

Chapter 2

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,

$$\langle A(t)B(t') \rangle = \langle A(t - t')B(0) \rangle \quad (2.1)$$

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 . This is a model that 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 taking the Fourier transform

$$S_{AB}(\omega) = \int d\tau \langle A(t + \tau)B(t) \rangle e^{-i\omega\tau} \quad (2.2)$$

(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 symmetric in τ (provided the average is taken in a stationary state). This implies that the spectrum is even in ω . We keep the operator order here because we want to deal also with normally ordered averages, for example $\langle a^\dagger(t)a(t') \rangle$.

2.1 Power spectrum and signal analysis

The Eq.(2.2) 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:

$$\mu_A(\omega) = \int d\tau \frac{e^{-i\omega\tau} - 1}{-i\tau} \langle A(t+\tau)A(t) \rangle \quad (2.3)$$

... and Khintchine showed that the autocorrelation function has a spectral representation

$$\langle A(t+\tau)A(t) \rangle = \int \frac{d\mu_A(\omega)}{2\pi} e^{i\omega\tau} \quad (2.4)$$

where $d\mu_A(\omega)$ is a certain positive integration measure. It is clear that $\mu_A(\omega)$ 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:

$$S_A(\omega, t) = \lim_{T \rightarrow \infty} \frac{1}{T} \left\langle \left| \int_{t-T/2}^{t+T/2} dt' e^{i\omega t'} A(t') \right|^2 \right\rangle \quad (2.5)$$

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.(2.5) 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.

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

$$A_\varphi = \int dt \varphi^*(t) A(t) \quad (2.6)$$

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

$$\langle A_\varphi^\dagger A_\varphi \rangle = \int \frac{d\omega}{2\pi} S_A(\omega) |\tilde{\varphi}(\omega)|^2 \quad (2.7)$$

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 exist 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 (2.5) 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?

2.2 Examples

2.2.1 Single-mode laser

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

$$C_a(\tau) = \lim_{t \rightarrow \infty} \langle \hat{a}^\dagger(t + \tau) \hat{a}(t) \rangle \quad (2.8)$$

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

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

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

$$\tilde{a}(\omega) = \int dt e^{i\omega t} \hat{a}(t) \quad (2.9)$$

and show that for a stationary correlation function,

$$\langle \tilde{a}^\dagger(\omega) \tilde{a}(\omega') \rangle = S_a(\omega) 2\pi \delta(\omega - \omega') \quad (2.10)$$

where $S_a(\omega)$ is the Fourier transform of $C_a(\tau)$ [Eq.(2.8)].

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

$$\langle \hat{a}^\dagger(t + \tau) \hat{a}(t) \rangle = \langle \hat{a}^\dagger \hat{a} \rangle e^{i\omega_L \tau} \quad (2.11)$$

and the spectrum is monochromatic

$$S_a(\omega) = \langle \hat{a}^\dagger \hat{a} \rangle \delta(\omega - \omega_L) \quad (2.12)$$

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:

- the measurement time T is finite. The δ -function in Eq.(2.12) 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.

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.

2.2.2 Driven two-level atom

The spectrum of *resonance fluorescence* is a fundamental quantity for quantum optics. It 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 appear for a harmonic oscillator, be it quantized or damped, as long as it is linearly driven.

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

$$\mathbf{j}(\mathbf{x}, t) = e^{-i\omega_L t} \mathbf{d}_{ge} \left(-i\omega_L + \frac{d}{dt} \right) \sigma(t) + \text{h.c.} \quad (2.13)$$

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)

$$\mathbf{E}(\mathbf{x}, t) = -\frac{\mathbf{d} - \hat{\mathbf{R}}(\hat{\mathbf{R}} \cdot \mathbf{d})}{4\pi\epsilon_0 c^2} e^{-i\omega_L(t-R/c)} \left(-i\omega_L + \frac{d}{dt} \right)^2 \frac{\sigma(t - R/c)}{R} + \text{h.c.} \quad (2.14)$$

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.(??) are much slower. We thus have

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

According to photodetector theory, we need the normally ordered autocorrelation function of the electric field,

$$\lim_{t \rightarrow \infty} \langle \mathbf{E}^-(\mathbf{x}, t + \tau) \cdot \mathbf{E}^+(\mathbf{x}, t) \rangle \propto C_\sigma(\tau) = \lim_{t \rightarrow \infty} \langle \sigma^\dagger(t + \tau) \sigma(t) \rangle e^{i\omega_L \tau} \quad (2.15)$$

The proportionality factor is simply ignored in many treatments. The spectrum of the resonance fluorescence is thus related to the normally ordered dipole autocorrelation. Note that in the stationary limit $t \rightarrow \infty$, the delay time R/c drops out from the autocorrelation function. 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. 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.(2.12) emerges:

$$S_\sigma(\omega) = \langle \sigma^\dagger \sigma \rangle \delta(\omega - \omega_A) \quad (2.16)$$

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 (2.16) is of course a too crude approximation because it ignores the natural linewidth of the atomic transition.

2.2.3 Intensity correlations

Finally, *intensity correlations* provide our last example. In the context of resonance fluorescence, they give an experimental check that one deals with a single emitter, not a large number. 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 multiphoton 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

$$C_I(\tau) = \langle : I(t + \tau)I(t) : \rangle = \langle E^\dagger(t)E^\dagger(t + \tau)E(t + \tau)E(t) \rangle \quad (2.17)$$

where the symbols $: \dots :$ 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

$$G_2(\tau) = \langle \sigma^\dagger(t)\sigma^\dagger(t + \tau)\sigma(t + \tau)\sigma(t) \rangle \quad (2.18)$$

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

$$\begin{aligned} G_2(\tau) &= \text{tr} [|e\rangle\langle g| U^\dagger(\tau) |e\rangle\langle e| U(\tau) |g\rangle\langle e| \rho(t)] \\ &= |\langle e|U(\tau)|g\rangle|^2 \rho_{ee}(t) \end{aligned} \quad (2.19)$$

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.

2.2.4 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

$$\langle A(t + \tau)B(t) \rangle \quad (2.20)$$

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 $P(A(t + \tau) = a | “B(t) \text{ has been measured}”)$. This gives the following recipe for the correlation function as a multi-average

$$\langle A(t + \tau)B(t) \rangle = \int da db a P(A(t + \tau) = a | \dots) b P(B(t) = b) \quad (2.21)$$

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.

2.3 Schawlow-Townes spectrum of the laser

Up to now, we have assumed that the laser mode oscillates at the frequency ω_L . This is not strictly true. Its frequency spectrum has a finite width. In particular, one observes experimentally that the linewidth gets narrower far above threshold. The corresponding limit of the laser linewidth has been derived in the 1960/70’s by Schawlow and Townes. We sketch here a derivation of this formula.

2.3.1 Idea: phase diffusion

The spectrum of the laser is related to the autocorrelation function of the laser mode operator a :

$$S(\omega) = \lim_{t \rightarrow \infty} \int d\tau e^{-i\omega\tau} \langle a^\dagger(t+\tau)a(t) \rangle \quad (2.22)$$

where the average is taken in the stationary state that is reached at large times. We shall denote this state ρ_{ss} or by the subscript $\langle \dots \rangle_{\text{ss}}$. If the mode were evolving freely at the laser frequency, $a(t+\tau) = a(t)e^{-i\omega_L t}$, then the spectrum would be a δ -peak, $S(\omega) = \langle a^\dagger a \rangle_{\text{ss}} 2\pi\delta(\omega - \omega_L)$ and proportional to the mean photon number. This is no longer true when we take into account that the gain mechanism also involves “spontaneous emission” where atoms of the pumping medium emit a spontaneous photon (not stimulated, hence no phase relation with the existing laser field). This gives the laser field a “fluctuating amplitude” that we have to characterize.

The main idea of Schawlow & Townes is that the laser field is essentially subject to phase fluctuation, but not to intensity fluctuations. In a semiclassical description, we thus have a mode amplitude $a(t) = \sqrt{\bar{n}_{\text{ss}}} e^{i\phi(t) - i\omega_L t}$, where only the phase is fluctuating. We shall see below that the fluctuations of the laser phase are “diffusive” – the phase makes a “random walk”. In terms of a distribution function $P(\phi, t)$, this behaviour is described by a diffusion equation,

$$\frac{\partial}{\partial t} P = D \frac{\partial^2}{\partial \phi^2} P \quad (2.23)$$

where D is called “phase diffusion coefficient”. You may remember this equation from heat conduction. Its solution, for an initial state with a fixed phase ϕ_0 , is given by a gaussian distribution

$$P(\phi, t | \phi_0) = \frac{1}{\sqrt{4\pi Dt}} e^{-(\phi - \phi_0)^2 / 4Dt}, \quad t > 0. \quad (2.24)$$

The width of the gaussian is $2Dt$ and increases with time. For very large times, the distribution is completely flat. (This solution neglects the fact that the phase is only defined in the interval $[0, 2\pi]$. See the exercises for this case.) For $t \rightarrow 0$, one recovers a δ -function centered at ϕ_0 .

With this result, we can compute the temporal correlation function of the laser field,

$$\langle a^\dagger(t+\tau)a(t) \rangle = \bar{n}_{\text{ss}} e^{i\omega_L \tau} \langle e^{-i[\phi(t+\tau) - \phi(t)]} \rangle,$$

which gives us the spectrum by a Fourier transform with respect to τ . We can assume that the initial phase $\phi(t)$ has some value ϕ_0 and that the later phase $\phi = \phi(t + \tau)$ is a random variable with the distribution $P(\phi, \tau|\phi_0)$ of Eq.(2.24). Taking the average, we find

$$\langle a^\dagger(t + \tau)a(t) \rangle = \bar{n}_{\text{ss}} e^{i\omega_L \tau} \int d\phi P(\phi, \tau|\phi_0) e^{-i(\phi - \phi_0)} = \bar{n}_{\text{ss}} e^{i\omega_L \tau} e^{-D\tau}.$$

The temporal correlation function thus decays exponentially with a coherence time $\tau_c = 1/D$. This argument applies only for $\tau > 0$. But if the laser state is stationary, we can argue for $\tau < 0$ that

$$\langle a^\dagger(t + \tau)a(t) \rangle = \langle a^\dagger(t)a(t - \tau) \rangle = \langle a^\dagger(t - \tau)a(t) \rangle^* \quad (2.25)$$

by taking the hermitean conjugate of the operator product. The spectrum (2.22) is thus obtained from a “half-sided Fourier transform”

$$S(\omega) = 2 \operatorname{Re} \int_0^\infty d\tau e^{-i\omega\tau} \langle E^*(t + \tau)E(t) \rangle = \frac{2I_{\text{ss}}D}{(\omega - \omega_L)^2 + D^2} \quad (2.26)$$

The laser spectrum is centered at ω_L with a “Lorentzian” lineshape and a width of the order of D . *The laser linewidth is thus limited by the phase diffusion coefficient.*

2.3.2 Diffusion coefficient

See Sec.3.1 for details. The quantum theory of the laser provides an estimate for the rate of phase diffusion and hence the linewidth of the laser. This is the so-called Schawlow-Townes limit

$$D \approx \frac{\kappa}{4\bar{n}_{\text{ss}}} \quad (2.27)$$

where κ is the linewidth of the laser cavity and \bar{n}_{ss} the average photon number.

2.4 Quantum regression hypothesis

Material covered by a student talk.

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.

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 $\rho(t) = \rho_{ss}$. We need to know also the density operator $P(t) = P_{ss}$ for system + bath. Then, without any further approximation

$$\begin{aligned} \langle a^\dagger(t + \tau)a(t) \rangle &= \text{tr}_{\text{SB}} \left\{ e^{iH\tau} a e^{-iH\tau} a P_{ss} \right\} \\ &= \text{tr}_{\text{S}} \left\{ a \underbrace{\text{tr}_{\text{B}} \left(e^{-iH\tau} a P_{ss} e^{iH\tau} \right)}_{=: \varrho(\tau|a\rho_{ss})} \right\} \end{aligned} \quad (2.28)$$

Here, we identify a reduced “density matrix” of the system, $\varrho(\tau|a\rho_{ss})$ that is derived by the same system+bath procedure that we discussed in the first Chapter: “blow up” the system density operator to a system+bath state, evolve this state under the (total) Hamiltonian and trace out the bath. The difference to the usual picture is that the initial density operator $a\rho_{ss}$ (for the system) or aP_{ss} is not a proper one, since it is not hermitean. This difference is not really a problem if we remember that by linearity, we could extend a positive map to “skew states” as well, like $\rho = |\psi\rangle\langle\chi|$. So we get the following

Quantum regression hypothesis. If the stationary state of system + bath can be described by the factorized form $P_{ss} = \rho_{ss} \otimes \rho_{\text{B}}$, then the correlation function evolves in the time difference τ according to the same master equation as the one-time average:

$$\langle a^\dagger(t + \tau)a(t) \rangle = \text{tr} \left(a^\dagger \varrho(\tau|a\rho_{ss}) \right) = \text{tr} \left(a^\dagger e^{\mathcal{L}\tau} a \rho_{ss} \right) \quad (2.29)$$

$$\frac{d}{d\tau}\varrho = \mathcal{L}\varrho, \quad \varrho(0|a\rho_{ss}) = a\rho_{ss} \quad (2.30)$$

The only difference is that one has to evolve a skew operator to find the “conditional state” $\varrho(\tau|a\rho_{ss})$. The “Liouville (super)operator” \mathcal{L} used here contains both the commutator with the Hamiltonian and the dissipative Lindblad terms.

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.(2.25)], one thus gets the required spectra from the usual dissipative “forward-in-time” evolution.

We apply quantum regression in the next section to study another cornerstone of quantum optics, the spectrum of resonance fluorescence.

2.5 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”.² 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.

²Depending on the order of magnitude of the radiative decay time, one also uses the names “luminescence” or “phosphorescence”.

2.5.1 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 \mathbf{x} from the atom, is given by Eq.(2.14).

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.(2.14), this field is proportional to the dipole itself, and we therefore compute its autocorrelation function in normal order

$$G_1(\tau, t) = \langle \sigma^\dagger(t + \tau)\sigma(t) \rangle \quad (2.31)$$

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

$$\begin{aligned} S(\omega - \omega_L) &\equiv \int_{-\infty}^{+\infty} d\tau e^{-i(\omega - \omega_L)\tau} \langle \sigma^\dagger(t + \tau)\sigma(t) \rangle \\ &= 2 \operatorname{Re} \int_0^{+\infty} d\tau e^{-i(\omega - \omega_L)\tau} \langle \sigma^\dagger(t + \tau)\sigma(t) \rangle, \end{aligned} \quad (2.32)$$

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.2.4.) For the time being, we have worked with Heisenberg operators. The average in the dipole correlation function can be written as

$$\begin{aligned} \langle \sigma^\dagger(t + \tau)\sigma(t) \rangle &= \operatorname{tr}_{AF} [U^\dagger(\tau)\sigma^\dagger(t)U(\tau)\sigma(t)\rho_{AF}(t)] \\ &= \operatorname{tr}_A [\sigma^\dagger(t) \operatorname{tr}_F (U(\tau)\sigma(t)\rho_{AF}(t)U^\dagger(\tau))] \end{aligned} \quad (2.33)$$

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

$$\rho_A(t + \tau) = \text{tr}_F \left(U(\tau) \rho_{AF}(t) U^\dagger(\tau) \right) \quad (2.34)$$

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),

$$\rho_{AF}(t) = \rho_A^{(\text{st})} \otimes \rho_F^{(\text{eq})} \quad (2.35)$$

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 (2.33) and (2.34), we observe that both the density matrix and the atomic operator

$$P_-(\tau) \equiv \text{tr}_F \left(U(\tau) \sigma(t) \rho_{AF}(t) U^\dagger(\tau) \right) \quad (2.36)$$

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”.³ The only difference is the initial state that involves an additional dipole operator

$$P_-(0) \equiv \sigma \rho_A^{(\text{st})}. \quad (2.37)$$

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

$$\frac{d}{d\tau} P_- = \frac{1}{i\hbar} [H_A, P_-] + \mathcal{L}[P_-]. \quad (2.38)$$

³We 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 (2.38) 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:⁴

$$P_-(\tau) = \sum_{\lambda} P_-^{(\lambda)} e^{-\lambda t} \quad (2.39)$$

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

$$P_-(\tau) = P_-^{(\text{st})} + \text{terms that vanish as } \tau \rightarrow \infty \quad (2.40)$$

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).

⁴More 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)

$$\begin{pmatrix} -\gamma/2 & \Delta & 0 \\ -\Delta & -\gamma/2 & -\Omega \\ 0 & \Omega & -\gamma \end{pmatrix}. \quad (2.41)$$

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

$$\left(\frac{1}{2}\gamma - \lambda\right)^2 (\gamma - \lambda) + \Omega^2 \left(\frac{1}{2}\gamma - \lambda\right) + \Delta^2 (\gamma - \lambda) = 0 \quad (2.42)$$

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

2.5.2 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

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

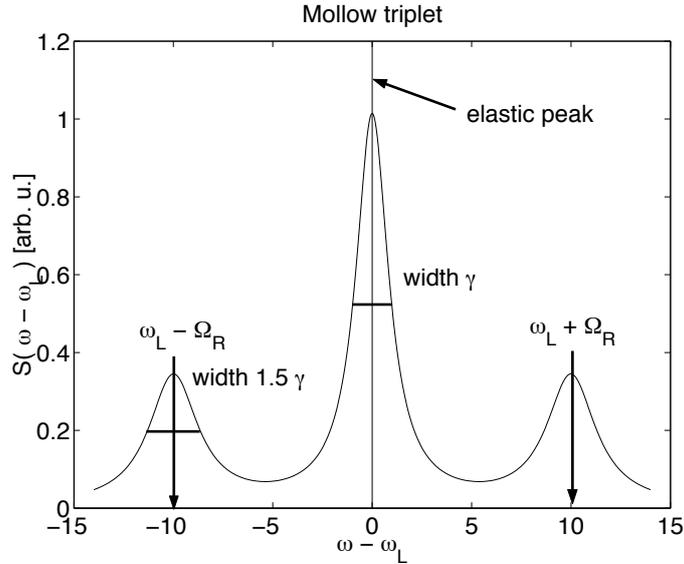


Figure 2.1: The emission spectrum of a driven two-level atom (the Mollow triplet). Parameters: detuning $\Delta = 0$, Rabi frequency $\Omega = 10\gamma$, zero temperature. The ratio between the peak heights is 1 : 3 : 1. The peak surfaces are in the ratio 1 : 2 : 1.

as shown schematically in Fig. 2.2. 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

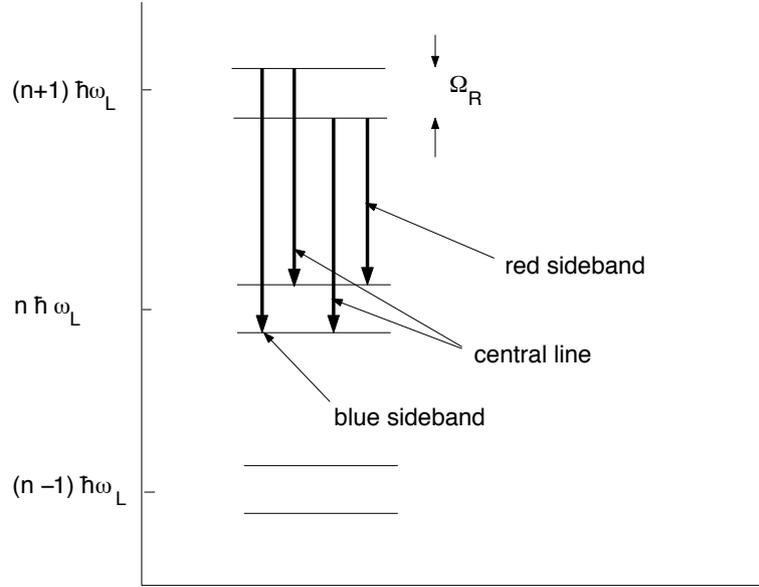


Figure 2.2: Interpretation of the Mollow triplet in terms of transitions between dressed states.

“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 $e^{-2i\omega_L t}$.

2.5.3 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

$$G_2(\tau, t) = \langle \sigma^\dagger(t)\sigma^\dagger(t + \tau)\sigma(t + \tau)\sigma(t) \rangle \quad (2.43)$$

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

$$G_2(\tau, t) = \text{tr}[\sigma^\dagger\sigma e^{\mathcal{L}\tau}P(t)], \quad P(t) = \sigma\rho(t)\sigma^\dagger = \sigma\rho^{(\text{st})}\sigma^\dagger \quad (2.44)$$

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 (details?) that

$$\text{classical: } G_2(0, t) \geq |G_1(0, t)|^2 > 0 \quad (2.45)$$

where the first-order correlation function G_1 was defined in Eq.(2.31). 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.

2.6 Squeezing correlations in a cavity

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

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 $\langle \dots \rangle$)

$$\langle : A_{\text{in}}(t + \tau) A_{\text{in}}(t) : \rangle = |E|^2 g_1(\tau) \quad (2.46)$$

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

$$A_{\text{out}}(t) = \frac{1}{2}(X_\theta(t) + iY_\theta(t)) e^{i\theta - i\omega_0 t} \quad (2.47)$$

where ω_0 is the carrier frequency and θ a phase controlled by the reference beam in the homodyne measurement. (Eq.(2.47) 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:

$$\langle X_\theta(t + \tau) X_\theta(t) \rangle, \quad \langle Y_\theta(t + \tau) Y_\theta(t) \rangle. \quad (2.48)$$

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

$$A_{\text{out}}(t) = \sqrt{\frac{\hbar\omega_0}{2\varepsilon_0}} \int \frac{d\omega}{2\pi} a(\omega) e^{-i\omega t}, \quad [a(\omega), a(\omega')^\dagger] = 2\pi\delta(\omega - \omega') \quad (2.49)$$

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

$$\langle X_\theta(t + \tau) X_\theta(t) \rangle_{\text{vac}} = \frac{\hbar\omega_0}{2\varepsilon_0} \delta(\tau) \quad (2.50)$$

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

Link between output field correlations and input field autocorrelation, normal and time order.

Model for damped cavity filled with a nonlinear medium.

Quantum Langevin equation and Fourier solution.

Results of the squeezing spectrum.

2.7 The fluctuation–dissipation theorem

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).

2.7.1 Statement

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

$$C_{AB}(t, t') := \langle A(t)B(t') \rangle - \langle A(t) \rangle \langle B(t') \rangle \quad (2.51)$$

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

$$\langle \dots \rangle := \frac{1}{Z} \text{tr}(e^{-\beta H} \dots), \quad (2.52)$$

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)⁵

$$S_{AB}(\omega) := \int_{-\infty}^{+\infty} d\tau C_{AB}(\tau) e^{i\omega\tau} \quad (2.53)$$

⁵In the SS 2010 lecture, we used the opposite sign convention for ω .

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 $\chi_{AB}(\omega)$:

$$\boxed{S_{AB}(\omega) = \frac{1}{i\omega} \frac{\hbar\omega}{1 - e^{-\beta\hbar\omega}} (\chi_{AB}(\omega) - \chi_{BA}(\omega)^*)} \quad (2.54)$$

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” $f(t)$:

$$\langle A(t) \rangle_f - \langle A \rangle_{\text{eq}} = \int_{-\infty}^{+\infty} \chi_{AB}(\tau) f(t - \tau) d\tau \quad (2.55)$$

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

$$\chi_{AB}(\omega) = \int_0^{+\infty} \chi_{AB}(\tau) e^{i\omega\tau} d\tau, \quad (2.56)$$

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.

2.7.2 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,

$$\frac{d}{dt}A(t) = -\frac{i}{\hbar} [A(t), H] + \frac{i}{\hbar} [A(t), B(t)] f(t), \quad (2.57)$$

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.(2.52) leads to

$$\langle A(t) \rangle_f = \langle A_{\text{free}}(t) \rangle + \frac{i}{\hbar} \int_0^{+\infty} \langle [A_{\text{free}}(t), B_{\text{free}}(t - \tau)] \rangle f(t - \tau) d\tau, \quad (2.58)$$

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.(2.58) is actually time-independent in the canonical ensemble defined in Eq.(2.52) 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.(2.55),

$$\chi_{AB}(\tau) = \begin{cases} \frac{i}{\hbar} \langle [A(t), B(t - \tau)] \rangle & \text{for } \tau \geq 0, \\ 0 & \text{for } \tau < 0, \end{cases} \quad (2.59)$$

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

$$\chi_{AB}(\omega) = \frac{i}{\hbar} \int_0^{+\infty} e^{i\omega\tau} \langle [A(\tau), B(0)] \rangle d\tau. \quad (2.60)$$

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 (2.53), by forming the combination

$$-i\hbar \{ \chi_{AB}(\omega) - \chi_{BA}(-\omega) \} = \int_{-\infty}^{+\infty} e^{i\omega\tau} \{ C_{AB}(\tau) - C_{BA}(-\tau) \} d\tau. \quad (2.61)$$

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:

$$\int_{-\infty}^{+\infty} e^{i\omega\tau} C_{BA}(-\tau) d\tau = e^{-\beta\hbar\omega} \int_{-\infty}^{+\infty} e^{i\omega\tau} C_{AB}(\tau) d\tau. \quad (2.62)$$

In the classical theory, $\hbar = 0$ and operator ordering is irrelevant. Using this in the second term of Eq.(2.61), we get the FDT:

$$-i\hbar \{\chi_{AB}(\omega) - \chi_{BA}(-\omega)\} = (1 - e^{-\beta\hbar\omega}) S_{AB}(\omega). \quad (2.63)$$

With $\chi_{BA}(-\omega) = \chi_{BA}(\omega)^*$, which follows from the reality of $\chi_{BA}(\tau)$, this reduces to Eq.(2.54). 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 (2.62), we start from the right hand side and write the equilibrium average of Eq. (2.52) explicitly

$$e^{i\omega\tau} \langle A(\tau)B(0) \rangle = e^{i\omega\tau} Z^{-1} \text{Tr} \left[e^{-\beta H} e^{iH\tau/\hbar} A e^{-iH\tau/\hbar} B \right] \quad (2.64)$$

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.(2.64) becomes

$$\begin{aligned} & e^{\beta\hbar\omega} e^{i\omega\tau} Z^{-1} \text{Tr} \left[e^{-\beta H} e^{(\beta+i\tau/\hbar)H} A e^{-(\beta+i\tau/\hbar)H} B \right] \\ &= e^{\beta\hbar\omega} e^{i\omega\tau} Z^{-1} \text{Tr} \left[e^{-\beta H} B e^{iH\tau/\hbar} A e^{-iH\tau/\hbar} \right] \\ &= e^{\beta\hbar\omega} e^{i\omega\tau} \langle B(0)A(\tau) \rangle, \end{aligned} \quad (2.65)$$

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. (2.62).

2.7.3 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

$$\begin{aligned} \langle \hat{D}_j(\omega) \hat{D}_k^*(\omega') \rangle &= D_{jk}(\omega) 2\pi \delta(\omega - \omega') \\ &= \frac{1}{i\omega} \left[\frac{\hbar\omega}{1 - e^{-\hbar\omega/k_B T}} \right] [\alpha_{jk}(\omega) - \alpha_{kj}^*(\omega)] \delta(\omega - \omega') \end{aligned} \quad (2.66)$$

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

$$D_{ij}(\omega) = \frac{2\hbar}{1 - e^{-\hbar\omega/k_B T}} \text{Im} \alpha_{jk}(\omega) \quad (2.67)$$

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

$$\lim_{T \rightarrow 0} \frac{1}{1 - e^{-\hbar\omega/k_B T}} = \begin{cases} 1 & \omega > 0 \\ 1/2 & \omega = 0 \\ 0 & \omega < 0 \end{cases} \quad (2.68)$$

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

$$\lim_{T \rightarrow \infty} \frac{\hbar\omega}{1 - e^{-\hbar\omega/k_B T}} = k_B T. \quad (2.69)$$

2.7.4 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

$$\hat{E}_j(\mathbf{r}, \omega) = \sum_k G_{jk}(\mathbf{r}, \mathbf{r}'; \omega) \hat{D}_k(\omega) \quad (2.70)$$

The FDT for the fluctuating fields is then given by

$$\langle \hat{E}_j(\mathbf{r}, \omega) \hat{E}_k^*(\mathbf{r}', \omega') \rangle = \frac{2\hbar}{1 - e^{-\hbar\omega/k_B T}} \text{Im}\{G_{jk}(\mathbf{r}, \mathbf{r}'; \omega)\} 2\pi\delta(\omega - \omega') \quad (2.71)$$

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.(2.71) coincides with the result of elementary field quantization.

The advantage of Eq.(2.71) 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.(2.66, ??). The FDT (2.71) 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.(2.66). 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 (2.71). For a more detailed discussion, see Henry & Kazarinov (1996); Eckhardt (1984).

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