

Chapter 4

Correlations and Spectra

4.1 Fluctuations and correlations

Introduction

We have so far analyzed so-called ‘one-time averages’ of an open quantum system, $\langle A(t) \rangle = \text{tr } A\rho(t)$, given by the density operator at time t , or via generalized Heisenberg equations. What can we say about time correlation functions like $\langle A(t)B(t') \rangle$ where two times enter? It is clear that we are dealing here with Heisenberg operators $A(t)$ and $B(t')$. We have encountered these correlations when discussing vacuum fluctuations. Indeed, these correlation functions can be interpreted in analogy to classical stochastic processes where one deals with random variables (instead of operators) and where $\langle A(t)B(t') \rangle$ is a measure of the correlations between the variables A and B at different times.

In practise, one often studies stationary cases in the wide sense where the correlations depend only on the time difference,

$$\langle A(t)B(t') \rangle = \langle A(t - t')B(0) \rangle \quad (4.1)$$

One can easily show by cyclic permutations under the trace that this property is true if $A(t)$ and $B(t')$ evolve in time by a Hamiltonian H that commutes with the (initial) state ρ . Such a Hamiltonian model is not general enough to account for dissipation and coupling to an unobserved environment, of course. In the stationary case, we *define* a ‘‘correlation spectrum’’

by expanding the correlation function in a Fourier integral

$$\langle A(t + \tau)B(t) \rangle = \int \frac{d\omega}{2\pi} S_{AB}(\omega) e^{i\omega\tau} \quad (4.2)$$

(the sign in the exponential is conventional). There are also different conventions regarding the operator order: a symmetrized operator product is useful to make the mapping to classical physics easy. In that case, the correlation function $\frac{1}{2}\langle A(t + \tau)B(t) + B(t)A(t + \tau) \rangle$ is real and depends only on τ (provided the average is taken in a stationary state). This implies that the real part of the spectrum is even in ω . We keep the operator order here because we want to deal also with normally ordered averages, for example $\langle a^\dagger(t)a(t') \rangle$.

Exercise. Show that for a single mode (frequency ω_c) in a coherent state $|\alpha\rangle$ the quadrature autocorrelation $\langle X(t)X(t') \rangle$ has a spectrum with intensities $|\alpha|^2$ and $|\alpha|^2 + 1$ at the frequencies $\omega = \omega_c$ and $-\omega_c$.

4.1.1 Power spectrum and signal analysis

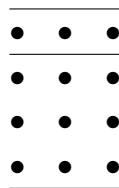
The Eq.(4.2) we used to define the spectrum is actually a theorem, named after Wiener and Khintchine. In many physics textbooks, it does not become clear what is the *theorem* behind this formula. In brief, Wiener showed that the following integral exists for a certain class of autocorrelation functions . . .

$$\mu_A(\omega) = \int d\tau \frac{e^{-i\omega\tau} - 1}{-i\tau} \langle A(t + \tau)A(t) \rangle \quad (4.3)$$

and Khintchine showed that a function $C_A(\tau)$ has a spectral representation

$$C_A(\tau) = \int \frac{d\mu_A(\omega)}{2\pi} e^{i\omega\tau} \quad (4.4)$$

with a certain positive integration measure $d\mu_A(\omega)$. . . if and only if $C_A(\tau)$ is the autocorrelation function of a stochastic process, i.e., $C_A(\tau) = \langle A(t + \tau)A(t) \rangle$. In this context,



$\mu_A(\omega)$ is the integrated spectrum – a quantity that is less singular than the spectral density (which may contain δ functions).

E. Wolf in his books (Born & Wolf, 1959; Mandel & Wolf, 1995) introduces the spectrum via a time-limited Fourier transform:

$$S_A(\omega, t) = \lim_{T \rightarrow \infty} \frac{1}{T} \left\langle \left| \int_{t-T/2}^{t+T/2} dt' e^{i\omega t'} A(t') \right|^2 \right\rangle \quad (4.5)$$

which is a formula well-suited for actual data processing. The time integrals cover only a finite window T and the average can be calculated by shifting this window across a time series (time averaging). In practice, Eq.(4.5) can be used if T is larger than the correlation time τ_c of the process. It is a simple exercise to show that it coincides with the Wiener-Khinchine formula. The theorem is a statement of how probable it is that this analysis of a time series gives the spectrum defined in terms of the autocorrelation function.

A third definition of the power spectrum appears in the mathematical theory of (linear) filters. This can be physically motivated by remembering that we never actually observe the value $A(t)$ of an observable, but some time average. This can be formalized by considering a “detector function” $\varphi(t)$ and forming the integral

$$A_\varphi = \int dt \varphi^*(t) A(t) \quad (4.6)$$

The spectrum then appears as the link between the Fourier transform of the detector and the variance of the filtered signal (we assume that $\langle A_\varphi \rangle = 0$)

$$\langle A_\varphi^\dagger A_\varphi \rangle = \int \frac{d\omega}{2\pi} S_A(\omega) |\tilde{\varphi}(\omega)|^2 \quad (4.7)$$

where $\tilde{\varphi}(\omega)$ is the Fourier transform of the detector function (it exists by assumption because the detector function is smooth and integrable). The advantage of this formulation is that the integrals always exists because $\tilde{\varphi}(\omega)$ has built-in cutoffs at small and large frequencies if it is integrable. And that in actual experiments, we only have access to the spectral density in a certain range of frequencies (defined by the resolution of the detectors). By taking for $\varphi(t)$ an exponential $e^{i\omega t}$ restricted to a finite interval, one recovers the definition (4.5) of Wolf and the prefactor $1/T$ that appears there.

Question. The previous equation was formulated for a stationary process (autocorrelation function depends on the time difference only). How can it be generalized to non-stationary processes?

4.1.2 An interpretation via a sequence of measurements

To conclude, let us formulate a “standard interpretation” of correlation functions that appears in the literature and that may be taken with care in the quantum context. For the general correlation

$$\langle A(t + \tau)B(t) \rangle \quad (4.8)$$

one makes the reasoning: with some probability distribution $P(B(t) = b)$ one measures the value b for the observable B at time t . Then, given this measurement and evolving the system forward over a time step τ , one gets a value a for the observable A with some conditional probability $P(A(t + \tau) = a | \text{“}B(t) \text{ has been measured”})$. This gives the following recipe for the correlation function as a multi-average

$$\langle A(t + \tau)B(t) \rangle = \int da db a P(A(t + \tau) = a | \dots) b P(B(t) = b) \quad (4.9)$$

Note however, that in quantum mechanics, the measurement at time t in general perturbs the system. This can only be avoided if we write the stationary state (at time t) in a diagonal expansion over eigenstates of the operator B . The conditional probability then involves the time evolution of one of these eigenstates over a time step τ . If this trick is not applied, it may happen that the state after the first “measurement” is not a physical state (it is not a hermitean density operator, but something skew). In addition, at intermediate times, it may not be appropriate to talk about the observable B having a value b with a positive probability: this line of thought has somewhat the flavour of a “hidden variable” which we know is problematic from the EPR paradox.

4.1.3 Examples

Single-mode laser

The frequency spectrum of a **single-mode laser** is given by the Fourier transform of the autocorrelation function

$$C_a(\tau) = \lim_{t \rightarrow \infty} \langle \hat{a}^\dagger(t + \tau) \hat{a}(t) \rangle \quad (4.10)$$

where the limit $t \rightarrow \infty$ ensures that the system has reached a steady state.¹

Exercise. Introduce the (formal) Fourier transform of the mode operator

$$\tilde{a}(\omega) = \int dt e^{i\omega t} \hat{a}(t) \quad (4.11)$$

and show that for a stationary correlation function, the Fourier transform has the covariance

$$\langle \tilde{a}^\dagger(\omega) \tilde{a}(\omega') \rangle = S_a(\omega) 2\pi \delta(\omega - \omega') \quad (4.12)$$

where $S_a(\omega)$ is the Fourier transform of $C_a(\tau)$ [Eq.(4.10)].

The simplest case corresponds to a “freely evolving mode” (frequency ω_L where

$$\langle \hat{a}^\dagger(t + \tau) \hat{a}(t) \rangle = \langle \hat{a}^\dagger \hat{a} \rangle e^{i\omega_L \tau} \quad (4.13)$$

and the spectrum is monochromatic

$$S_a(\omega) = \langle \hat{a}^\dagger \hat{a} \rangle 2\pi \delta(\omega - \omega_L) \quad (4.14)$$

Its amplitude is given by the stationary average photon number $\langle \hat{n} \rangle = \langle \hat{a}^\dagger \hat{a} \rangle$.

In a physical system, there are several reasons why the spectrum is not monochromatic:

¹The time difference τ cannot be too large, of course, otherwise one would again get sensitive to initial transients. This can be made more precise with the mathematical tools of “filter theory” and wavelet analysis. The physicist helps himself by introducing a measurement interval of length T which overlaps with the stationary regime, and by reducing the Fourier transform to this interval. The spectrum is then defined in the limit $T \rightarrow \infty$ after suitable normalization.

- the measurement time T is finite. The δ -function in Eq.(4.14) has a width $\mathcal{O}(1/T)$. This is a “trivial” limitation, and one takes in practice the limit $T \rightarrow \infty$.
- losses from the laser cavity at a rate κ . This gives a width $\mathcal{O}(\kappa)$ which can be interpreted in terms of time-frequency uncertainty, now for the “lifetime of a photon” in the cavity.
- spontaneous (not stimulated) emission of the active laser medium. This gives a fundamental limit to the frequency width of the laser, named after Schawlow & Townes. The broadening is of the order $\mathcal{O}(\kappa/\langle\hat{n}\rangle)$ and decreases as the laser intensity is raised. It fundamentally arises due to the discrete nature of the photons.

4.2 Quantum regression hypothesis

The approach discussed so far illustrates that correlation functions like $\langle A(t+\tau)B(t) \rangle$, in their dependence on the *time difference* τ have a dynamics that is very similar to the one-time averages that we calculated with the master equation. Indeed, for the phase diffusion, we assumed that between t and $t+\tau$, the phase (and even the full laser amplitude) evolves according to the same diffusion (or Fokker-Planck) equation as if nothing special happened at $t=0$. The problem here is that one sometimes *interprets* the correlation function by saying “at time t , we have measured the operator A and at the later time $t+\tau$, we measure B ”. This is not exactly true as the operators do not coincide with their expectation values nor with their eigenvalues (results of projective measurements).²

To elaborate this further, let us write down the meaning of the correlation function $\langle a^\dagger(t+\tau)a(t) \rangle$ (any other choice of operators would give the same physics) in the language of “system + bath” dynamics. At the time t , we identify the Heisenberg and Schrödinger picture and use the stationary density operator $\rho(t) = \rho_{\text{st}}$. We need to know also the density operator

²Since about 20 years, Yakir Aharonov has developed a formalism of ‘weak measurements’, intermediate between projective measurements and ‘learning nothing at all’. The motivation was to learn how a quantum system can be described ‘between’ preparation and detection. For a review, see Dressel & al. (2014) and Aharonov & al. (2014).

$P(t) = P_{\text{st}}$ for system + bath. Then, without any further approximation

$$\begin{aligned}\langle a^\dagger(t + \tau)a(t) \rangle &= \text{tr}_{\text{SB}} \left\{ e^{iH\tau} a e^{-iH\tau} a P_{\text{st}} \right\} \\ &= \text{tr}_{\text{S}} \left\{ a \underbrace{\text{tr}_{\text{B}} \left(e^{-iH\tau} a P_{\text{st}} e^{iH\tau} \right)}_{=: \varrho(\tau|a\rho_{\text{st}})} \right\}\end{aligned}\quad (4.15)$$

where tr_{B} is the ‘partial trace’ over the bath Hilbert space. The result is called a ‘reduced density matrix’ for the system, $\varrho(\tau|a\rho_{\text{st}})$. It is all we need to predict the system observable a^\dagger at time $t + \tau$.

The same concept can be used to construct the evolution of the system state (in the sense of a density operator) when it interacts with a bath. Take some initial state,

$$P(0) = \rho(0) \otimes \rho_B(\text{eq}) \quad (4.16)$$

typically taken as the uncorrelated tensor product with a reference state $\rho_B(\text{eq})$ for the bath. To justify this approximation, a quote from Melvin Lax (2000): ‘A bath is by definition a large system that is characterised by few parameters that do not change when it is put into contact with a system.’ If we want to construct a time evolution (master equation) for the system alone, then we better pick just one state for the bath, characterised with only a few parameters (like its temperature). More information about the bath is simply not available. Quoting Lax again: ‘A reservoir’s properties should be anonymous. Its properties should be describable by a small number of parameters, often only one, the temperature.’ (The word ‘anonymous’ is used in the same sense as characters like the ‘Men in Black’ or the ‘Graue Männer’ (M. Ende, *Momo*) are anonymous, and one cannot say more about them than that they look like man in a business suit.)

The S+B state $P(0)$ evolves according to the S+B (total) Hamiltonian

$$P(t) = e^{-iH\tau} P(0) e^{iH\tau} \quad (4.17)$$

and we ‘trace out the bath’ to get a density operator for the system alone:

$$\rho(t) = \text{tr}_{\text{B}} \left(e^{-iH\tau} \rho(0) \otimes \rho_B(\text{eq}) e^{iH\tau} \right) \quad (4.18)$$

This is the Nakajima-Zwanzig approach to the master equation: ‘blow up’ the system state $\rho(0)$ to a system+bath state $P(0)$, evolve in time and

project back into the system Hilbert space. The master equation is the ‘generator’ of this evolution:

$$\rho(t) = e^{\mathcal{L}t}\rho(0), \quad \frac{d\rho}{dt} = \mathcal{L}\rho \quad (4.19)$$

The “Liouville (super)operator” \mathcal{L} used here contains both the commutator with the Hamiltonian and the dissipative Lindblad terms.

We can now come back to the density operator ϱ in Eq.(4.15):

$$\varrho(\tau|a\rho_{\text{st}}) = \text{tr}_{\text{B}}\left(e^{-iH\tau} aP_{\text{st}} e^{iH\tau}\right)$$

It is derived by the same Nakajima-Zwanzig procedure if we may approximate the operator aP_{st} by a tensor product

$$aP_{\text{st}} \approx a\rho_{\text{st}} \otimes \rho_{\text{B}}(\text{eq}) \quad (4.20)$$

where $\rho_{\text{B}}(\text{eq})$ is the state of the bath (same as before).

The difference to the usual picture is that the initial density operator $a\rho_{\text{st}}$ (for the system) or aP_{st} (for system + bath) is not a proper one, since it is not hermitean: we call it a ‘skew state’ in the following. This difference is not really a problem if one remembers that by the linearity of the master equation, we can extend it to “skew states” as well, like $\rho = |\psi\rangle\langle\chi|$. We can write, for example:

$$|\psi\rangle\langle\chi| + |\chi\rangle\langle\psi| = \frac{1}{2}(|\psi\rangle + |\chi\rangle)(\langle\psi| + \langle\chi|) - \frac{1}{2}(|\psi\rangle - |\chi\rangle)(\langle\psi| - \langle\chi|) \quad (4.21)$$

To get the non-symmetric skew operator $|\psi\rangle\langle\chi|$, we continue this construction with the superposition states $|\psi\rangle \pm i|\chi\rangle$. The only technical problem is that this linear combination of true states involves negative (and eventually complex) coefficients, although linear combinations of density operators are physically only defined for real and positive coefficients (a ‘convex sum’)

$$p\rho_1 + (1-p)\rho_2 \quad (4.22)$$

Accepting this procedure, we get the following

Quantum regression hypothesis: If the stationary state of system + bath can be described by the factorized form $P_{\text{st}} = \rho_{\text{st}} \otimes \rho_{\text{B}}$, then the correlation

function evolves in the time difference τ according to the same master equation as the one-time average:

$$\langle a^\dagger(t + \tau)a(t) \rangle = \text{tr} \left(a^\dagger \varrho(\tau|a\rho_{\text{st}}) \right) = \text{tr} \left(a^\dagger e^{\mathcal{L}\tau} a\rho_{\text{st}} \right) \quad (4.23)$$

$$\frac{d}{d\tau} \varrho = \mathcal{L}\varrho, \quad \varrho(0|a\rho_{\text{st}}) = a\rho_{\text{st}} \quad (4.24)$$

The only difference is that one has to evolve a skew operator to find the “conditional state” $\varrho(\tau|a\rho_{\text{st}})$.

The word “regression” is due to Onsager who formulated a “regression hypothesis” in classical physics: “fluctuations of a system in equilibrium (correlations) show the same dynamics as deviations from equilibrium (one-time averages)”. This hypothesis is sometimes called “theorem” because the assumptions beyond the validity of the master equation seem minimal. One thing to check is that the system+bath approach gives exactly the same reduced dynamics even for skew states (the bath average of the coupling vanishes in first order, for example). An exception occurs when “memory terms” play a role, but then even the Lindblad master equation does not apply: the generator \mathcal{L} cannot be constant in time. So within the second-order perturbation theory and in the Markov (no memory) limit, we have good reasons to use the quantum regression “theorem”. Combined with the stationarity of the correlations [see Eq.(3.49)], one thus gets the required spectra from the usual dissipative “forward-in-time” evolution.

4.3 Resonance fluorescence

Overview

As an example, we propose to study here the spectrum of the light emitted by a laser-driven two-level atom. This emission is usually called “fluorescence”.³ Its spectrum shows characteristic deviations from the radiation of a classical dipole. These demonstrate that both the atom and the field are genuinely quantum-mechanical systems. Therefore, resonance fluorescence is a key problem in quantum optics.

³Depending on the order of magnitude of the radiative decay time, one also uses the names “luminescence” or “phosphorescence”.

4.3.1 Dipole correlations and Bloch equations

Emission spectrum

It is a simple exercise to show that the electric field operator contains two contributions: the “free field” that is independent of the atom, and a “source field” that involves the atomic dipole operator. (For the proof, use the operator-valued Maxwell equations.) Using some approximations, the far field at distance x from the atom, is given by Eq.(??).

We have learnt before that the spectrum of the emitted light is the Fourier transform of the field correlation function. As we have seen in Eq.(??), this field is proportional to the dipole itself, and we therefore compute its autocorrelation function in normal order

$$G_1(\tau, t) = \langle \sigma^\dagger(t + \tau)\sigma(t) \rangle \quad (4.25)$$

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 hermitean conjugate of the autocorrelation function, it can be shown that

$$\begin{aligned} S(\omega - \omega_L) &\equiv \int_{-\infty}^{+\infty} d\tau e^{-i(\omega - \omega_L)\tau} \langle \sigma^\dagger(t + \tau)\sigma(t) \rangle \\ &= 2 \operatorname{Re} \int_0^{+\infty} d\tau e^{-i(\omega - \omega_L)\tau} \langle \sigma^\dagger(t + \tau)\sigma(t) \rangle, \end{aligned} \quad (4.26)$$

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 $\omega - \omega_L$.

Quantum regression formula

(This section repeats the reasoning of Sec.4.2.) For the time being, we have worked with Heisenberg operators. The average in the dipole correlation function can be written as

$$\begin{aligned} \langle \sigma^\dagger(t + \tau)\sigma(t) \rangle &= \operatorname{tr}_{AF} [U^\dagger(\tau)\sigma^\dagger(t)U(\tau)\sigma(t)\rho_{AF}(t)] \\ &= \operatorname{tr}_A [\sigma^\dagger(t) \operatorname{tr}_F (U(\tau)\sigma(t)\rho_{AF}(t)U^\dagger(\tau))] \end{aligned} \quad (4.27)$$

where $U(\tau)$ is the complete unitary time evolution and $\rho_{AF}(t)$ the atom+field density matrix at time t .

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

$$\rho_A(t + \tau) = \text{tr}_F(U(\tau)\rho_{AF}(t)U^\dagger(\tau)) \quad (4.28)$$

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

$$\rho_{AF}(t) = \rho_A^{(\text{st})} \otimes \rho_F^{(\text{eq})} \quad (4.29)$$

where $\rho_A^{(\text{st})}$ is the stationary density matrix for the atom and $\rho_F^{(\text{eq})}$ an equilibrium state for the field (the environment). Recall that this is actually an approximation based on the Markov assumption: correlations between atom and field decay rapidly.

Comparing (4.27) and (4.28), we observe that both the density matrix and the atomic operator

$$P_-(\tau) \equiv \text{tr}_F(U(\tau)\sigma(t)\rho_{AF}(t)U^\dagger(\tau)) \quad (4.30)$$

are determined by the same combination of evolving with the complete time evolution and taking the trace. We can therefore *use the same master equation* that we derived for ρ_A to compute the time-dependence of $P_-(\tau)$. This statement is the “quantum regression theorem”.⁴ The only difference is the initial state that involves an additional dipole operator

$$P_-(0) \equiv \sigma\rho_A^{(\text{st})}. \quad (4.31)$$

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 operator $P_-(\tau)$ is

$$\frac{d}{d\tau}P_- = \frac{1}{i\hbar}[H_A, P_-] + \mathcal{L}[P_-]. \quad (4.32)$$

⁴We shall use the word “formula” because there are people who insist that it is based on an approximation and hence not a real theorem.

Eigenvalues of the Bloch equations

The explicit solution of (4.32) involves some algebraic manipulations that are not very illuminating, and we shall give only a sketch of the most important techniques and results.

One of the main ideas is to write $P_-(\tau)$ as a sum of eigenvectors of the master equation:⁵

$$P_-(\tau) = \sum_{\lambda} P_-^{(\lambda)} e^{-\lambda t} \quad (4.33)$$

Each of these vectors 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 λ 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. It is easy to see that the corresponding eigenfunction $P_-^{(\text{st})}$ is the stationary density matrix $\rho_A^{(\text{st})}$ (simply because its time derivative vanishes by construction). For the operator $P_-(\tau)$, we thus also find an eigenvalue $\lambda = 0$, and

$$P_-(\tau) = P_-^{(\text{st})} + \text{terms that vanish as } \tau \rightarrow \infty \quad (4.34)$$

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).

⁵More precisely, the matrices are “right eigenvectors” of a superoperator \mathcal{L} that combines the Hamiltonian commutator and the Lindblad parts of the master equation: $\mathcal{L}P_-^{(\lambda)} = -\lambda P_-^{(\lambda)}$. Since \mathcal{L} is in general not symmetric (hermitean), one must distinguish between left and right eigenvectors.

For the operator $P_-(\tau)$, 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 because it is preserved under the master equation generated by \mathcal{L}):

$$P_-^{(\text{st})} = \rho_A^{(\text{st})} \text{tr} P_-(0) = \rho_A^{(\text{st})} \text{tr} (\sigma \rho_A^{(\text{st})}) = \rho_A^{(\text{st})} \rho_{eg}^{(\text{st})}$$

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

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

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

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

This expectation value generates the closest approximation to a “classical” field that is possible for this quantum system. It is called the “elastically scattered light” as it has the same frequency as the driving laser.

The other eigenvalues of the master equation provide the “inelastic” components of the fluorescence spectrum. If we expand the operator $P_-(\tau)$ 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)

$$\begin{pmatrix} -\gamma/2 & \Delta & 0 \\ -\Delta & -\gamma/2 & -\Omega \\ 0 & \Omega & -\gamma \end{pmatrix}. \quad (4.35)$$

The eigenvalues $-\lambda$ of this matrix are the solutions of the cubic equation

$$\left(\frac{1}{2}\gamma - \lambda\right)^2 (\gamma - \lambda) + \Omega^2 \left(\frac{1}{2}\gamma - \lambda\right) + \Delta^2 (\gamma - \lambda) = 0 \quad (4.36)$$

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 (exercise!)

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

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. 4.1. The heights of the peaks can be obtained from a more detailed analysis, and is discussed in the exercises.

4.3.2 Mollow spectrum

The peaks in the Mollow spectrum can be interpreted in terms of transitions between the dressed states of the Jaynes Cummings Paul model. 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

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

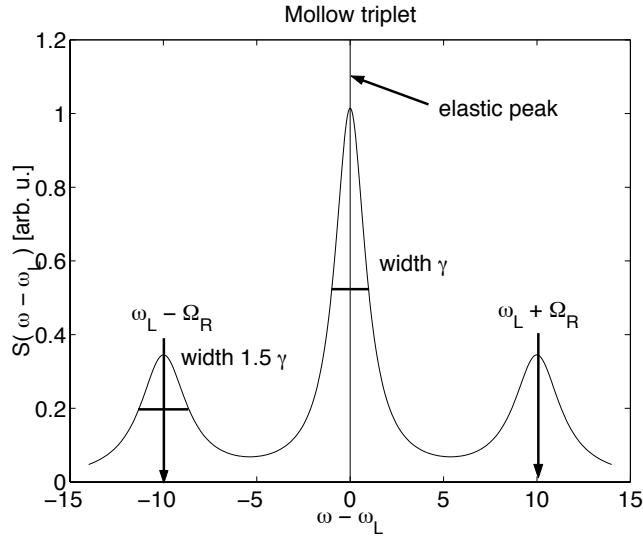


Figure 4.1: The emission spectrum of a driven two-level atom (the Mollow triplet). Parameters: detuning $\Delta = 0$, Rabi frequency $\Omega = 10 \gamma$, zero temperature. The ratio between the peak heights is 1 : 3 : 1. The peak surfaces are in the ratio 1 : 2 : 1.

as shown schematically in Fig. 4.2. 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 spin label of the state changes under the transition: from $|n, +\rangle$ to $|n - 1, -\rangle$ or from $|n, -\rangle$ to $|n - 1, +\rangle$. Here, the fluorescence photon is shifted in frequency 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. Therefore, the emission spectrum contains, in addition to the carrier at the laser frequency (as expected from the stationary state) sidebands at $\omega_L \pm \Omega_R$. As a general remark, this inelastic spectrum illustrates that a two-level atom is the simplest nonlinear medium one can imagine. Indeed, the correlation function $\langle \sigma(t + \tau) \sigma(t) \rangle$ is nonzero and gives the source strength for a

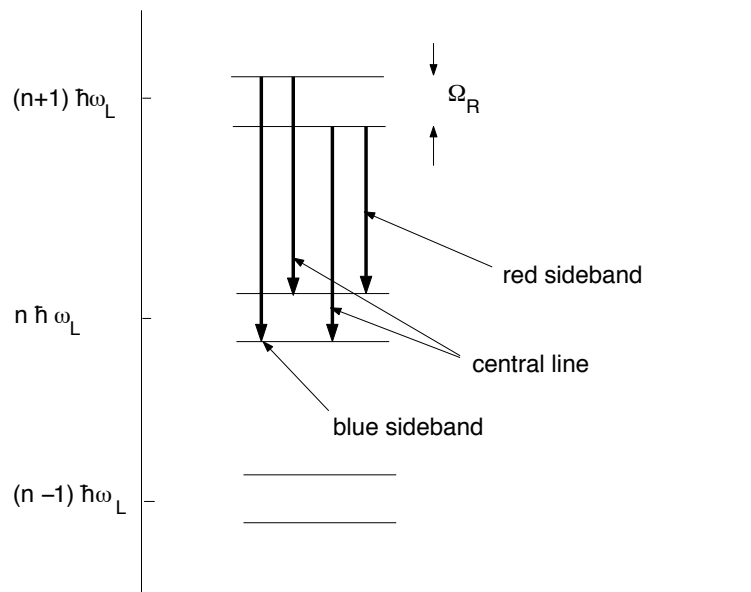


Figure 4.2: Interpretation of the Mollow triplet in terms of transitions between dressed states.

“coherent two-photon field”. The photons are emitted in pairs with a sum frequency $\omega_1 + \omega_2 = 2\omega_L$ since $\langle \sigma(t + \tau)\sigma(t) \rangle$ is proportional to $e^{-2i\omega_L t}$.

A few numerical results based on the solution of the Bloch equations. The numerics has been ‘upgraded’ to allow for skew input states where the Bloch vector is complex.

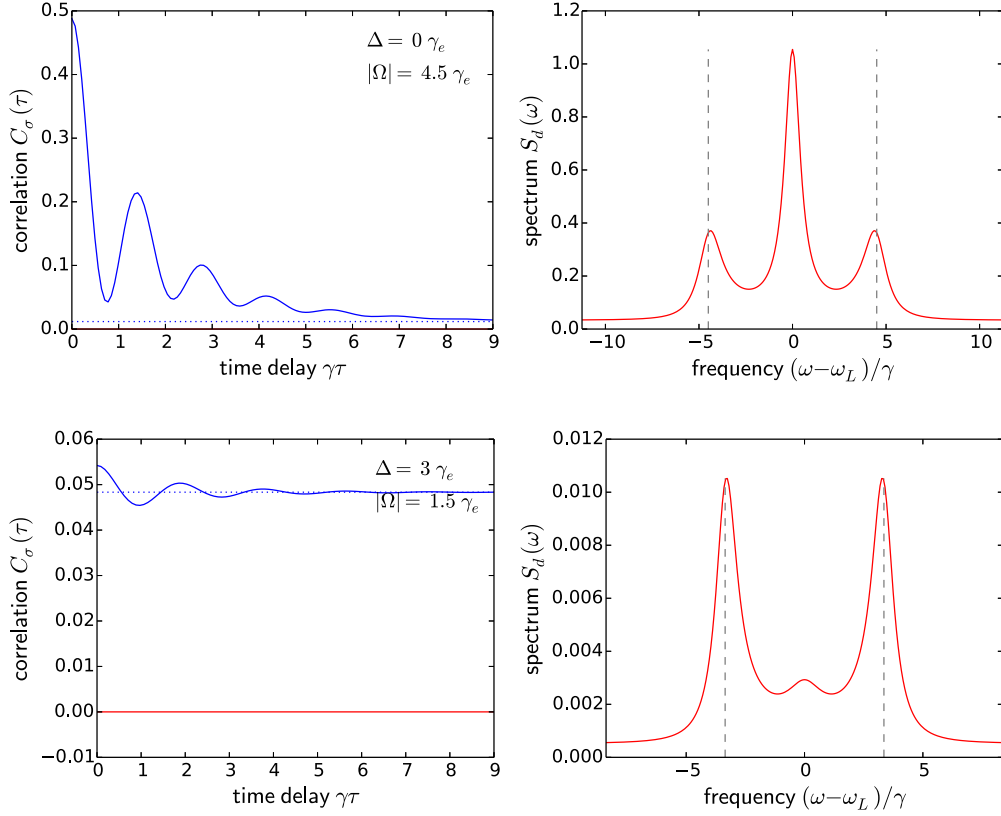


Figure 4.3: (*left*) Dipole correlation function $\langle \sigma^\dagger(\tau)\sigma(0) \rangle$. Blue (red): real (imaginary) part. (*right*) Spectrum: Mollow triplet. (*top row*) Laser on resonance, driving the atom into saturation. For a weaker Rabi frequency, the sidebands merge with the central line, leaving a single peak. (*bottom*) Laser off-resonance, weaker Rabi frequency. The side bands appear at the ‘generalised Rabi frequency’ $(|\Omega|^2 + \Delta^2)^{1/2}$ (dashed lines). For a stronger Rabi frequency, the central line gets stronger. The dotted line on the *left* gives the asymptotic value of the correlation, computed analytically from the stationary solution of the Bloch equations.

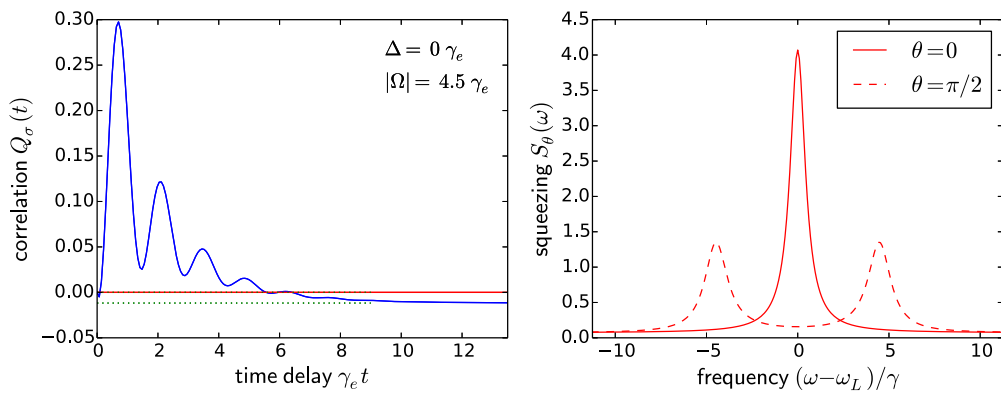


Figure 4.4: (left) Squeezing correlation $Q(\tau) = \langle \sigma(\tau)\sigma(0) \rangle$. Note the zero value at $\tau = 0$. (right) Spectrum of the corresponding quadratures $X_\theta = e^{i\theta}\sigma + e^{-i\theta}\sigma^\dagger$: the quadrature in phase with the laser ($\theta = 0$) peaks at the central line, the quadrature out of phase peaks at the sidebands. Parameter regime: laser on resonance, driving the atom into saturation.

4.3.3 Photon correlations and antibunching

Intensity correlation function: probability to detect one photon at time t and another one at time $t + \tau$. Glauber's theory of photodetection (any book on quantum optics): normal and time-ordered operator average

$$G_2(\tau, t) = \langle \sigma^\dagger(t) \sigma^\dagger(t + \tau) \sigma(t + \tau) \sigma(t) \rangle \quad (4.37)$$

Quantum regression formula: expectation value of $\sigma^\dagger \sigma$ at time step τ after dissipative evolution

$$G_2(\tau, t) = \text{tr}[\sigma^\dagger \sigma e^{\mathcal{L}\tau} P(t)], \quad P(t) = \sigma \rho(t) \sigma^\dagger = \sigma \rho^{(\text{st})} \sigma^\dagger \quad (4.38)$$

we have denoted by $e^{\mathcal{L}\tau}$ the solution of the master equation over the time step τ . Observe that the starting point $P(t)$ is a correct quantum state, it is generated from the equilibrium state by projecting to the ground state. (This is not a skew operator as we encountered it for the dipole correlation function.)

Hence physical interpretation of G_2 correlation (up to normalization): probability to find the atom in the excited state after starting in the ground state. Immediate consequence: $G_2(0, t) = 0$ because some delay is needed to excite the atom. This is called *anti-bunching*: the photons have the tendency not to arrive closely together (“bunched”).

This is a “non-classical” feature, because if σ were a classical random variable, one can show from the Schwarz inequality (exercise) that

$$\text{classical:} \quad G_2(0, t) \geq |G_1(0, t)|^2 > 0 \quad (4.39)$$

where the first-order correlation function G_1 was defined in Eq.(4.25). In practice, the “classical limit” is reached if there are many emitters that are excited by the laser and that radiate independently one of the other. At any time $\tau \geq 0$, one then finds some excited emitter that is able to emit a photon. In this situation, anti-bunching can be used as a signature that just one emitter is present in the excitation region. This is used, for example, in biophysical systems where photon correlations can be used to measure the (position-dependent) decay rates γ inside a focused laser spot with a weak drive amplitude.

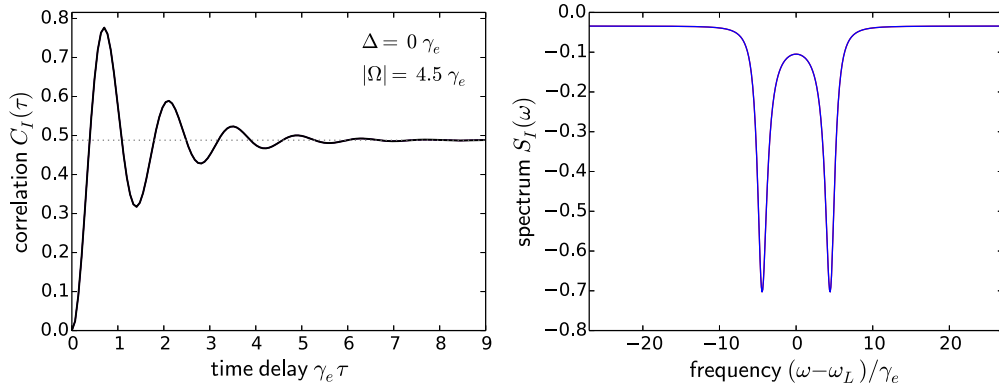


Figure 4.5: (*left*) Intensity correlation $G_2(\tau) = \langle \sigma^\dagger(0)\sigma^\dagger(\tau)\sigma(\tau)\sigma(0) \rangle$. Note the ‘anti-bunching hole’ at $\tau = 0$, characteristic for a single emitter. (*right*) Corresponding spectrum: negative intensities characteristic for squeezing (non-classical light). Parameter regime: laser on resonance, driving the atom into saturation.

4.4 Squeezing correlations in a cavity

Taken from Chapter 7 in the quantum optics book by Walls & Milburn (1994) and Chapter 14 in the book by Orszag (2000).

Split of the system into cavity (one operator a) and input/output fields $A_{\text{in}}(t)$, $A_{\text{out}}(t)$. Correlation functions for input and output fields. Quantum Langevin equations for cavity operator, including squeezing Hamiltonian for cavity field, cavity damping rate and input noise. Calculation of stationary correlations in Fourier space. Discussion of homodyne measurement of quadratures, of the corresponding noise spectra. Definition of squeezing in the frequency domain, shot-noise reference level.

The input field $A_{\text{in}}(t)$ may contain, in addition to its quantum noise, an incident laser beam. More generally, we can write for the normally ordered correlation (denoted by $: \dots :$)

$$\langle : A_{\text{in}}(t + \tau) A_{\text{in}}(t) : \rangle = |E|^2 g_1(\tau) \quad (4.40)$$

where $|E|^2$ is proportional to the laser intensity and $g_1(\tau)$ its normalized autocorrelation function. Its Fourier transform describes the spectrum of the laser.

The output field $A_{\text{out}}(t)$ is what the experimenter can measure. By performing a homodyne measurement (see the chapter on quantum states of light and the beam splitter in QO I), one can measure quadrature operators that are defined as

$$A_{\text{out}}(t) = \frac{1}{2}(X_{\theta}(t) + iY_{\theta}(t)) e^{i\theta - i\omega_0 t} \quad (4.41)$$

where ω_0 is the carrier frequency and θ a phase controlled by the reference beam in the homodyne measurement. (Eq.(4.41) gives only the positive-frequency component of the output field.) In radio communications, the quadratures $X_{\theta}(t)$ describe the envelope (or frequency modulation) of the carrier signal that oscillates at ω_0 , they typically evolve on a much slower time scale. The same applies to optical signals. This justifies an expansion of the light field in a narrow frequency band around ω_0 .

The following autocorrelation functions of the quadratures provide information about squeezing:

$$\langle X_{\theta}(t + \tau)X_{\theta}(t) \rangle, \quad \langle Y_{\theta}(t + \tau)Y_{\theta}(t) \rangle. \quad (4.42)$$

They can be measured from the fluctuations of the signal difference at the two output ports of a homodyne detector. For a field in the vacuum state, these correlations are equal and given by the so-called shot-noise limit. With the normalization

$$A_{\text{out}}(t) = \sqrt{\frac{\hbar\omega_0}{2\varepsilon_0}} \int \frac{d\omega}{2\pi} a(\omega) e^{-i\omega t}, \quad [a(\omega), a(\omega')^{\dagger}] = 2\pi\delta(\omega - \omega') \quad (4.43)$$

one gets the following shot-noise level for the quadrature autocorrelation

$$\langle X_{\theta}(t + \tau)X_{\theta}(t) \rangle_{\text{vac}} = \frac{\hbar\omega_0}{2\varepsilon_0} \delta(\tau) \quad (4.44)$$

which corresponds to a flat spectrum $S_{\text{SN}} = \hbar\omega_0/2\varepsilon_0$.

System-bath formalism for cavity losses, input and output noise operators yields the equations of motion for the cavity field [Eqs.(14.44, 48) in Orszag (2000)], the quantum Langevin equations

$$\frac{da}{dt} = -\frac{i}{\hbar} [a, H] - \frac{\kappa}{2} a + \sqrt{\kappa} A_{\text{in}}(t) \quad (4.45)$$

$$\frac{da}{dt} = -\frac{i}{\hbar} [a, H] + \frac{\kappa}{2} a - \sqrt{\kappa} A_{\text{out}}(t) \quad (4.46)$$

The second equation can be read as the time-reversed version: “given the output field at a later time, what can we infer for the cavity field at an earlier time”. This is getting more and more imprecise as one moves into the past, because of the damping. Combining these two equations, we get

$$A_{\text{out}} = -A_{\text{in}} + \sqrt{\kappa}a \quad (4.47)$$

The fact that the output is related to the field *at the same time* t is due to the *Markov approximation* that has been applied in deriving these quantum Langevin equations: losses are determined by the value of the field at the same time. In general, they also depend on past values of the field; the non-Markovian formalism is still subject to current research.

Model for damped cavity filled with a nonlinear medium:

$$H = \hbar\omega_c a^\dagger a + \frac{i\hbar}{2} \left(\varepsilon e^{-i\omega_p t} a^{\dagger 2} - \varepsilon^* e^{-i\omega_p t} a^2 \right) \quad (4.48)$$

where ε is proportional to the amplitude of the ‘pump field’ at frequency ω_p . Switch to rotating frame $a(t) = \tilde{a}(t) e^{-i\omega_p t/2}$, $A_{\text{in}}(t) = \tilde{A}_{\text{in}}(t) e^{-i\omega_p t/2}$ and find from Eq.(4.45)

$$\frac{d\tilde{a}}{dt} = -i\delta_c \tilde{a} + \varepsilon \tilde{a}^\dagger - \frac{\kappa}{2} \tilde{a} + \sqrt{\kappa} \tilde{A}_{\text{in}}(t) \quad (4.49)$$

where $\delta_c = \omega_c - \omega_p/2$ is related to the detuning of the pump relative to twice the cavity resonance. We drop the tilde and compute the equations of motion for the following quadrature operators

$$a = \frac{x + iy}{2} e^{i\theta} \quad (4.50)$$

where the phase reference is half the phase of the pump field: $\varepsilon = |\varepsilon| e^{2i\theta}$.

Details to be continued ... see the Chapter by Orszag (2000) for the final result of the squeezing spectrum (for $\delta_c = 0$):

$$\langle : X_{\text{out}}(\omega), X_{\text{out}}(\omega') : \rangle = \langle X_{\text{out}}(\omega) X_{\text{out}}(\omega') \rangle \quad (4.51)$$

$$\begin{aligned} & - \langle X_{\text{out}}(\omega) \rangle \langle X_{\text{out}}(\omega') \rangle - \langle X_{\text{out}}(\omega) X_{\text{out}}(\omega') \rangle_{\text{vac}} \\ & = S_{X_{\text{out}}}(\omega) S_{\text{SN}} \delta(\omega + \omega') \end{aligned} \quad (4.52)$$

Measured relative to the shot noise, one gets for the two quadratures [Fig.4.6]:

$$S_{X_{\text{out}}}(\omega) = \frac{2\kappa|\varepsilon|}{\omega^2 + (\kappa/2 - |\varepsilon|)^2}$$

$$S_{Y_{\text{out}}}(\omega) = \frac{-2\kappa|\varepsilon|}{\omega^2 + (\kappa/2 + |\varepsilon|)^2} \quad (4.53)$$

‘Perfect squeezing’ corresponds to a normalised spectrum $S_Y = -1$: it happens for $|\varepsilon| = \kappa/2$ and $\omega = 0$. Note that this is accompanied by a diverging spectrum in the orthogonal quadrature. For $|\varepsilon| > \kappa/2$, an instability appears to happen that is not described by this linearised theory. It is likely that by taking into account ‘pump depletion’, one gets a finite result. This corresponds to the pump photons that are converted into pairs of cavity photons by the nonlinear process (spontaneous parametric down conversion).

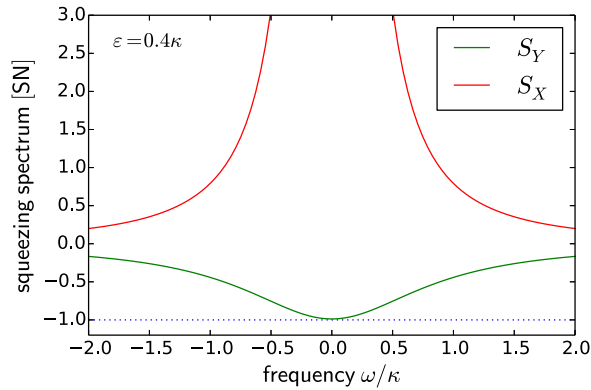


Figure 4.6: Noise spectrum of output quadratures X_{out} and Y_{out} , normalised to shot noise.

4.5 The fluctuation–dissipation theorem

(Material not covered in SS 2019.)

The fluctuation-dissipation theorem (FDT) is one of the key results of linear response theory. It is actually a statement that appears in many different forms, the common feature being a link between, on the one hand, the fluctuations of some system observable in thermal equilibrium and, on the other, a response function for this observable when the system is brought out of equilibrium by an external perturbation. One of the early statements of the FDT is, for example, the Einstein relation between the diffusion coefficient and the friction coefficient for a Brownian particle Einstein (1905). We outline in this section a proof of the FDT for quantum fluctuations, following Callen & Welton (1951).

4.5.1 Statement

Consider two observables A and B and their two-time correlation function

$$C_{AB}(t, t') := \langle A(t)B(t') \rangle - \langle A(t) \rangle \langle B(t') \rangle \quad (4.54)$$

where $A(t)$ is an operator in the Heisenberg picture and the average $\langle \dots \rangle$ is taken in thermal equilibrium ($\beta = 1/k_B T$, $Z = \text{tr}(e^{-\beta H})$)

$$\langle \dots \rangle := \frac{1}{Z} \text{tr}(e^{-\beta H} \dots), \quad (4.55)$$

with H the system Hamiltonian. Since equilibrium is a stationary state, the one-time average $\langle A(t) \rangle = \langle A \rangle_{\text{eq}}$ is actually time-independent and the correlation function $C_{AB}(t, t')$ only depends on the time difference $\tau = t - t'$. We consider its Fourier transform (the cross-correlation spectrum)⁶

$$S_{AB}(\omega) := \int_{-\infty}^{+\infty} d\tau C_{AB}(\tau) e^{i\omega\tau} \quad (4.56)$$

The spectrum exists whenever the observables $A(t)$ and $B(t')$ decorrelate sufficiently fast as the time difference $|\tau|$ increases, as expected on physical grounds. If $A = B$, $S_{AA}(\omega) = S_A(\omega)$ specifies the strength of the fluctuations of A in a frequency band around ω .

The FDT links the correlation spectrum to the response function $\chi_{AB}(\omega)$:

$$S_{AB}(\omega) = \frac{1}{i\omega} \frac{\hbar\omega}{1 - e^{-\beta\hbar\omega}} (\chi_{AB}(\omega) - \chi_{BA}(\omega)^*) \quad (4.57)$$

The response function χ_{AB} connects the deviation of the observable A from its equilibrium value to a perturbation described by the interaction Hamiltonian $H_{\text{int}}(t) = -f(t)B$. To first order in the (c-number valued) “force” $f(t)$:

$$\langle A(t) \rangle_f - \langle A \rangle_{\text{eq}} = \int_{-\infty}^{+\infty} \chi_{AB}(\tau) f(t - \tau) d\tau \quad (4.58)$$

By causality, $\chi_{AB}(\tau)$ vanishes for $\tau < 0$ and decreases to zero for $\tau \rightarrow \infty$. As a consequence, its Fourier transform $\chi_{AB}(\omega)$,

$$\chi_{AB}(\omega) = \int_0^{+\infty} \chi_{AB}(\tau) e^{i\omega\tau} d\tau, \quad (4.59)$$

⁶In the SS 2010 lecture, we used the opposite sign convention for ω .

is analytical in the upper half of the complex ω -plane.

In the literature, the FDT is often stated in terms of a symmetrized correlation function. We retain a specific operator ordering here because these correlation functions directly provide photodetector signals or excited state decay rates in the context of quantum electrodynamics. It also turns out that the genuine contribution of quantum fluctuations can be identified in a simple way.

4.5.2 Proof

We first connect the response function to a commutator. The equation of motion for the Heisenberg operator A reads, in the presence of the external force,

$$\frac{d}{dt}A(t) = -\frac{i}{\hbar} [A(t), H] + \frac{i}{\hbar} [A(t), B(t)] f(t), \quad (4.60)$$

where H is the Hamiltonian without the perturbation. The first term generates the free evolution of the operator, $A_{\text{free}}(t) = e^{iHt}A(0)e^{-iHt}$, while the second is due to the perturbation. Solving to first order in $f(t)$ and taking the expectation value according to Eq.(4.55) leads to

$$\langle A(t) \rangle_f = \langle A_{\text{free}}(t) \rangle + \frac{i}{\hbar} \int_0^{+\infty} \langle [A_{\text{free}}(t), B_{\text{free}}(t - \tau)] \rangle f(t - \tau) d\tau, \quad (4.61)$$

where the time dependence of the ‘free’ operators is generated by the evolution under H . This will be the case for any time-dependent operator in the following, and we therefore drop the subscript ‘free’.

The first term in Eq.(4.61) is actually time-independent in the canonical ensemble defined in Eq.(4.55) and therefore equal to the equilibrium value $\langle A \rangle_{\text{eq}}$. From the second term, we can read off the response function introduced in Eq.(4.58),

$$\chi_{AB}(\tau) = \begin{cases} \frac{i}{\hbar} \langle [A(t), B(t - \tau)] \rangle & \text{for } \tau \geq 0, \\ 0 & \text{for } \tau < 0, \end{cases} \quad (4.62)$$

The response function is thus itself a correlation function between operators; it does not depend on t because of the stationarity of equilibrium. It can be checked directly that for hermitean A and B , $\chi_{AB}(\tau)$ is real as it is in classical linear response theory where it links two real quantities.

Taking the Fourier transform of χ_{AB} , we get an expression for the response function in terms of a field correlation spectrum

$$\chi_{AB}(\omega) = \frac{i}{\hbar} \int_0^{+\infty} e^{i\omega\tau} \langle [A(\tau), B(0)] \rangle d\tau. \quad (4.63)$$

By causality, the time integral is running over one half of the real axis only. We can make an integral over all τ appear, as it occurs in the spectrum (4.56), by forming the combination

$$-i\hbar \{\chi_{AB}(\omega) - \chi_{BA}(-\omega)\} = \int_{-\infty}^{+\infty} e^{i\omega\tau} \{C_{AB}(\tau) - C_{BA}(-\tau)\} d\tau. \quad (4.64)$$

We added and subtracted $\langle A(\tau)\rangle\langle B(0)\rangle$ under the integral to generate the correlation functions $C_{AB}(\tau)$ and $C_{BA}(-\tau)$, whose Fourier integrals exist separately. The following identity that we prove below, allows to permute operators occurring in correlation spectra computed in the canonical ensemble:

$$\int_{-\infty}^{+\infty} e^{i\omega\tau} C_{BA}(-\tau) d\tau = e^{-\beta\hbar\omega} \int_{-\infty}^{+\infty} e^{i\omega\tau} C_{AB}(\tau) d\tau. \quad (4.65)$$

In the classical theory, $\hbar = 0$ and operator ordering is irrelevant. Using this in the second term of Eq.(4.64), we get the FDT:

$$-i\hbar \{\chi_{AB}(\omega) - \chi_{BA}(-\omega)\} = (1 - e^{-\beta\hbar\omega}) S_{AB}(\omega). \quad (4.66)$$

With $\chi_{BA}(-\omega) = \chi_{BA}(\omega)^*$, which follows from the reality of $\chi_{BA}(\tau)$, this reduces to Eq.(4.57). The spectrum $S_{AA}(\omega)$ is positive, as it should, for a passive medium with a non-negative ‘‘absorption rate’’, $\omega \text{Im} \chi_{AA}(\omega) \geq 0$.

To prove the identity (4.65), we start from the right hand side and write the equilibrium average of Eq. (4.55) explicitly

$$e^{i\omega\tau} \langle A(\tau)B(0)\rangle = e^{i\omega\tau} Z^{-1} \text{Tr} \left[e^{-\beta H} e^{iH\tau/\hbar} A e^{-iH\tau/\hbar} B \right] \quad (4.67)$$

One shifts the integration path into the complex τ -plane, putting $\tau \mapsto \tau - i\hbar\beta$. The convergence of the τ -integral at $|\tau| \rightarrow \infty$ is ensured by subtracting $e^{i\omega\tau} \langle A(\tau)\rangle\langle B(0)\rangle$. Along the shifted path, Eq.(4.67) becomes

$$\begin{aligned} & e^{\beta\hbar\omega} e^{i\omega\tau} Z^{-1} \text{Tr} \left[e^{-\beta H} e^{(\beta+i\tau/\hbar)H} A e^{-(\beta+i\tau/\hbar)H} B \right] \\ &= e^{\beta\hbar\omega} e^{i\omega\tau} Z^{-1} \text{Tr} \left[e^{-\beta H} B e^{iH\tau/\hbar} A e^{-iH\tau/\hbar} \right] \\ &= e^{\beta\hbar\omega} e^{i\omega\tau} \langle B(0)A(\tau)\rangle, \end{aligned} \quad (4.68)$$

using cyclic permutation under the trace. A similar argument shows that the last correlation function is equal to $\langle B(-\tau)A(0)\rangle$, leading to $C_{BA}(-\tau)$. The τ -integral then yields the right hand side of Eq. (4.65).

4.5.3 Dipole fluctuations of a polarizable particle

Our first illustration of the FDT will also serve later to compute the dispersion interaction between polarizable particles. We take two components of the electric dipole operator, D_j and D_k , and ask for the cross-correlation spectrum $S_{D_j D_k}(\omega)$ that we denote $D_{jk}(\omega)$ in the following. For the FDT, we need the response function for the dipole subject to the interaction Hamiltonian $-D_k f(t)$. It is clear that this represents the excitation of the dipole by an electric field $\mathbf{E}(t) = \mathbf{e}_k f(t)$, linearly polarized along the x_k -axis. The corresponding response function is well known in frequency space: it is the polarizability tensor $\alpha_{jk}(\omega)$. We use the convention that in frequency space, the induced dipole moment is given by $\hat{D}_j(\omega) = \sum_k \alpha_{jk}(\omega) \hat{E}_k(\omega)$. For the Fourier transform $\hat{D}_j(\omega)$ of the dipole moment, the FDT yields the following correlation function

$$\begin{aligned} \langle \hat{D}_j(\omega) \hat{D}_k^*(\omega') \rangle &= D_{jk}(\omega) 2\pi \delta(\omega - \omega') \\ &= \frac{1}{i\omega} \left[\frac{\hbar\omega}{1 - e^{-\hbar\omega/k_B T}} \right] [\alpha_{jk}(\omega) - \alpha_{kj}^*(\omega)] \delta(\omega - \omega') \end{aligned} \quad (4.69)$$

If the dipole polarizability is symmetric (which can be shown to be true if the dipole matrix elements are real), we thus get

$$D_{ij}(\omega) = \frac{2\hbar}{1 - e^{-\hbar\omega/k_B T}} \text{Im} \alpha_{jk}(\omega) \quad (4.70)$$

It is important to notice that dissipation $\propto \text{Im} \alpha_{jk}(\omega)$ does not vanish at absolute zero ($T = 0$ K). However, the remaining fluctuations affect only *positive* frequencies! This can easily be seen by the following limit

$$\lim_{T \rightarrow 0} \frac{1}{1 - e^{-\hbar\omega/k_B T}} = \begin{cases} 1 & \omega > 0 \\ 1/2 & \omega = 0 \\ 0 & \omega < 0 \end{cases} \quad (4.71)$$

This asymmetry illustrates the fundamental difference between classical and quantum fluctuation spectra. Indeed, classical quantities commute and this leads to a spectral density that is symmetric in ω . This result is recovered from the FDT in the high-temperature limit where

$$\lim_{T \rightarrow \infty} \frac{\hbar\omega}{1 - e^{-\hbar\omega/k_B T}} = k_B T. \quad (4.72)$$

4.5.4 The electromagnetic vacuum

Our second example is the electromagnetic field and its fluctuations. (See the book by Novotny & Hecht (2006) for details.) For the correlation between the

fields $E_j(\mathbf{r})$ and $E_k(\mathbf{r}')$, we need the response function to the interaction Hamiltonian $-E_k(\mathbf{r}')f(t)$. This corresponds physically to a point dipole moment located at position \mathbf{r}' and oriented along the x_k -axis. The response function in question is thus given, in frequency space, by the Green function $\mathbf{G}(\mathbf{r}, \mathbf{r}'; \omega)$. We use the convention that (Fourier transform of) the electric field $\hat{E}_j(\mathbf{r}, \omega)$ radiated by a point dipole moment $\hat{D}_k(\omega)$ located at \mathbf{r}' is given by

$$\hat{E}_j(\mathbf{r}, \omega) = \sum_k G_{jk}(\mathbf{r}, \mathbf{r}'; \omega) \hat{D}_k(\omega) \quad (4.73)$$

The FDT for the fluctuating fields is then given by

$$\langle \hat{E}_j(\mathbf{r}, \omega) \hat{E}_k^*(\mathbf{r}', \omega') \rangle = \frac{2\hbar}{1 - e^{-\hbar\omega/k_B T}} \text{Im}\{G_{jk}(\mathbf{r}, \mathbf{r}'; \omega)\} 2\pi\delta(\omega - \omega') \quad (4.74)$$

using the reciprocity of the Green tensor, $G_{kj}(\mathbf{r}', \mathbf{r}; \omega) = G_{jk}(\mathbf{r}, \mathbf{r}'; \omega)$ (Jackson, 1975). This result establishes the correspondence between field fluctuations (left side) and dissipation (right side) which is expressed in terms of the imaginary part of the Green's function. It can be checked, using a direct calculation in the plane-wave basis, that Eq.(4.74) coincides with the result of elementary field quantization.

The advantage of Eq.(4.74) is that it applies very generally to an arbitrary arrangement of objects, described by a spatially varying linear permittivity and permeability. Absorbing objects are also allowed for: they would play in the quantum theory the role of operator Langevin sources for the field via their polarization fluctuations, see, e.g., Eqs.(4.69, ??). The FDT (4.74) is strictly valid, however, only at equilibrium, i.e. when the field and the sources are at the same temperature. In this case, it can also be derived directly from the fluctuations of dipolar sources. These give rise to radiation with a strength proportional to Eq.(4.69). For a given distribution of sources, the Green function $\mathbf{G}(\mathbf{r}, \mathbf{r}_0; \omega)$ maps the dipole fluctuations at \mathbf{r}_0 onto the electromagnetic field at \mathbf{r} . Using properties of \mathbf{G} and the requirement of equilibrium between the field and its sources (no net energy transport), one can derive (4.74). For a more detailed discussion, see Henry & Kazarinov (1996); Eckhardt (1984).

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