

Chapter 3

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.3.1 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.3.1 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”.

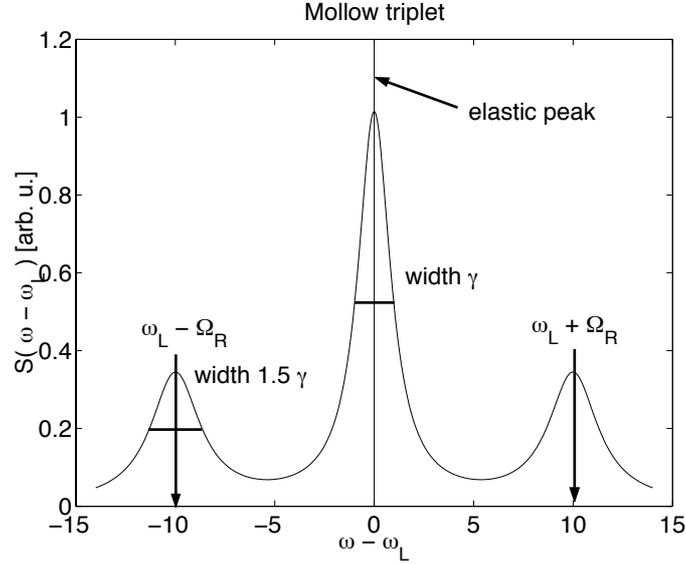


Figure 3.1: The emission spectrum of a driven two-level atom (the Mollow triplet). Parameters: detuning $\Delta = 0$, Rabi frequency $\Omega = 10\gamma$, zero temperature. The ratio between the peak heights is 1 : 3 : 1. The peak surfaces are in the ratio 1 : 2 : 1.

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

3.1 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

$$\mathbf{j}(\mathbf{x}, t) = e^{-i\omega_L t} \mathbf{d}_{ge} \delta(\mathbf{x} - \mathbf{x}_A) \left(-i\omega_L + \frac{d}{dt} \right) \sigma(t) + \text{h.c.} \quad (3.1)$$

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)

$$\mathbf{E}(\mathbf{x}, t) = -\frac{\mathbf{d} - \hat{\mathbf{R}}(\hat{\mathbf{R}} \cdot \mathbf{d})}{4\pi\epsilon_0 c^2} e^{-i\omega_L(t-R/c)} \left(-i\omega_L + \frac{d}{dt}\right)^2 \frac{\sigma(t-R/c)}{R} + \text{h.c.} \quad (3.2)$$

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.(3.2) 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.(3.2) 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,

$$\lim_{t \rightarrow \infty} \langle \mathbf{E}^-(\mathbf{x}, t + \tau) \cdot \mathbf{E}^+(\mathbf{x}, t) \rangle \propto C_\sigma(\tau) = \lim_{t \rightarrow \infty} \langle \sigma^\dagger(t + \tau) \sigma(t) \rangle e^{i\omega_L \tau} \quad (3.3)$$

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

$$S_d(\omega - \omega_L) = \lim_{t \rightarrow \infty} \int d\tau \langle \sigma^\dagger(t + \tau) \sigma(t) \rangle e^{-i(\omega - \omega_L)\tau} \quad (3.4)$$

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.(3.4) 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.(2.103) emerges:

$$S_\sigma(\omega) = \langle \sigma^\dagger \sigma \rangle \delta(\omega - \omega_A) \quad (3.5)$$

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 (3.5) 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

$$C_I(\tau) = \langle : I(t + \tau) I(t) : \rangle = \langle E^\dagger(t) E^\dagger(t + \tau) E(t + \tau) E(t) \rangle \quad (3.6)$$

where the symbols $\langle \cdots \rangle$ 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

$$G_2(\tau) = \langle \sigma^\dagger(t) \sigma^\dagger(t + \tau) \sigma(t + \tau) \sigma(t) \rangle \quad (3.7)$$

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

$$\begin{aligned} G_2(\tau) &= \text{tr} \left[|e\rangle\langle g| U^\dagger(\tau) |e\rangle\langle e| U(\tau) |g\rangle\langle e| \rho(t) \right] \\ &= |\langle e| U(\tau) |g\rangle|^2 \rho_{ee}(t) \end{aligned} \quad (3.8)$$

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.

3.2 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

$$\begin{aligned}\langle \sigma^\dagger(t + \tau)\sigma(t) \rangle &= \text{tr}_{SB} \left[U^\dagger(\tau)\sigma^\dagger(t)U(\tau)\sigma(t)P_{SB}(t) \right] \\ &= \text{tr}_S \left[\sigma^\dagger(t) \text{tr}_B \left(U(\tau)\sigma(t)P_{SB}(t)U^\dagger(\tau) \right) \right]\end{aligned}\quad (3.9)$$

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

$$\rho(t + \tau) = \text{tr}_B \left(U(\tau)P_{SB}(t)U^\dagger(\tau) \right) \quad (3.10)$$

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),

$$P_{SB}(t) \approx \rho_{\text{st}} \otimes \rho_B \quad (3.11)$$

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 (3.9) and (3.10), we observe that both the density matrix and the atomic operator

$$\varrho_\sigma(\tau) \equiv \text{tr}_B \left(U(\tau)\sigma(t)P_{SB}(t)U^\dagger(\tau) \right) \quad (3.12)$$

are determined by the same Nakajima-Zwanziger map (Sec.2.2.2): 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

$$\varrho_\sigma(0) \equiv \sigma\rho_{\text{st}}. \quad (3.13)$$

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

$$\frac{d}{d\tau}\varrho_\sigma = \frac{1}{i\hbar} [H_A, \varrho_\sigma] + \mathcal{L}[\varrho_\sigma]. \quad (3.14)$$

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

3.3 Eigenvalues of the master equation

The explicit solution of (3.14) 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)

$$\begin{pmatrix} -\gamma/2 & \Delta & 0 \\ -\Delta & -\gamma/2 & -\Omega \\ 0 & \Omega & -\gamma \end{pmatrix}. \quad (3.15)$$

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

$$\left(\frac{1}{2}\gamma - \lambda\right)^2 (\gamma - \lambda) + \Omega^2 \left(\frac{1}{2}\gamma - \lambda\right) + \Delta^2 (\gamma - \lambda) = 0 \quad (3.16)$$

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. 3.1. 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.3.2. 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).

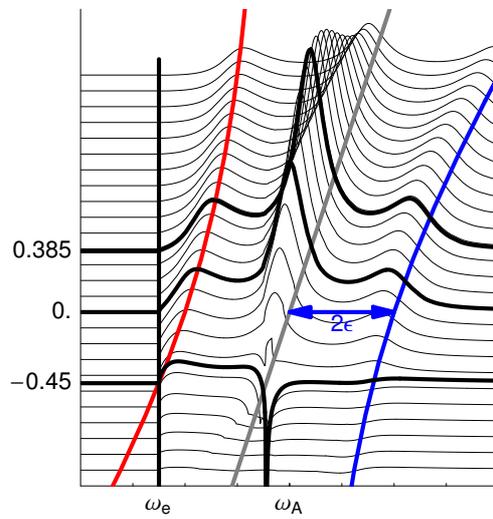


Figure 3.2: Family of spectra for different detunings $\Delta = \omega_L - \omega_A$, marked by the numbers on the left (in units of the Rabi frequency $\Omega = 2\epsilon$). The calculation is made for a photon bath with an inhomogeneous spectral density: there are no available modes for $\omega \leq \omega_e$ (thick vertical line). The red, gray, and blue lines trace the spectrum at the positions of three peaks of the Mollow spectrum.

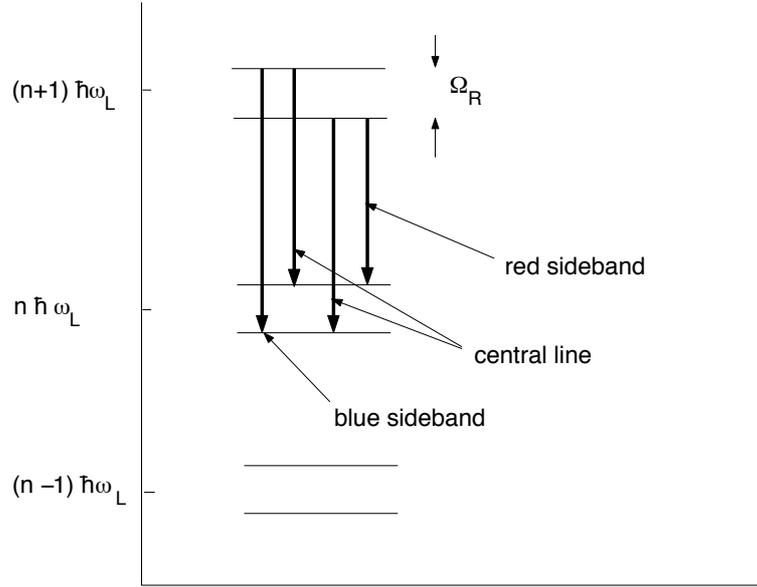


Figure 3.3: Interpretation of the Mollow triplet in terms of transitions between dressed states.

3.4 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 ???. 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. 3.3. 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 $\omega_L \pm \Omega_R$.

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