sigma_3 \mathcal{E}_0(t) + \int_0^t d\tau C(\tau) \sigma_3(t) \sigma(t - \tau) \quad (2.31)$$

Now comes the *key observation*: under the time integral occur two very different functions. The correlation function $C(\tau)$ is very narrow in τ . The atomic operator $\sigma(t - \tau)$ contains a “fast free evolution” (generated by the first term in its equation of motion), but once this is factored out, we anticipate that its evolution is “slow”:

$$\sigma(t - \tau) = e^{-i\omega_A(t-\tau)} \tilde{\sigma}(t - \tau) \approx e^{-i\omega_A(t-\tau)} \tilde{\sigma}(t) = e^{+i\omega_A \tau} \sigma(t) \quad (2.32)$$

Here, we introduced temporarily the dipole operator $\tilde{\sigma}(t')$ (in an interaction picture).

The main benefit of this approximation is that only atomic operators at time t appear in the equation of motion. The two-level commutation rules³ give $\sigma_3(t)\sigma(t) = -\sigma(t)$, and we end up with

$$\frac{d\sigma}{dt} = -i\omega_A \sigma + i\sigma_3 \mathcal{E}_0(t) - \sigma(t) \int_0^t d\tau C(\tau) e^{i\omega_A \tau} \quad (2.33)$$

The upper integration limit t is actually the difference between our initial time and the actual time where the equation of motion is computed. We now let this time difference be much larger than the correlation time τ_c . This is consistent with the assumption that the atomic dynamics is slow on the scale of the field’s correlation time. Then the integrand is effectively

³A pedantic remark: operator products evolve as products under the Heisenberg equations of motion. This is because taking the commutator with a product is compatible with the product rule of (time) derivatives.

zero at the upper limit, and we get a constant complex number

$$\gamma + i\delta\omega_A = \int_0^{t \gg \tau_c} d\tau C(\tau) e^{i\omega_A \tau} = \frac{S(\omega_A)}{2} + i\mathcal{P} \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \frac{S(\omega)}{\omega - \omega_A} \quad (2.34)$$

where $S(\omega)$ is the Fourier transform of the correlation function $C(\tau)$ and \mathcal{P} means the principal part of the integral. Putting this complex number into the equation of motion (2.33):

$$\frac{d\sigma}{dt} = -i(\omega_A + \delta\omega_A)\sigma(t) - \gamma\sigma(t) + i\sigma_3\mathcal{E}_0(t), \quad (2.35)$$

we see that the real part γ leads to an exponential decay of the dipole operator $\langle\sigma\rangle$: this is called spontaneous decay (or natural linewidth). While $\delta\omega_A$ gives a shift of the atomic frequency, called the Lamb shift.

Spontaneous decay rate and Lamb shift

Explicit calculation: spectrum of vacuum fluctuations

$$S(\omega) = 2\pi \sum_k |g_k|^2 \delta(\omega_k - \omega) \quad (2.36)$$

Take a plane wave expansion and sum over the polarization vectors $\mathbf{u}_{\mathbf{k}\lambda}$ in the coupling constants g_k

$$\sum_{\lambda} |\mathbf{d}_{\text{eg}} \cdot \mathbf{u}_{\mathbf{k}\lambda}|^2 = |\mathbf{d}_{\text{eg}}|^2 - |\mathbf{d}_{\text{eg}} \cdot \hat{\mathbf{k}}|^2 \quad (2.37)$$

where $\hat{\mathbf{k}}$ is the unit vector along \mathbf{k} . This formula arises because the $\mathbf{u}_{\mathbf{k}\lambda}$ are perpendicular to \mathbf{k} . Integration over the angles of \mathbf{k} gives

$$\int d\Omega(\hat{\mathbf{k}}) (|\mathbf{d}_{\text{eg}}|^2 - |\mathbf{d}_{\text{eg}} \cdot \hat{\mathbf{k}}|^2) = 4\pi |\mathbf{d}_{\text{eg}}|^2 - \frac{4\pi}{3} |\mathbf{d}_{\text{eg}}|^2 = \frac{8\pi}{3} |\mathbf{d}_{\text{eg}}|^2 \quad (2.38)$$

The integral over the length of \mathbf{k} is trivial because of the δ -function in Eq.(4.3), its length is fixed to $|\mathbf{k}| = \omega/c$. Putting everything together, we get

$$S(\omega) = \frac{2\pi}{3\hbar^2} |\mathbf{d}_{\text{ge}}|^2 \frac{\hbar\omega}{2\epsilon_0} \frac{8\pi\omega^2}{(2\pi c)^3} \quad (2.39)$$

where the last factor is the density of field modes per $d\omega$ and volume. We can thus say that the spontaneously decaying atom is a “detector” for vacuum field fluctuations. The decay rate γ

$$\gamma = \frac{1}{2}S(\omega_A) = \frac{|\mathbf{d}_{ge}|^2(\omega_A/c)^3}{6\pi\hbar\epsilon_0} \quad (2.40)$$

is also called the “natural linewidth” of the atomic transition $|g\rangle \leftrightarrow |e\rangle$ because it gives the width in frequency of the spontaneous emission spectrum. In order of magnitude, with $|\mathbf{d}_{ge}| \sim ea_0$ (electron charge \times atom size)

$$\frac{\gamma}{\omega_A} \sim \alpha_{\text{fs}}(a_0/\lambda_A)^2 \sim \alpha_{\text{fs}}^3 \quad (2.41)$$

with fine structure constant $\alpha_{\text{fs}} = e^2/4\pi\epsilon_0\hbar c \approx 1/137.04$ and wavelength λ_A of resonant transition. Hence, indeed decay is “slow” on the scale of the optical period.

Frequency shift $\delta\omega_A$ is related to asymmetry of vacuum spectrum around the transition frequency. Interpretation from second order perturbation theory: modes slightly below the atomic resonance, $\omega_k \leq \omega_A$ tend to push the level $|e\rangle$ upwards. Modes above resonance: push downwards. Null effect if spectrum is flat.

Actual calculation requires UV cutoff ω_{uv} and interaction Hamiltonian beyond the resonance (rotating wave) approximation (Hans Bethe \sim 1948, calculation of the Lamb shift). Order of magnitude:

$$\delta\omega_A \approx \gamma \log(\omega_{\text{uv}}/\omega_A) \quad (2.42)$$

and a consistent cutoff is the electron rest mass, $\omega_{\text{uv}} \sim m_e c^2/\hbar$. Hence comparable to the natural linewidth. Note: relativistic theory required, where wavelengths up to electron Compton wavelength $\hbar/m_e c$ are involved. This goes beyond the “long wavelength approximation” because $\hbar/m_e c \sim \alpha_{\text{fs}} a_0 \ll a_0$.

Atomic populations

Actually, we did not yet show that γ is the rate of decay for the excited state. For this, we need the equation of motion for the occupations of the two energy levels. This is described by the atomic operator σ_3 , also called

the “inversion” because $\langle \sigma_3 \rangle > 0$ when the excited state is more occupied than the ground state.

Heisenberg equation of motion

$$\begin{aligned} \frac{d\sigma_3}{dt} &= +2i \sum_k [g_k^* a_k^\dagger \sigma - g_k \sigma^\dagger a_k] \\ &= 2i [\mathcal{E}_0^\dagger(t) \sigma - \sigma^\dagger \mathcal{E}_0(t)] \\ &\quad - 2 \int_0^t dt' [C^*(t-t') \sigma^\dagger(t') \sigma(t) + C(t-t') \sigma^\dagger(t) \sigma(t')] \end{aligned} \quad (2.43)$$

where we brought the operator products in normal order (first line) and inserted the formal solution for $a_k(t)$ (second line). We apply to the t' -integral the same prescription as before and get

$$\begin{aligned} &\int_0^t dt' [C^*(t-t') e^{i\omega_A(t'-t)} \sigma^\dagger(t') \sigma(t) + C(t-t') e^{-i\omega_A(t'-t)} \sigma^\dagger(t) \sigma(t')] \\ &\approx (\gamma - i\delta\omega_A) \sigma^\dagger(t) \sigma(t) + (\gamma + i\delta\omega_A) \sigma^\dagger(t) \sigma(t) = \gamma(\sigma_3(t) + \mathbb{1}) \end{aligned} \quad (2.44)$$

where the frequency shift drops out. By construction, the operator $\sigma_3(t) + \mathbb{1}$ gives the occupation of the excited state. From Eqs.(2.43, 2.44), we can thus read off the decay rate 2γ for the excited state population, while the ground state remains stable.

Finally, the equation for the inversion operator becomes

$$\frac{d\sigma_3}{dt} = -2\gamma(\sigma_3(t) + \mathbb{1}) + 2i [\mathcal{E}_0^\dagger(t) \sigma - \sigma^\dagger \mathcal{E}_0(t)] \quad (2.45)$$

Combined with the equation of motion for the atomic dipole operator

$$\frac{d\sigma}{dt} = -(\gamma + i\omega_A)\sigma + i\sigma_3 \mathcal{E}_0(t) \quad (2.46)$$

we have thus found the “optical Bloch equations”.

In Eq.(2.46), we have made the replacement $\omega_A + \delta\omega_A \mapsto \omega_A$ for the atomic frequency. This is called “renormalization”: we combine the shift induced by the coupling to the vacuum field with the “naked” transition frequency into the frequency that can be physically observed. Recall that in reality, we can never “switch off” the coupling to the vacuum. Hence the “naked” two-level atom that we started with is actually a theoretical artefact.

Approximations

Let us collect the approximations that we had to make.

1. weak coupling between atom and field. We used second order perturbation theory.
2. effective evolution of the atom occurs at long timescales. We found $1/\gamma_e \approx 10^{-9}$ s $\gg \tau_c \approx 10^{-12}$ s. This allows to choose $\tau_c \ll \Delta t \ll 1/\gamma_e$. Note that the field correlation time τ_c is only “short” when the field has a “broad” frequency spectrum $S(\omega)$ (width much larger than γ_e).
3. the period of the emitted radiation is much shorter than $1/\gamma_e$. O.k. for atoms where $\omega_A \gg \gamma_e$.
4. the field state is unaffected by the presence of the atom. Radiated photons escape from the atom and do not interact a second time. The field is a large “reservoir”.

2.5.3 Application 1: Bloch equations

Note that we did not specify yet the state of the light field: it is in fact encoded in the operator $\mathcal{E}_0(t)$ that depends on the initial field operators $a_k(0)$. Two examples will be studied now, the first one being an atom driven by a laser field. We have argued that to a good approximation, we can assume that the light field is in a coherent state $|\alpha_L\rangle$. We assume that at $t = 0$, the total system is in the product state $|\psi(0), \alpha_L\rangle$ and take the expectation value of the Bloch equation. This gives the dynamics of the Bloch vector as follows (optical Bloch equations in the proper sense)

$$\frac{ds}{dt} = -(\gamma + i\omega_A)s + (i/2)s_3\Omega e^{-i\omega_L t} \quad (2.47)$$

$$\frac{ds_3}{dt} = -2\gamma(s_3(t) + 1) + i \left[\Omega^*(t)e^{i\omega_L t} s - s^* \Omega e^{-i\omega_L t} \right] \quad (2.48)$$

where $\Omega/2 = \langle \alpha_L | \mathcal{E}_0(0) | \alpha_L \rangle$ is the (complex) Rabi frequency and ω_L the frequency of the laser mode.

These equations have time-dependent coefficients, but this can be removed by making a transformation into a “rotating frame”. We make the

replacement

$$s(t) \mapsto s(t)e^{-i\omega_L t} \quad (2.49)$$

where the “new” $s(t)$ satisfies Bloch equations with time-independent coefficients

$$\begin{cases} \frac{ds}{dt} = -(\gamma - i\delta)s + (i/2)s_3\Omega \\ \frac{ds_3}{dt} = -2\gamma(s_3(t) + 1) + i[\Omega^*s - s^*\Omega] \end{cases} \quad (2.50)$$

where $\delta = \omega_L - \omega_A$ is the “laser detuning”. Note that the symbols are not the same throughout the books: the Rabi frequency Ω or the decay rate γ can differ by a factor of 2 (or -2), the detuning can have the opposite sign.

The Bloch equations are a “workhorse” of atomic physics and quantum optics. They are used to compute light absorption, excitation spectra, population transfer, radiation forces on atoms etc. In the exercises, you compute the stationary state of the Bloch equations (attention with the different sign for δ : this one should be correct)

$$s_{ss} = \frac{-i(\Omega/2)(\gamma + i\delta)}{\gamma^2 + \delta^2 + \Omega^2/2} \quad (2.51)$$

$$s_{3,ss} = \frac{-(\gamma^2 + \delta^2)}{\gamma^2 + \delta^2 + \Omega^2/2} \quad (2.52)$$

Discussion: average atomic dipole operator (induced by laser field), average inversion. Line broadening.

Exercise: total excitation N , does not commute when laser included. Expectation value of \dot{N} in stationary state, interpretation as total scattered intensity.

Exercise: spectrum of spontaneous emission, from formal solution. Need formal solution of atomic dipole operator, Eq.(2.57).

2.5.4 Application 2: the Glauber photodetector

Roy Glauber (Nobel prize 2005) developed in the 1960s the theory of photodetection. His main result is that the signal of a photodetector is proportional to

$$I(t) \propto \langle E^{(-)}(t)E^{(+)}(t) \rangle \quad (2.53)$$

where $E^{(+)}(t)$ is the positive frequency part of the electric field operator at the detector position. This signal is constructed in such a way that if the field is in the vacuum state, a detector gives no signal: perfectly reasonable. But due to the presence of vacuum fluctuations (also nonzero expectation values of a product of field operators!), not easy to implement in the theory.

We can recover the relevant features of Glauber's theory with our two-level atom. We shall actually show that under suitable approximations,

$$I(t) \propto \langle \mathcal{E}_0^\dagger(t) \mathcal{E}_0(t) \rangle \quad (2.54)$$

where $\mathcal{E}_0(t)$ is the re-scaled electric field operator we introduced in Eq.(2.30). Note that it contains positive frequency components only, and also only those modes that are near-resonant with the atomic transition frequency ω_A . Indeed, Glauber's model for a photodetector is a two-state system that is prepared in the ground state. Incident light can be absorbed, leading to some population in the excited state. This population is then "rapidly removed" from the system. A physical example: the excited atom is ionized and the free electron moves away (it cannot come back to recombine into the ground state). This is actually the process that happens in a photomultiplier ("avalanche photodiode").

So let us see what our Heisenberg equations (2.45, 2.46) give when the atom is in the ground state and the field is in an arbitrary state. We are interested in the rate of change of the excited state population:

$$I := \frac{dp_e}{dt} = \frac{d}{dt} \langle \frac{\sigma_3 + \mathbb{1}}{2} \rangle = \frac{1}{2} \frac{d\langle \sigma_3 \rangle}{dt} \quad (2.55)$$

Since the atom is in the ground state, the operator $\sigma_3 + \mathbb{1}$ that appears in Eq.(2.45) averages to zero. We are left with

$$I = i \langle \mathcal{E}_0^\dagger(t) \sigma(t) - \sigma^\dagger(t) \mathcal{E}_0(t) \rangle \quad (2.56)$$

and insert the formal solution for the atomic dipole operator [similar to Eq.(2.27)]:

$$\sigma(t) = \sigma(0) e^{-(\gamma+i\omega_A)t} + i \int_0^t dt' \sigma_3(t') \mathcal{E}_0(t') e^{-(\gamma+i\omega_A)(t-t')} \quad (2.57)$$

This gives

$$\begin{aligned}
I &= - \int_0^t dt' \left[\langle \mathcal{E}_0^\dagger(t) \sigma_3(t') \mathcal{E}_0(t') \rangle e^{-(\gamma+i\omega_A)(t-t')} + \text{h.c.} \right] \\
&= \int_0^t d\tau \left[\langle \mathcal{E}_0^\dagger(t) \mathcal{E}_0(t-\tau) \rangle e^{-(\gamma+i\omega_A)\tau} + \text{h.c.} \right]
\end{aligned} \tag{2.58}$$

where in the second line, we used the approximation that the inversion $\sigma_3(t')$ is evolving slowly and took its expectation value in the initial state (atom in the ground state, not correlated with the field). If we also assume that the expectation value evolves slowly with time t , $\langle \mathcal{E}_0^\dagger(t) \mathcal{E}_0(t-\tau) \rangle \approx \langle \mathcal{E}_0^\dagger(t+\tau) \mathcal{E}_0(t) \rangle$ [this is strictly true for a stationary field, but stationary fields do not give not very interesting signals on a photodetector], we can combine the ‘+h.c.’ term into the single integral

$$I = \int_{-t}^t dt' \langle \mathcal{E}_0^\dagger(t) \mathcal{E}_0(t+\tau) \rangle e^{-\gamma|\tau|+i\omega_A\tau} \tag{2.59}$$

We observe that the photodetector signal is similar to the Fourier transform of the two-time field correlation function. We can already confirm that the signal is given by a normally ordered expectation value of field operator. Let us consider two limits.

Narrow-band detector

If the decay time $1/\gamma$ is “very long” (compared to the correlation time of the field), the photodetector signal is essentially given by the field spectrum, taken at the transition frequency ω_A . Indeed, the τ -integral in Eq.(2.59) picks from the positive frequency operator $\mathcal{E}_0(t+\tau)$ those components that evolve like $e^{-i\omega_A\tau}$. A narrow band photodetector is thus simply a ‘spectrometer’, and the quantity it measures is the Fourier transform of the temporal correlation (or coherence) function. Experimentally, this situation can be achieved by placing a narrow-band frequency filter in front of a photodetector.

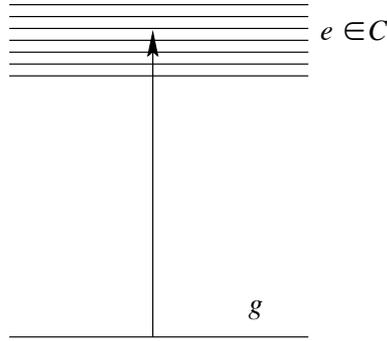


Figure 2.5: Two-level model for a fast photodetector. The signal corresponds to a sum of transition rates into all components of the upper state (quasi)continuum.

Fast detector

In some photodetectors, the excited state has a very short lifetime $1/\gamma$. This happens for example when the excited state actually ionizes and gives away its electron. The excitation rate I is then actually an integral over all components of the excited state ‘continuum’, as sketched in Fig.2.5. Under the integral, the exponential $e^{-\gamma|\tau|}$ becomes very short-ranged in τ , and we can make the replacement

$$e^{-\gamma|\tau|} \mapsto \frac{2}{\gamma} \delta(\tau) \quad (2.60)$$

This leads to Glauber’s formula

$$I(t) = \eta \langle \mathcal{E}_0^\dagger(t) \mathcal{E}_0(t) \rangle \quad (2.61)$$

where the expectation value of the instantaneous intensity operator $\mathcal{E}_0^\dagger(t) \mathcal{E}_0(t)$ appears, in normal order, of course. The prefactor η actually is a number characteristic for the detector and is called “quantum efficiency”. It must be determined experimentally. If the intensity is scaled to “photons per second”, then η gives the detection probability per photon, and I the “rate of detected photons per second”.

Intensity (photon) correlations

Generalization to multiple detection events: $I(t)$ interpreted as probability per unit time to detect one photon at time t . Probability to detect one

photon at time t_1 and another one at t_2 :

$$I(t_1, t_2) = \eta^2 \langle \mathcal{E}_0^\dagger(t_1) \mathcal{E}_0^\dagger(t_2) \mathcal{E}_0(t_2) \mathcal{E}_0(t_1) \rangle, \quad t_2 > t_1 \quad (2.62)$$

time- and normal-ordered operator product. Interpretation.

Temporal coherence

Definition in terms of factorization of $\langle \mathcal{E}_0^\dagger(t) \mathcal{E}_0(t') \rangle$ (first-order temporal coherence). And for second-order.

Examples: monochromatic field always first-order coherent. Black-body radiation not. Two-photon state is second-order coherent, but not in first-order.

Bibliography

- G. Boedeker & C. Henkel (2012). Validity of the quantum regression theorem for resonance fluorescence in a photonic crystal, *Ann. Phys. (Berlin)* **524** (12), 805–13. Highlight ‘Resonance fluorescence spectra near a photonic bandgap’ by Y. Lai, p.A179-180.
- C. Cohen-Tannoudji (1977). Atoms in strong resonant fields. in R. Balian, S. Haroche & S. Liberman, editors, *Frontiers in Laser Spectroscopy (Les Houches XXVII 1975)*, page 3. North Holland, Amsterdam.
- J. Dalibard & C. Cohen-Tannoudji (1985). Atomic motion in laser light: connection between semiclassical and quantum descriptions, *J. Phys. B* **18**, 1661–1683.
- R. Hanbury Brown & R. Q. Twiss (1956). Correlation between photons in two coherent beams of light, *Nature* **177** (4497), 27–29.
- L. Mandel & E. Wolf (1995). *Optical coherence and quantum optics*. Cambridge University Press, Cambridge.
- P. Meystre & M. Sargent III (1999). *Elements of Quantum Optics*. Springer, Berlin, 3rd edition.