

Chapter 2

Spectra of Radiating Systems

We have already noted that a possible way of measuring fluctuations of the field amplitudes is to detect the intensity of the radiation field emitted by an ensemble of atoms. More useful is the power spectrum, which is attributed to fluctuations of the field amplitudes and gives variations in the field intensity as a function of frequency. As demonstrated in the previous chapter, the calculation of the power spectrum requires the knowledge of the normally-ordered two-time correlation function of the electric field amplitudes evaluated at the position of a detector. In this chapter, we shall take a closer look at the relation of the spectrum to the correlation functions of the source variables and undertake a more detailed discussion of the calculation of the spectrum of radiation field emitted from the atoms. We shall be dealing entirely with power spectra of stationary and quasistationary fields, i.e. fields whose properties are independent of the origin of time. Under this condition, all the two-time correlation functions will involve components that depend on the time arguments only through their difference. Also, we shall follow the definition (1.70) of the power spectrum, which employs a simplified physical model of the detection process. In this model, the radiation field is measured with an ideal Fabry–Perot interferometer of bandwidth $\Gamma \rightarrow 0$, and its spectral range much larger than the width of the frequency spectrum of the measured field. The results presented here, however, can be easily extended to include finite bandwidth effects of the interferometer.

2.1 Emission Power Spectrum

Let us begin by considering the normal and time-ordered second-order correlation function of the electric field amplitudes evaluated at the position of a photodetector. As a consequence of the arrangements (1.85) and (1.86), we observe that the correlation function of the field amplitudes at a suitably located detector can be completely determined by the normally ordered correlation function of the source field amplitudes. Under these conditions and for the case of atomic source of the radiation field, we can apply the result (1.31) and write the correlation function of the source field

amplitudes as a multiple sum of terms involving correlation functions of the atomic dipole operators. The normal and time-ordered correlation function of the electric field amplitudes at the photodetector can then be written in the form as follows:

$$\begin{aligned} \langle \hat{\mathbf{E}}^{(-)}(t) \cdot \hat{\mathbf{E}}^{(+)}(t') \rangle &= \langle \hat{\mathbf{E}}_s^{(-)}(t) \cdot \hat{\mathbf{E}}_s^{(+)}(t') \rangle \\ &= \sum_{n,m=1}^N \sum_{i,k>j,l} \Psi_{ij}^*(\mathbf{r} - \mathbf{R}_n) \cdot \Psi_{lk}(\mathbf{r} - \mathbf{R}_m) \langle \hat{\Lambda}_{ij}^n(t) \hat{\Lambda}_{lk}^m(t') \rangle e^{ik_0 \bar{\mathbf{r}} \cdot \mathbf{R}_{nm}}. \end{aligned} \quad (2.1)$$

By taking the double truncated Fourier transform of the two-time correlation function (2.1) and the limit of long detection time, we obtain an expression for the stationary power spectrum of the field radiated per unit solid angle and measured at a point \mathbf{r} in the radiation zone

$$\begin{aligned} S(\mathbf{r}, \omega) &= \lim_{T \rightarrow \infty} \frac{1}{T} \sum_{n,m=1}^N \sum_{i,k>j,l} \Phi_{ijkl}^{nm} e^{ik_0 \bar{\mathbf{r}} \cdot \mathbf{R}_{nm}} \\ &\quad \times \int_0^T dt \int_0^T dt' \langle \hat{\Lambda}_{ij}^n(t) \hat{\Lambda}_{lk}^m(t') \rangle e^{i\omega(t-t')}, \end{aligned} \quad (2.2)$$

where Φ_{ijkl}^{nm} are the effective initial radiation rates defined in (1.96). This general expression shows that the spectrum of the radiation field emitted by an arbitrary atomic system can be described as having its origin in the correlation functions of the atomic dipole operators. Thus, our concern with the calculation of the spectrum is then with the evaluation of the two-time atomic correlation function. This two-time correlation function is obtained by superposing contributions from different atoms and different atomic transitions together with possible interference terms involving dipole transitions in different atoms and/or different transitions in the same atom.

Usually the source field is composed of a coherent component, corresponding to a field elastically scattered by the source atoms, and an incoherent (noise) component, corresponding to a field produced by fluctuations of the atomic dipoles. In practice this will almost certainly be the case if the source atoms are driven by a coherent field. Therefore, it is often necessary to distinguish between these two contributions to the source field. It is accomplished by expressing the atomic dipole operators $\hat{\Lambda}_{ij}^n(t)$ as the sum of its expectation value $\langle \hat{\Lambda}_{ij}^n(t) \rangle$ and its fluctuations $\delta \hat{\Lambda}_{ij}^n(t)$, so that

$$\hat{\Lambda}_{ij}^n(t) = \langle \hat{\Lambda}_{ij}^n(t) \rangle + \delta \hat{\Lambda}_{ij}^n(t), \quad (2.3)$$

with $\langle \delta \hat{\Lambda}_{ij}^n(t) \rangle = 0$. Accordingly, we can write the atomic correlation function, appearing in (2.2), as a sum of two qualitatively different parts

$$\langle \hat{\Lambda}_{ij}^n(t) \hat{\Lambda}_{lk}^m(t') \rangle = \langle \hat{\Lambda}_{ij}^n(t) \rangle \langle \hat{\Lambda}_{lk}^m(t') \rangle + \langle \delta \hat{\Lambda}_{ij}^n(t) \delta \hat{\Lambda}_{lk}^m(t') \rangle. \quad (2.4)$$

The first term on the right-hand side of (2.4) is given by the product of the expectation values of the atomic dipole operators and we therefore refer to it as the coherent part of the oscillating atomic dipole moments. The second term is associated with the fluctuations of the atomic dipole moments produced by the coupling of the atoms to the vacuum field. Thus, the effects of the vacuum fluctuations can be reflected through spectral properties of the radiated field, and can be clearly distinguished from coherent effects.

With the result (2.4), we may then clearly resolve the spectrum into two parts. On substituting (2.4) into (2.2), we find that the contribution of the mean dipole operators gives the coherent part of the spectrum

$$S_{\text{coh}}(\mathbf{r}, \omega) = \lim_{T \rightarrow \infty} \frac{1}{T} \sum_{n,m=1}^N \sum_{i,k>j,l} \Phi_{ijkl}^{nm} e^{ik_0 \bar{\mathbf{r}} \cdot \mathbf{R}_{nm}} \times \int_0^T dt \int_0^T dt' \langle \hat{A}_{ij}^n(t) \rangle \langle \hat{A}_{lk}^m(t') \rangle e^{i\omega(t-t')}, \quad (2.5)$$

and the contribution of the quantum fluctuations or quantum noise of the atomic dipoles gives the incoherent part of the spectrum

$$S_{\text{in}}(\mathbf{r}, \omega) = \lim_{T \rightarrow \infty} \frac{1}{T} \sum_{n,m=1}^N \sum_{i,k>j,l} \Phi_{ijkl}^{nm} e^{ik_0 \bar{\mathbf{r}} \cdot \mathbf{R}_{nm}} \times \int_0^T dt \int_0^T dt' \langle \delta \hat{A}_{ij}^n(t) \delta \hat{A}_{lk}^m(t') \rangle e^{i\omega(t-t')}. \quad (2.6)$$

From (2.5) and (2.6) it is apparent that the calculation of the coherent part of the spectrum only involves expectation values of the atomic transition operators, whereas the calculation of the incoherent part of the spectrum requires the knowledge of the two-time correlation function of the fluctuation operators. In an alternative way, one can calculate the incoherent part by subtracting the coherent part from the power spectrum $S(\mathbf{r}, \omega)$. To go further with the evaluation of the correlation functions and thereby the power spectrum, requires the knowledge of an explicit model of the source atoms.

There are a number of theoretical approaches that can be used to calculate the two-time correlation function of the atomic operators. A common method, which we shall be using in the succeeding chapters of the book, involves an application of the master equation describing the evolution of an atomic system and the quantum regression theorem [1]. We shall present some details of this method in Chap. 3, where we calculate and analyse power spectra for the specific source consisting of a single atom.

For the sake of completeness, let us briefly consider the relation between the stationary power spectrum, which is the frequency composition of the emitted field, and the intensity of the entire field emitted by the atoms into an element of solid

angle $d\Omega_r$ around the direction \mathbf{r} . By integrating (2.5) and (2.6) over the spectral frequency ω we obtain, respectively, the coherent and incoherent contributions to the radiation intensity of the emitted field. Since ω only appears in the exponential, the integration in the limit of the detection time $T \rightarrow \infty$ gives the delta function $\delta(t - t')$ and thus the two-time correlation functions reduce to single-time expectation values. The pertinent formula for the stationary value of the coherent part of the radiation intensity, per unit solid angle, is

$$\begin{aligned} \langle \hat{I}(\mathbf{r}) \rangle_{\text{coh}} &= \int_{-\infty}^{+\infty} d\omega S_{\text{coh}}(\mathbf{r}, \omega) = \sum_{n,m=1}^N \sum_{i,k>j,l} \Phi_{ijkl}^{nm} e^{ik_0 \bar{\mathbf{r}} \cdot \mathbf{R}_{nm}} \\ &\times \lim_{T \rightarrow \infty} \frac{1}{T} \int_0^T dt \langle \hat{A}_{ij}^n(t) \rangle \langle \hat{A}_{lk}^m(t) \rangle, \end{aligned} \quad (2.7)$$

and for the incoherent part is given by

$$\begin{aligned} \langle \hat{I}(\mathbf{r}) \rangle_{\text{in}} &= \int_{-\infty}^{+\infty} d\omega S_{\text{in}}(\mathbf{r}, \omega) = \sum_{n,m=1}^N \sum_{i,k>j,l} \Phi_{ijkl}^{nm} e^{ik_0 \bar{\mathbf{r}} \cdot \mathbf{R}_{nm}} \\ &\times \lim_{T \rightarrow \infty} \frac{1}{T} \int_0^T dt \langle \delta \hat{A}_{ij}^n(t) \delta \hat{A}_{lk}^m(t) \rangle. \end{aligned} \quad (2.8)$$

These equations express the stationary intensity of the emitted field in terms of expectation values and one-time correlation functions of the various transition operators. For the incoherent part populations of the atomic states are involved, since expanding the summation over the atomic energy states yields

$$\sum_{i,k>j} \langle \hat{A}_{ij}^n(t) \hat{A}_{jk}^n(t) \rangle = \sum_{i>j} \langle \hat{A}_{ii}^n(t) \rangle + \sum_{i \neq k > j} \langle \hat{A}_{ij}^n(t) \hat{A}_{jk}^n(t) \rangle, \quad (2.9)$$

where $\langle \hat{A}_{ii}^n(t) \rangle = \varrho_{ii}^n$ are populations of the atomic excited states $|i_n\rangle$. Accordingly, the single-atom ($n = m$) contributions to the intensity come from populations of the atomic states and coherences between different atomic transitions. Multiatom contributions with $n \neq m$ cannot be expressed in terms of populations of the single-atom states. They can rather be expressed in term of populations of entangled states of the multiatom system.

2.1.1 Coherent (Elastic) Part of the Spectrum

In the preceding section, we have shown that it is possible to separate the power spectrum into the coherent and incoherent components. It is accomplished by first splitting the two-time correlation function of the atomic operators into coherent

and noise parts. The spectrum is then derived as a sum of the double truncated Fourier transforms of the two parts of the correlation function. We now focus on the coherent part of the spectrum and, for later convenience, we shall introduce a certain simplification which is applicable to an arbitrary atomic system. We shall then find that the stationary coherent spectrum is composed of an infinitely narrow line, represented by a Dirac δ function, or a series of infinitely narrow lines.

Let us consider the relation (2.5) to examine the coherent spectrum when the source field is stationary in time. If the source atoms are driven by an external field, the induced atomic dipole moments continuously oscillate in time at the frequency of the driving field. As far as atoms are concerned, radiative processes such as spontaneous radiative decay, are very slow requiring on the average many millions of cycles of dipole oscillations before they are completed. Therefore, we can extract from the atomic transition operators the rapidly oscillating terms, and write their expectation values as

$$\begin{aligned}\langle \hat{A}_{ij}^n(t) \rangle_s &= \langle \hat{A}_{ij}^n(t) \rangle e^{-i\omega_L t}, \\ \langle \hat{A}_{ji}^n(t) \rangle_s &= \langle \hat{A}_{ji}^n(t) \rangle e^{i\omega_L t}, \quad i > j,\end{aligned}\quad (2.10)$$

where ω_L is the frequency of the driving field, which does not necessarily coincide with the atomic transition frequencies ω_{ij} , and $\langle \hat{A}_{ij}^n(t) \rangle_s$ are expectation values of slowly varying transition operators, which change slowly compared with variations arising from the periodic term $\exp(i\omega_L t)$.

Written in terms of the slowly varying expectation values, the coherent part of the spectrum (2.5) becomes

$$\begin{aligned}S_{\text{coh}}(\mathbf{r}, \omega) &= \lim_{T \rightarrow \infty} \frac{1}{T} \sum_{n,m=1}^N \sum_{i,k>j,l} \Phi_{ijlk}^{nm} e^{ik_0 \bar{\mathbf{r}} \cdot \mathbf{R}_{nm}} \\ &\times \int_0^T dt \int_0^T dt' \langle \hat{A}_{ij}^n(t) \rangle_s \langle \hat{A}_{lk}^m(t') \rangle_s e^{i(\omega - \omega_L)(t-t')}. \quad (2.11)\end{aligned}$$

Equation (2.10) shows directly that the atomic dipole moments continue to oscillate in time at the driving frequency and, even after a long time, the expectation values cannot be strictly stationary. However, we may assume that a stationary state is reached after a long time, in the sense that $\langle \hat{A}_{ij}^n(t) \rangle_s$ are independent of t , and each of the expectation values in (2.11) becomes independent of time also. If it applies, the expectation values of the slowly varying operators can be replaced by their stationary, time-independent values.

It is then possible to perform the integration explicitly by extracting the time-independent expectation values from the integrals. Provided $T \rightarrow \infty$, the double integral can be well approximated by a Dirac δ function, and then the coherent spectrum (2.11) simplifies to

$$S_{\text{coh}}(\mathbf{r}, \omega) = \sum_{n,m=1}^N \sum_{i,k>j,l} \Phi_{ijkl}^{nm}(\mathbf{r}) \langle \hat{A}_{ij}^n \rangle_s \langle \hat{A}_{lk}^m \rangle_s \pi \delta(\omega - \omega_L) , \quad (2.12)$$

where $\Phi_{ijkl}^{nm}(\mathbf{r}) = \Phi_{ijkl}^{nm} \exp(ik_0 \bar{\mathbf{r}} \cdot \mathbf{R}_{nm})$ and $\langle \hat{A}_{ij}^n \rangle_s$ are stationary coherences, i.e., the time-independent parts of the average atomic transition operators.

The expression (2.12) shows that the coherent spectrum of a stationary field emitted by an atomic source consists of a delta function contribution giving an infinitely narrow line at the frequency of the driving field.¹ It obviously corresponds to a field that is elastically scattered by the atomic system. The intensity of the line depends on magnitudes of the stationary atomic coherences. We should mention that in experimental practice, the exact delta line would not be observed. It would rather show up as a peak of a finite width imposed by the finite spectral resolution of the detecting apparatus.

The procedure of calculating the coherent spectrum is more complicated if the system is not strictly stationary, but rather is in a quasistationary state, even in the long-time limit. The qualifying term *quasistationary* means that the atomic quantities are explicit function of time. In this case, the above formalism has to be modified. Usually, the time dependence arises from nonzero detunings of the driving field frequency from the atomic transition frequencies. It may also arise from a time modulation of the driving field frequency or amplitude. In general, the time dependence may be quite complicated and involve many oscillatory contributions. However, in many practical cases, it occurs that the system oscillates harmonically with a frequency determined by a single parameter δ . The problem is then solved by applying the Floquet method that enables to obtain a formal analytic expression for the stationary part of the time-dependent expectation value $\langle \hat{A}_{ij}^n(t) \rangle_s$. It is done by expanding the time-dependent expectation values into a sum of contributions oscillating at the frequency δ and its harmonics, by means of a Fourier series

$$\langle \hat{A}_{ij}^n(t) \rangle_s = \sum_{p=-\infty}^{\infty} \langle \hat{A}_{ij}^n \rangle_s^{(p)} e^{ip\delta t} , \quad (2.13)$$

where p is an integer which determines the order of the harmonics, $\langle \hat{A}_{ij}^n \rangle_s^{(p)}$ are the expansion coefficients that vary slowly with time, and δ is an oscillation (modulation) parameter. As we have already mentioned, the parameter δ can be associated with a detuning of the driving field from atomic resonances or with a time modulation of the driving field frequency and/or amplitude. In the case of a continuous pulse-train driving field, the parameter δ is given by the pulse repetition frequency.

Now that we have determined the method of the explicit treatment of the time-dependent correlation functions, we expand each of the expectation values appearing in (2.11) into a series of slowly varying amplitudes that oscillate at the frequency δ and its harmonics, and arrive at the expression

¹The delta function occurs only for an infinite observation time. In practice, for a finite observation time T , the width of the contribution is of order T^{-1} .

$$\begin{aligned}
S_{\text{coh}}(\mathbf{r}, \omega) = & \lim_{T \rightarrow \infty} \frac{1}{T} \sum_{n,m=1}^N \sum_{i,k>j,l} \Phi_{ijkl}^{nm}(\mathbf{r}) \sum_{p,r=-\infty}^{\infty} \int_0^T dt \int_0^T dt' \\
& \times \langle \hat{A}_{ij}^n \rangle_s^{(p)} \langle \hat{A}_{lk}^m \rangle_s^{(r)} e^{i(p+r)\delta t} e^{i(\omega - \omega_L - r\delta)(t-t')}. \quad (2.14)
\end{aligned}$$

The spectrum generally consists of terms which depend on time t and the time difference $t - t'$. For a fixed $t - t'$ and for $p \neq -r$, we see that the time dependent factor $\exp[i(p+r)\delta t]$ continuously oscillate in time and becomes negligible after a sufficiently long time t . Thus, in the long-time limit, the terms with $p \neq -r$ make a negligible contribution to the spectrum. There are however “diagonal” terms involving harmonics with $p = -r$. These terms are independent of t , and therefore make a significant contribution to the spectrum even in the long-time limit. This shows that the system eventually settles in a stationary state. Hence, for a sufficiently long detection time T the resultant “stationary” coherent spectrum is given by

$$\begin{aligned}
S_{\text{coh}}(\mathbf{r}, \omega) = & \sum_{n,m=1}^N \sum_{i,k>j,l} \Phi_{ijkl}^{nm}(\mathbf{r}) \\
& \times \sum_{r=-\infty}^{\infty} \langle \hat{A}_{ij}^n \rangle_s^{(-r)} \langle \hat{A}_{lk}^m \rangle_s^{(r)} \pi \delta(\omega - \omega_L - r\delta). \quad (2.15)
\end{aligned}$$

From this we see that the spectral distribution of the coherently scattered field differs from that of the driving field. It exhibits a series of infinitely sharp peaks located at frequencies $r\delta$ separated by a constant spacing δ . This implies that the coherent scattering on a quasistationary system is not in general elastic. We may conclude that a periodic time modulation of the expectation values introduces new infinitely sharp components at multiples of the modulation frequency δ . The new components reflect the presence of parametric resonances, which are due to the oscillations introduced into the atomic dipole moments at harmonic frequencies $\omega_L + r\delta$. The intensity of a given coherent peak depends on the properties of the stationary Fourier amplitudes of the atomic coherences, whose explicit forms are normally determined from the master equation of a specific atomic system.

As we have already stated, the coherent spectrum is composed of infinitely sharp peaks. Nevertheless, the coherent part of the radiation intensity, obtained after integration of (2.15) over all spectral frequencies, is finite and proportional to the stationary harmonics of the average atomic coherences.

In summary of this section, the expressions (2.12) and (2.15) are our formal results for the coherent part of the power spectrum of a radiation field emitted by stationary and quasistationary systems. The results are instructive because they show how the expectation values of the atomic transition operators and their stationary components enter the spectrum in a simple explicit way. They have simple mathematical structures and allow for a particularly transparent physical interpretation of the coherent spectrum. We emphasize the complete generality of these equations that they are applicable to arbitrary atomic systems and hold for any initial state of the system.

2.1.2 Incoherent (Noise) Part of the Spectrum

Thus far our discussion has been limited to the coherent part of the power spectrum. However, more important is the incoherent part. It arises from quantum fluctuations both of the field and of the atomic source and therefore its measurement may be thought as a method of their detection. Earlier in this chapter, we have defined the incoherent part of the spectrum as a double Fourier transform of the two-time correlation function of the fluctuation operators. Before proceeding further with an examination of the incoherent spectrum for specific atomic systems, we stop for a moment to introduce certain general simplifications to the procedure of calculating the spectrum, which are common for all atomic systems. To be specific, we shall introduce the simplifications to the general form (2.6) of the incoherent part of the power spectrum without a consideration of detailed atomic dynamics.

When dealing with the coherent spectrum it was convenient to introduce slowly oscillating operators that are free from the rapid oscillations at optical frequencies characterizing the atomic operators. In analogy, the common rapid oscillations at the optical frequency ω_L can be removed from the fluctuation operators by transforming to slowly varying operators

$$\delta S_{ij}^n(t) = \delta \hat{A}_{ij}^n(t) e^{-i\omega_L t}, \quad \delta S_{ji}^n(t) = \delta \hat{A}_{ji}^n(t) e^{i\omega_L t}, \quad i > j, \quad (2.16)$$

where, as before, ω_L is the frequency of the driving field. In terms of these new operators, the incoherent part of the power spectrum becomes

$$\begin{aligned} S_{\text{in}}(\mathbf{r}, \omega) = & \lim_{T \rightarrow \infty} \frac{1}{T} \sum_{n,m=1}^N \sum_{i,k>j,l} \Phi_{ijkl}^{nm} e^{ik_0 \bar{\mathbf{r}} \cdot \mathbf{R}_{nm}} \\ & \times \int_0^T dt \int_0^T dt' \langle \delta S_{ij}^n(t) \delta S_{lk}^m(t') \rangle e^{i(\omega - \omega_L)(t-t')}. \end{aligned} \quad (2.17)$$

Although the above expression is rather simple in form, the integrals cannot be evaluated before a detailed solution for the two-time correlation function of the slowly varying operators has been obtained. However, there are two situations in which this formula can further be simplified without a consideration of the solution for the correlation function. The first is the case of steady or stationary systems for which the correlation function depends on the time arguments only through their difference $t' - t$. From this property and employing the basic expression (2.17), giving the incoherent spectrum for a long detection time, it follows that by taking $T \rightarrow \infty$ and by changing the variables of integration, the t integral in (2.17) can be carried out explicitly and then the expression for the incoherent spectrum simplifies to

$$S_{\text{in}}(\mathbf{r}, \omega) = 2\text{Re} \sum_{n,m=1}^N \sum_{i,k>j,l} \Phi_{ijkl}^{nm}(\mathbf{r}) \times \int_0^\infty d\tau \langle \delta S_{ij}^n(0) \delta S_{lk}^m(\tau) \rangle e^{i(\omega - \omega_L)\tau}, \quad (2.18)$$

where $\tau = t - t'$ is the delay time, and we have shifted the time origin to $t = 0$ as in the long-time limit the correlation function is independent of the time origin.

Integrating (2.18) over all directions, specified by the solid angles $d\Omega_r$, we obtain the incoherent spectrum of the total radiation field per unit time

$$S_{\text{in}}(\omega) = \int d\Omega_r S_{\text{in}}(\mathbf{r}, \omega) = 2\text{Re} \sum_{n,m=1}^N \sum_{i,k>j,l} \sqrt{\gamma_{ij}\gamma_{kl}} \cos \theta_{ik} \times \int_0^\infty d\tau \langle \delta S_{ij}^n(0) \delta S_{lk}^m(\tau) \rangle e^{i(\omega - \omega_L)\tau}. \quad (2.19)$$

The expression (2.19) is one of the fundamental equations in atomic spectroscopy. It is used to determine spectral features of the radiation field emitted by an arbitrary stationary system. This general property makes this expression extremely flexible in that it may be applied in various situations ranging from a single two-level atom to multiatom and multi-level systems in arbitrary spatial distributions and in arbitrary configurations of the energy levels. As we shall explore in this book, the expression (2.19) may also be used to obtain an information about certain quantum features of the field.

This is as far as we can get with the calculation of the stationary spectrum without an explicit calculation of the atomic correlation function. To go further requires a master equation treatment of the stationary atomic correlation function and the quantum regression theorem. Briefly, for a given system, an explicit form of the correlation function can be established from the master equation by deriving a set of coupled differential equations for the density matrix elements and solving it under the stationary condition. Having computed the correlation function, the integration can be performed from which the spectrum of the emitted field may be readily determined for a specific direction of observation \mathbf{r} . It is worth noting that depending on whether the correlation function is real or complex, the spectrum can be symmetric or asymmetric about its line center, $\omega = \omega_L$. If the correlation function is real, the incoherent spectrum will be symmetric about its line centre, and will be asymmetric if it is not. These points will be further elaborated in the course of specific examples discussed in the next chapter.

Now let us examine the incoherent spectrum for the second situation in which the system is in a quasistationary state. In this case, the two-time correlation function depends explicitly on time t , and we shall not find it possible to proceed with the calculation of the spectrum in quite such a straightforward manner as before for the stationary case. Actually, we have already considered this kind of problem when we have calculated the coherent spectrum of a quasistationary field oscillating slowly

with some frequency δ characterizing the radiating system. We have seen that the temporal oscillations of the one-time atomic variables significantly altered the nature of the coherent field. In this case, it was appropriate to treat the spectrum by employing a Fourier decomposition of the atomic variables in terms of the frequency δ and its harmonics. For the incoherent spectrum we might expect that a similar procedure could be used. This is true; however, we shall see shortly that the calculation of the incoherent spectrum is more complicated than the evolution of the simple expectation values of the atomic operators. It involves two-time correlation functions and, in general, may require a double Fourier decomposition.

Suppose we consider a two-time correlation function $Y(t, t')$, which has a periodic dependence on both t and t' , with a period of oscillation δ . Now because the correlation function depends on two different times, we may introduce a double Fourier decomposition taken with respect to the same variable δ . Consequently, the two-time correlation function may be written as

$$\begin{aligned} Y(t, t') &= \sum_{p=-\infty}^{\infty} \sum_{r=-\infty}^{\infty} Y^{(p,r)}(t, t') e^{ip\delta t} e^{ir\delta t'} \\ &= \sum_{p=-\infty}^{\infty} \sum_{r=-\infty}^{\infty} Y^{(p,r)}(t, t + \tau) e^{i(p+r)\delta t} e^{ir\delta \tau}, \end{aligned} \quad (2.20)$$

where $Y^{(p,r)}(t, t')$ are two-dimensional slowly varying Fourier amplitudes and, as before, $\tau = t' - t$ is the delay time.

We call the above expansion the double Fourier expansion simply because it is the product of two individual Fourier series, one in time t and the other in time t' , or in the delay time τ . From (2.20), we see that the correlation function is periodic in both t and τ , and responds at the fundamental frequency δ and its harmonics. In the long-time limit of $t \rightarrow \infty$, the amplitudes $Y^{(p,r)}(t, \tau)$ do not vary with time, that is $dY^{(p,r)}(t, \tau)/dt \approx 0$, and then the Fourier amplitudes become independent of the first argument yielding $Y^{(p,r)}(t, \tau) = Y^{(p,r)}(0, \tau)$. In addition, the t dependent exponential factors cause all terms with $r \neq -p$ to vanish as $t \rightarrow \infty$, except the terms with $r = -p$ for which the factors reduce to unity. Hence, in the limit of long times t , the double Fourier expansion reduces to

$$Y(\tau) = \lim_{t \rightarrow \infty} Y(t, t + \tau) = \sum_{p=-\infty}^{\infty} Y^{(p,-p)}(0, \tau) e^{-ip\delta \tau}. \quad (2.21)$$

First note that in the stationary limit, in which the two-time correlation function depends only on the time difference τ , the double Fourier expansion reduces to a simpler “diagonal” form. This expansion also tells us that in the stationary limit a system described by the correlation function $Y(t, t + \tau)$ will respond at the fundamental frequency δ at its harmonics. The response at the harmonics is what makes

the quasistationary system different from a stationary system, which responses only at the fundamental frequency δ .

To see how the periodic oscillations of the correlation function affect the incoherent spectrum, we apply the double Fourier decomposition to the two-time correlation function appearing in (2.17), and arrive at the expression

$$S_{\text{in}}(\mathbf{r}, \omega) = 2\text{Re} \sum_{n,m=1}^N \sum_{i,k>j,l} \Phi_{ijkl}^{nm} e^{ik_0 \bar{\mathbf{r}} \cdot \mathbf{R}_{nm}} \sum_{p=-\infty}^{\infty} \int_0^{\infty} d\tau \times \langle \delta S_{ij}^n(0) \delta S_{lk}^m(\tau) \rangle^{(p,-p)} e^{i(\omega - \omega_L - p\delta)\tau}. \quad (2.22)$$

Equation (2.22) gives the stationary part of the incoherent spectrum in terms of a two-dimensional array of functions. The most interesting feature of the spectrum is the appearance of resonances at harmonics of δ . This feature is not encountered for spectra of the radiation field emitted by a stationary system. Of course, the explicit form of the spectrum and its resonant structure depends crucially on the two-dimensional amplitudes. Once the amplitudes are known, the spectrum can be evaluated to any desired accuracy simply by performing the integration over τ . However, without further calculations two distinctly different types of spectra could be observed depending on whether the diagonal amplitudes are real or not. If the amplitudes are real, the spectrum will be symmetric about its line center, $\omega = \omega_L$. On the other hand, if the diagonal amplitudes are complex, the spectrum will be asymmetric and the imaginary parts of the amplitudes may lead to additional spectral lines or shifts of the harmonic resonances. Consequently, the spectrum can have quite different characteristics, which depend on the specific atomic system.

The method of the double Fourier decomposition, straightforward as it is, becomes cumbersome even in relatively simple problems because of the necessity of dealing with a two-dimensional array of functions. However, there is an equivalent alternative method of treating time dependence of two-time correlation functions, which in many cases is much easier to apply to the calculation of a stationary spectrum. This alternative method consists essentially in evaluating a stationary correlation function in terms of a single Fourier decomposition. For example, if the correlation function $Y(t, t + \tau)$, or its time evolution, depends only on the second argument $(t + \tau)$, we may expand $Y(t, t + \tau)$ into a Fourier series with respect to the time $t + \tau$ as

$$Y(t, t + \tau) = \sum_{p=-\infty}^{\infty} Y^{(p)}(\tau) e^{ip\delta(t+\tau)}. \quad (2.23)$$

Although this expansion differs from (2.20), we emphasize that (2.23) by itself is a perfectly valid mathematical expansion: unknown function Y can always be expressed in terms of a family of unknown functions $Y^{(p)}$. The procedure recognizes that we are actually interested only in the later (delay) time of the two-time correlation. The major advantage of using the decomposition (2.23) over (2.20) is that the real part of the Fourier transform of the stationary harmonic amplitude $Y^{(0)}$ gives

the stationary spectrum of the field emitted by the system described by the correlation function Y . This statement is readily verified by applying the above analysis to the correlation function appearing in (2.17). When the one-time expansion (2.23) is used in (2.17), and the integrals are expressed in terms of time t and time difference $\tau = t' - t$, we readily arrive at the following incoherent spectrum

$$S_{\text{in}}(\mathbf{r}, \omega) = \lim_{T \rightarrow \infty} \frac{1}{T} \sum_{n,m=1}^N \sum_{i,k>j,l} \Phi_{ijkl}^{nm} e^{ik_0 \bar{\mathbf{r}} \cdot \mathbf{R}_{nm}} \sum_{p=-\infty}^{\infty} \int_0^T dt \int_0^t d\tau \times \langle \delta S_{ij}^n(t) \delta S_{lk}^m(t + \tau) \rangle^{(p)} e^{ip\delta(t+\tau)} e^{i(\omega - \omega_L)\tau}. \quad (2.24)$$

The incoherent spectrum we have found is determined by the Fourier amplitudes which are accompanied by t -dependent exponential factors oscillating at harmonics of δ . Because of these factors, when the integration over the long detection time is performed, the terms with $p \neq 0$ make a negligible contribution to the spectrum except of the term with $p = 0$, which remains constant as $t \rightarrow \infty$. Accordingly, we find directly from (2.24) that the long-time spectrum is determined essentially by the temporally stationary harmonic and reaches a stationary value given by

$$S_{\text{in}}(\mathbf{r}, \omega) = 2\text{Re} \sum_{n,m=1}^N \sum_{i,k>j,l} \Phi_{ijkl}^{nm} e^{ik_0 \bar{\mathbf{r}} \cdot \mathbf{R}_{nm}} \times \int_0^{\infty} d\tau \langle \delta S_{ij}^n(0) \delta S_{lk}^m(\tau) \rangle^{(0)} e^{i(\omega - \omega_L)\tau}. \quad (2.25)$$

This simplified expression determines the stationary component of the incoherent spectrum of a quasistationary field emitted by an atomic system. It holds for an arbitrary quasistationary state of the system, and is applicable when the time dependence of the atomic correlation function can be expressed in terms of a single frequency parameter δ characterizing the system. Integrating (2.25) over all directions, we obtain the incoherent spectrum of the total radiation field per unit time

$$S_{\text{in}}(\omega) = \int d\Omega_{\mathbf{r}} S_{\text{in}}(\mathbf{r}, \omega) = 2\text{Re} \sum_{n,m=1}^N \sum_{i,k>j,l} \sqrt{\gamma_{ij}\gamma_{kl}} \cos \theta_{ik} \times \int_0^{\infty} d\tau \langle \delta S_{ij}^n(0) \delta S_{lk}^m(\tau) \rangle^{(0)} e^{i(\omega - \omega_L)\tau}. \quad (2.26)$$

The expression (2.26) will be extensively used in our study of spectral and quantum properties of the field emitted by quasistationary atomic systems. Although the parameter δ does not appear explicitly in the above expression for the stationary spectrum, it certainly will appear in explicit solutions for the time dependent harmonic of the atomic correlation function. As we shall see, the evaluation of the stationary harmonic requires the knowledge of the initial (stationary) values of the correlations, which are given in terms of single-time expectation values of the atomic operators.

They are obtained through another single Fourier decomposition, and then the resultant incoherent spectrum is to be thought of as a superposition of different harmonics of the single-time expectation values. The procedure just described and its applications for evaluation of incoherent spectra of radiation field emitted by specific atomic systems will be discussed in details in the next chapter.

To summarize, we have seen how one may simplify the calculations of incoherent spectra of the radiation field emitted by stationary and quasistationary systems without an explicit calculation of the two-time atomic correlation function. Problems involving stationary fields can be solved by straightforward application of the Fourier transform of the two-time correlation function of slowly varying parts of the atomic operators. In the case of quasistationary fields, the two-time correlation function is properly analyzed by a Fourier expansion, and there are two equivalent approaches available, which allow to study the spectrum analytically. For one, we employ a double Fourier decomposition taken with respect to the same variable characterizing the time dependence of a given system. In the second approach, the spectrum is obtained by employing a more direct approach, utilizing a single Fourier decomposition, which greatly simplifies the calculations. Once the time-dependence of the correlation function is known, it is only a matter of performing the integration over τ to derive the spectrum. It is worth noting, finally, that in general, when the problem involves quasistationary fields whose the time dependence is represented by several different parameters, it is not feasible to attempt a decomposition of the correlation function into a multi-dimensional Fourier series. The analytic description of such a problem is extremely difficult, it may result in multi-dimensional arrays of Fourier amplitudes. In this case, the time evolution is best analysed by numerical methods.

2.2 Absorption Spectrum of a Probe Field

In atomic and molecular spectroscopy one is frequently confronted with a situation in which, in addition to the power (emission) spectrum, changes in the absorption spectrum of a probe beam irradiating an atomic system are observed. The study of the absorption spectrum offers an opportunity for further tests of the role of quantum fluctuations in atomic spectroscopy.

The absorption spectrum is defined as the rate of energy absorption from the probe beam as a function of its frequency. As we shall see, it may be expressed quite generally in terms of a certain atomic correlation function, which is evaluated in the absence of the probe beam. This results from the weak field approximation, in which the probe intensity is assumed to be sufficiently weak that it does not appreciably perturb the atomic system. In addition to weak probe absorption spectra, one can also calculate absorption spectra for a strong probe beam. These spectra are termed “strong probe spectra” and may be significantly different from the spectra obtained with weak probe. However, one may be confused by the use of a strong probe to monitor an atomic system. This requires a word of explanation. If an intense field is used to probe an atomic system, one can argue that what we would normally call

‘strong probe’ is in fact a contradiction in terms. To put it another way, a strong probe can be viewed as a driving field for the atomic system. In this case, the system should be probed by another weak field. Then, we expect to see qualitatively different properties of the absorption spectrum from those observed in the absence of the strong probe field.

Let us proceed to a more detailed discussion of the procedure of evaluating the absorption spectrum. We stress that there are various ways to evaluate the spectrum. They arise from different experimental arrangements to observe absorptive properties of an atomic system. The method of calculation, we explore in this section, is based on the approach proposed by Mollow [2, 3] although our treatment is more general in some respects. It is appropriate for experiments where the intensity of the radiation field emitted from an atomic system is measured separately in the presence and in the absence of the probe beam. The difference between the two intensities measured as a function of the probe frequency represents the net of absorption at this frequency.

Following closely the approach of Mollow, we may describe the absorption spectrum of a probe beam by first determining the change it produces in the evolution of a probed system. Suppose that an atomic system, described by the total dipole operator $\hat{\mu}$, is monitored by a weak single-mode classical field of a tunable frequency ω_p . If coupling of the atomic system to all other modes of the electromagnetic field is neglected, the interaction between the probe beam and the atomic system, in the electric dipole and rotating-wave approximations, is given by the Hamiltonian

$$\hat{H}(t) = \sum_n -\hat{\mu}^n \cdot \mathbf{E}_p(\mathbf{R}_n, t) = -i\hbar \sum_n \sum_{i>j} g_{ij}^n(\mathbf{R}_n, t) \hat{A}_{ij}^n(t) + \text{H.c.}, \quad (2.27)$$

where we have used the projection operator representation for the total dipole operator and the mode representation for the probe beam. In (2.27), the atoms are treated as quantum systems represented by the transition operators $\hat{A}_{ij}^n(t)$, whereas the probe beam is treated as a single-mode classical field of the amplitude

$$\begin{aligned} \mathcal{E}_p(\mathbf{r}, t) &= \sqrt{\frac{\hbar\omega_p}{2\varepsilon_0\mathcal{V}}} \alpha_p \bar{\mathbf{e}}_p e^{i(\mathbf{k}_p \cdot \mathbf{r} - \omega_p t)} \\ &= \mathcal{E}_p \bar{\mathbf{e}}_p e^{i(\mathbf{k}_p \cdot \mathbf{r} - \omega_p t)}, \end{aligned} \quad (2.28)$$

and the coupling constant between the atoms and the field is given in terms of the matrix elements of the atomic transition dipole moments

$$g_{ij}^n(\mathbf{R}_n, t) = \hat{\mu}_{ij}^n \cdot \mathcal{E}_p(\mathbf{R}_n, t) / \hbar. \quad (2.29)$$

Here, α_p is the complex mode amplitude, $\bar{\mathbf{e}}_p$ is the unit polarization vector of the probe field mode, and $\hat{\mu}_{ij}^n$ is an atomic transition dipole moment interacting with the probe field. Finally, \mathbf{k}_p represents the propagation vector of the field. We point out here, that the probe field is assumed to be tuned near atomic resonances, in the sense that the detunings of the probe field are small compared to optical frequencies,

($|\omega_{ij} - \omega_p| \ll \omega_p$). Moreover, the choice of the classical description for the probe field has an advantage that it corresponds closely to a typical experimental situation in which atoms are probed by a coherent laser field.

With the coupling Hamiltonian (2.27), we are able to study the influence of a probe beam on the dynamics of the atomic system. It is conveniently studied in terms of the density operator describing the system, which obeys the Liouville–von Neumann equation

$$i\hbar \frac{\partial \varrho(t)}{\partial t} = [\hat{H}(t), \varrho(t)] . \quad (2.30)$$

Formally integrating (2.30) with respect to a measurable time interval, we find that to the lowest order in the probe beam amplitude the interaction (2.27) changes the density operator of the system by

$$\Delta \varrho(t) = \frac{1}{i\hbar} \int_{-\infty}^t dt' [\hat{H}(t'), \varrho(t)] , \quad (2.31)$$

where we have applied the Markov approximation in which we have replaced $\varrho(t')$ by $\varrho(t)$ under the integral. This is a reasonable approximation for our purposes here, since we look at the response of the system to a weak probe field. For this case, the probe absorption spectrum is determined by the rate at which the system changes linearly under the influence of the probe field [4]. It involves a trace of the time derivative of the interaction Hamiltonian over the internal states of the probed system

$$\begin{aligned} A(\omega_p, t) &= \text{Tr} \left[\frac{\partial \hat{H}(t)}{\partial t} \Delta \varrho(t) \right] \\ &= \frac{1}{i\hbar} \int_{-\infty}^t dt' \text{Tr} \left\{ \left[\frac{\partial \hat{H}(t)}{\partial t}, \hat{H}(t') \right] \varrho(t) \right\} . \end{aligned} \quad (2.32)$$

When the Hamiltonian (2.27) and its partial time derivative are used in (2.32), it is straightforward to show that in the long time limit the absorption spectrum of the weak field is given by the Fourier transform of the two-time commutator of the atomic dipole operators

$$\begin{aligned} A(\omega_p) &= \lim_{t \rightarrow \infty} A(\omega_p, t) = \sum_{n,m=1}^N \sum_{i>j} \sum_{k>l} (\hat{\mu}_{ij}^n \cdot \tilde{\mathcal{E}}_p) (\hat{\mu}_{kl}^m \cdot \tilde{\mathcal{E}}_p)^* \\ &\quad \times \int_{-\infty}^{\infty} d\tau [\hat{A}_{ji}^n, \hat{A}_{kl}^m(\tau)] e^{i\mathbf{k}_p \cdot \mathbf{R}_{nm}} e^{i\omega_p \tau} , \end{aligned} \quad (2.33)$$

where, as usual, $\mathbf{R}_{nm} = \mathbf{R}_n - \mathbf{R}_m$ is the vector distance between two atoms in the sample, and $\tilde{\mathcal{E}}_p = (2\omega_p \mathcal{E}_p / \hbar) \tilde{\mathbf{e}}_p$. In the derivation of (2.33), we have retained

only the slowly varying terms oscillating with the time difference $\tau = t - t'$ at the probe frequency ω_p . This simplification is obviously a form of the rotating-wave approximation, and for our purposes of a long-time absorption spectrum this is an excellent approximation.

The derivation of the absorption spectrum (2.33) has been kept very general in order to make it applicable to any system probed by a weak field. It is given in terms of a certain two-time atomic correlation function, which is quite different in form than the one which determines the power spectrum for the same system. The correlation function reflects absorptive as well as emissive processes which may take place when the system is probed by a weak field. Mathematically, the absorption spectrum is found by first calculating the expectation value of the commutator, and then taking the Fourier transform of the commutator with respect to τ . It is usually done using the master equation of the atomic system together with the quantum regression theorem. Note that the commutator is evaluated in the absence of the probe field, but the other fields such as the vacuum field and driving fields interacting with the atomic system are always present. The probe and driving fields can be tuned to the same atomic transitions, or alternatively, the probe can be tuned to an auxiliary level. In the first case a Mollow type, whereas in the second case an Autler–Townes type spectrum is monitored.

We recall that the expectation value of the commutator, appearing in (2.33), has an important interpretation in terms of directly measurable quantities. By definition, the commutator may be decomposed into two terms, and consequently we can write

$$\langle [\hat{A}_{ji}^n, \hat{A}_{kl}^m(\tau)] \rangle = \langle \hat{A}_{ji}^n \hat{A}_{kl}^m(\tau) \rangle - \langle \hat{A}_{kl}^m(\tau) \hat{A}_{ji}^n \rangle = A_1 - A_2. \quad (2.34)$$

The physical interpretation of the two terms on the right-hand side of (2.34) is as follows: The first term represents processes corresponding to absorption of the probe field by the atomic system. The second term is attributed to emissive processes corresponding to the stimulated emission into the probe field. The difference of these two terms yields the net absorption of the probe field, which is not necessarily positive. Indeed, when we combine the two terms, the resulting expectation value of the commutator may be positive, equal to zero, or negative. Physically, in the case of $A_1 > A_2$, the total number of absorption processes outweighs the total number of stimulated emission processes resulting in a positive absorption spectrum. Consequently, the probe field is absorbed by the system. In the case of $A_1 = A_2$, the number of absorption is equal to the number of stimulated emission processes resulting in zero net absorption of the probe field. For such a case one speaks of transparency of the system to the probe field. Finally, in the case of $A_1 < A_2$, the stimulated emission outweighs the absorption. Consequently, the absorption becomes negative, i.e. instead of being absorbed the probe field is amplified.

Amplification of a probe field usually requires an inversion of the atomic population on the probed atomic transition, but an amplification without population inversion is also possible. The later, however, requires more than two energy levels to be involved, or equivalently, at least two transition channels simultaneously monitored by the probe field. In this case, an amplification can be achieved through coherences

between the atomic levels which is traditionally understood as quantum interference between various transition channels in the probed system. If only two energy levels are involved, or equivalently the probe field is coupled to a single transition channel, such interference is impossible.

The role of population inversion and interference in the absorption process is seen more explicitly in the integrated absorption spectrum. When we integrate the absorption spectrum over all frequencies, we obtain the total absorption rate

$$\begin{aligned} \bar{A} = \int_{-\infty}^{\infty} d\omega_p A(\omega_p) &= 2\pi \sum_{n,m=1}^N \sum_{i>j} \sum_{k>l} (\hat{\mu}_{ij}^n \cdot \tilde{\mathcal{E}}_p)(\hat{\mu}_{kl}^m \cdot \tilde{\mathcal{E}}_p)^* \\ &\times (\langle \hat{A}_{ji}^n \hat{A}_{kl}^m \rangle - \langle \hat{A}_{kl}^m \hat{A}_{ji}^n \rangle) e^{i\mathbf{k}_p \cdot \mathbf{R}_{nm}}. \end{aligned} \quad (2.35)$$

If we separate the atomic correlation functions into single-atom and multiatom contributions, and further distinguish between populations and coherences in the single-atom terms, we arrive at the expression

$$\begin{aligned} \bar{A}/2\pi &= \sum_{n=1}^N \left\{ \sum_{i>j} |\hat{\mu}_{ij}^n \cdot \tilde{\mathcal{E}}_p|^2 (\langle \hat{A}_{jj}^n \rangle - \langle \hat{A}_{ii}^n \rangle) \right. \\ &\quad \left. + \sum_{i>j} \sum_{j \neq l} (\hat{\mu}_{ij}^n \cdot \tilde{\mathcal{E}}_p)(\hat{\mu}_{jl}^n \cdot \tilde{\mathcal{E}}_p)^* (\langle \hat{A}_{ji}^n \hat{A}_{il}^n \rangle - \langle \hat{A}_{lj}^n \hat{A}_{ji}^n \rangle) \right\} \\ &\quad + \sum_{n \neq m=1}^N \sum_{i>j} \sum_{k>l} (\hat{\mu}_{ij}^n \cdot \tilde{\mathcal{E}}_p)(\hat{\mu}_{kl}^m \cdot \tilde{\mathcal{E}}_p)^* \\ &\quad \times (\langle \hat{A}_{ji}^n \hat{A}_{kl}^m \rangle - \langle \hat{A}_{kl}^m \hat{A}_{ji}^n \rangle) e^{i\mathbf{k}_p \cdot \mathbf{R}_{nm}}. \end{aligned} \quad (2.36)$$

From the above expression, we see that the total absorption rate depends crucially on the population distribution and the coherences between the atomic levels. The first term is the contribution resulting from the difference between populations of the lower and upper levels in a given transition. The second term, proportional to the coherences between various atomic transitions, corresponds to absorption induced by quantum interference between the transitions. The final term is the multiatom contribution to the absorption. This arises from the collective interaction between the atoms.

It is apparent from (2.36) that an inequality of the populations gives rise to a net, positive or negative, absorption of the probe field. The negative absorption is understood physically as an amplification of the probe field. Thus, a population inversion ($\langle \hat{A}_{ii}^n \rangle > \langle \hat{A}_{jj}^n \rangle$) leads to the probe amplification on the $|i_n\rangle \longleftrightarrow |j_n\rangle$ transition. This is an example of amplification with population inversion. The contribution of the second term in (2.36), proportional to the difference between atomic coherences, may lead to a negative absorption rate even if the first term is positive (no population inversion). Since there is no population inversion between the upper and lower energy

levels, one obtains amplification without population inversion. We should note that the amplification with population inversion can be obtained on a single two-level transition with a negative population difference. However, the amplification without population inversion is unique to multi-level transitions and is observed when more than two energy levels are involved and nonzero coherences exist between the atomic levels. One can see from (2.36) that in order to have a significant amplification without population inversion, it is necessary for the populations to be nearly equal and the difference of the multi-level coherences to be large and negative.

Referring to the role of quantum fluctuations in the process of probe absorption, we stress that in the case of amplification with population inversion, it is spontaneous emission which imposes a serious restriction in creating an inversion between atomic levels. For example, in a single two-level system with ground state $|j_n\rangle$ and excited state $|i_n\rangle$, the stationary absorptive and emissive processes are governed by the balance condition

$$P_j \gamma_{ji} = P_i \gamma_{ij} , \quad (2.37)$$

where $P_j = \langle \hat{A}_{jj}^n \rangle$ and $P_i = \langle \hat{A}_{ii}^n \rangle$ are the steady-state populations of the ground and excited states, respectively. The parameter γ_{ji} is the absorptive and γ_{ij} is the emissive rate between the two energy states. It follows from (2.37) that inversion ($P_i > P_j$) can be produced only if $\gamma_{ji} > \gamma_{ij}$. This condition may not be achieved in two-level systems since the stimulated absorptive and emissive rates are the same and spontaneous emission contributes only to the emissive rate, giving $\gamma_{ij} > \gamma_{ji}$. population inversions involving the ground level can, however, be produced in multilevel systems where population can be transferred into level $|i_n\rangle$ through other channels (levels). If we introduce a third level $|k_n\rangle$ which has its absorptive rate γ_{jk} from the ground level and the spontaneous rate γ_{ki} to the excited level much larger than the emissive rate γ_{ij} , a pumping field applied to the $|j_n\rangle - |k_n\rangle$ transition will create a steady-state inversion on the $|i_n\rangle - |j_n\rangle$ transition. Using rate equations for the atomic populations, the ratio P_i/P_j of the steady-state populations of the excited and ground states can be expressed as

$$\frac{P_i}{P_j} = \frac{\gamma_{ki} \gamma_{jk}}{\gamma_{ij} (\gamma_{ki} + \gamma_{kj})} . \quad (2.38)$$

It is seen that the ratio (2.38) depends crucially on the spontaneous emission rate γ_{ij} which depopulates the upper state $|i_n\rangle$. Maximum inversion, with $P_i = 1$ and $P_j = 0$, is obtained for $\gamma_{ij} = 0$, when the population is said to be ‘shelved’ (trapped) in the state $|i_n\rangle$ from which it cannot decay to the lower state. Thus, in the case of maximum inversion one could expect maximum amplification of a probe field on the $|i_n\rangle - |j_n\rangle$ transition. However, there is another factor which can affect the magnitude of amplification of the probe field. Namely, the dipole moment of the transition, which determines the coupling strength of the probe field to the atom, must be nonzero. According to the balance condition (2.37), an increase of the population inversion can

be achieved by decreasing the emissive rate and the population can be completely inverted only if $\gamma_{ij} = 0$, that is, only if the state $|i_n\rangle$ is a trapping state. Since $\gamma_{ij} \sim |\mu_{ij}|^2$, the trapping results in the cancellation of the dipole moment of the probed transition, and then the inverted transition becomes transparent for the probe field. Therefore, in order to obtain a significant amplification one should produce a large population inversion and simultaneously maintain a strong coupling of the probe field to the inverted transition.

Equation (2.33) applies to the situation of a stationary system in which the commutator depends only on the time difference, and not on t . For a quasistationary system, when commutator depends on t , the treatment may follow the methods we used previously in Sect. 2.1.2 to evaluate the power spectrum. Briefly, if the commutator depends on t , we may first make a harmonic decomposition of the expectation value of the two-time commutator. Next, we substitute the decomposition into the time-dependent spectrum (2.32), and describe the correlation function in terms of the Fourier harmonics of the dipole correlation functions. Calculation of the absorption spectrum then follows exactly the approach that was illustrated in Sect. 2.1.2.

In the foregoing treatment of the absorption spectrum, we have confined our attention to the Mollow's approach in which a probed system is treated as a multi-level quantum system. There is, however, an alternative method of calculating the absorption spectrum in which the atomic system is treated as a homogeneous medium characterized by a (complex) susceptibility χ . The absorption spectrum is then calculated by taking an imaginary part of χ . This method is useful to study spatial propagation effects rather than temporal spectroscopic effects. A complete discussion of the method is out of the scope of this book, and we refer the interested reader to the text by Meystre and Sargent [5] for the complete account of the method.

2.3 Phase-Dependent Spectra and Their Measurements

We have seen that the emission spectrum measures the relative number of photons emitted by a system into vacuum field modes as a function of the spectral frequency of these modes. The absorption spectrum on the other hand measures the relative number of photons absorbed by a system from a probe field as a function of the probe field frequency. It should be noted that the definitions of the emission and absorption spectra involve number of photons which can be measured by direct photocounting techniques.

Additional characterization of the EM field can be found by measuring the amplitude of the field. However, the electric field amplitude is not easy to measure by a direct detection since it is a complex quantity that oscillates at an optical frequency, too fast to be detected by any macroscopic device. It is usually a slowly varying (real) quadrature amplitude of the field and the spectrum of the field-quadrature fluctuations which are measured. The physical quantities that display the field-quadrature fluctuations are Hermitian quadrature components of the electric field operator, which

are specified by a phase θ , an angular frequency ω_c and a wave-vector \mathbf{k} . They are defined in terms of the positive and negative frequency components of the EM field as

$$\begin{aligned}\hat{E}_\theta(\mathbf{r}, t) &= \hat{E}^{(+)}(\mathbf{r}, t) e^{i\xi} + \hat{E}^{(-)}(\mathbf{r}, t) e^{-i\xi}, \\ \hat{E}_{\theta+\pi/2}(\mathbf{r}, t) &= i \left(\hat{E}^{(+)}(\mathbf{r}, t) e^{i\xi} - \hat{E}^{(-)}(\mathbf{r}, t) e^{-i\xi} \right),\end{aligned}\quad (2.39)$$

where we have introduced the oscillatory factor

$$\xi = \omega_c t - \mathbf{k} \cdot \mathbf{r} + \theta \quad (2.40)$$

to make the quadrature components relatively slowly varying functions of time. The phase angle θ may be chosen arbitrary and the quadrature component $\hat{E}_\theta(\mathbf{r}, t)$ is in phase with θ , whereas $\hat{E}_{\theta+\pi/2}(\mathbf{r}, t)$ is $\pi/2$ out of phase. Thus, the component $\hat{E}_{\theta+\pi/2}(\mathbf{r}, t)$ can be obtained from $\hat{E}_\theta(\mathbf{r}, t)$ by incrementing the phase θ by $\pi/2$. Furthermore, according to (2.39) and (1.5), the electric field operator can be written in terms of the quadrature components as

$$\begin{aligned}\hat{E}(\mathbf{r}, t) &= \hat{E}_\theta(\mathbf{r}, t) \cos(\omega t - \mathbf{k} \cdot \mathbf{r} + \theta) \\ &\quad + \hat{E}_{\theta+\pi/2}(\mathbf{r}, t) \sin(\omega t - \mathbf{k} \cdot \mathbf{r} + \theta).\end{aligned}\quad (2.41)$$

This expression shows that the electric field can always be written as a sum of two slowly varying quadrature components oscillating $\pi/2$ out of phase.

The quadrature components do not commute. If, for simplicity, we assume that the detector measures a single polarization component of the field, we find from (2.39) and (1.7) that the quadrature components satisfy the commutation relation

$$\left[\hat{E}_\theta(\mathbf{r}, t), \hat{E}_{\theta+\pi/2}(\mathbf{r}, t) \right] = 2iC, \quad (2.42)$$

which is a particularly simple relation showing that the quadrature components behave as canonically conjugate variables, and

$$C = \sum_{\mathbf{k}} \frac{\hbar \omega_{\mathbf{k}}}{2\varepsilon_0 \mathcal{V}} \quad (2.43)$$

is a real positive number called the quantum (shot-noise) level or the standard quantum limit of the electric field fluctuations. In what follows, we will not consider the spatial effects, so we will suppress the position variable \mathbf{r} .

In terms of the detection theory, the commutation relation (2.42) implies that a simultaneous precise measurement of the two quadrature components is not possible, and from the standard quantum theory, a Heisenberg uncertainty principle applies to the quantum fluctuations in the quadrature components

$$\langle [\Delta \hat{E}_\theta(t)]^2 \rangle \langle [\Delta \hat{E}_{\theta+\pi/2}(t)]^2 \rangle \geq C^2, \quad (2.44)$$

where $\Delta \hat{E}_\theta(t) = \hat{E}_\theta(t) - \langle \hat{E}_\theta(t) \rangle$ is the fluctuation operator and the expectation value is taken over an arbitrary state of the field. The relation (2.44) means that the reduction in the fluctuations of one of the quadrature components occurs at the expense of increased fluctuations in the other component.

The variances $\langle [\Delta \hat{E}_\theta(t)]^2 \rangle$ and $\langle [\Delta \hat{E}_{\theta+\pi/2}(t)]^2 \rangle$ depend on the state of the field, and therefore could serve as distinguishing criteria for different states of the field. To see this, consider possible values of the variances for three different states of the field.

If, for instance, the field is in a chaotic (thermal) state, the fluctuations in both components are large such that

$$\langle [\Delta \hat{E}_\theta(t)]^2 \rangle \geq C \quad \text{and} \quad \langle [\Delta \hat{E}_{\theta+\pi/2}(t)]^2 \rangle \geq C. \quad (2.45)$$

Thus, variances of both quadrature components of a thermal field exceed the standard quantum limit of the electric field fluctuations.

In the case of the vacuum or a coherent state, the fluctuations are independent of θ , as there is no phase reference in the vacuum, and then the variances are equal to the standard quantum limit

$$\langle [\Delta \hat{E}_\theta(t)]^2 \rangle = \langle [\Delta \hat{E}_{\theta+\pi/2}(t)]^2 \rangle = C. \quad (2.46)$$

This relation expresses the fact that the fluctuations are equally distributed in the two quadrature components. Moreover, the product of the two variances equals the minimum possible value indicating that the vacuum (coherent) state is a minimum uncertainty state.

We may proceed further and introduce the idea of special states of the field for which fluctuations in one of the quadrature components may be reduced below the standard quantum limit of the field fluctuations [6–8]. These special states are called *squeezed states* of the field, and are determined by the requirement that either

$$\langle [\Delta \hat{E}_\theta(t)]^2 \rangle < C \quad \text{or} \quad \langle [\Delta \hat{E}_{\theta+\pi/2}(t)]^2 \rangle < C. \quad (2.47)$$

With the help of the commutator (1.7), the variances of the quadrature components can be written as

$$\begin{aligned} \langle [\Delta \hat{E}_\theta(t)]^2 \rangle &= C + \langle : [\Delta \hat{E}_\theta(t)]^2 : \rangle, \\ \langle [\Delta \hat{E}_{\theta+\pi/2}(t)]^2 \rangle &= C + \langle : [\Delta \hat{E}_{\theta+\pi/2}(t)]^2 : \rangle, \end{aligned} \quad (2.48)$$

where the colons denote normal ordering of the operators. In the vacuum or a coherent state of the field $\langle : [\Delta \hat{E}_\theta(t)]^2 : \rangle = \langle : [\Delta \hat{E}_{\theta+\pi/2}(t)]^2 : \rangle = 0$, independent of θ . Hence, we have an equivalent condition for squeezing

$$\langle : [\Delta \hat{E}_\theta(t)]^2 : \rangle < 0 \quad \text{or} \quad \langle : [\Delta \hat{E}_{\theta+\pi/2}(t)]^2 : \rangle < 0. \quad (2.49)$$

It is apparent that a squeezed field has a negative normally ordered variance in one of its quadratures. We shall call this as squeezing in the full sense, because it involves reduction of the fluctuations of the total field.

Note that it is fully consistent with quantum mechanics to shift fluctuations from one of the quadratures to its conjugate. Needless to say, both quadrature variances cannot fall below the quantum limit and beating the standard quantum limit in this way in $\langle [\Delta \hat{E}_\theta(t)]^2 \rangle$ is achieved at the expense of a corresponding increase in the fluctuations of the other quadrature $\langle [\Delta \hat{E}_{\theta+\pi/2}(t)]^2 \rangle$, for which in view of the inequality (2.44) we must have

$$\langle [\Delta \hat{E}_{\theta+\pi/2}(t)]^2 \rangle > C. \quad (2.50)$$

The distribution of the fluctuations among the quadrature components must be such that the product of the variances equals to or is larger than C . Thus, in a squeezed state, the increase of the variance $\langle [\Delta \hat{E}_{\theta+\pi/2}(t)]^2 \rangle$ may exceed the reduction of the variance $\langle [\Delta \hat{E}_\theta(t)]^2 \rangle$. In some cases, however, the field may be in a squeezed state for which the product of the variances is equal to C . Such a state is called the minimum uncertainty squeezed state. Obviously, minimum uncertainty squeezed states are only a subset of a much broader class of squeezed states.

In practice, the usual way to determine fluctuations of the quadrature components of an electric field is to test the criteria (2.45)–(2.49), to which we shall refer as measures of the total field fluctuations. We stress that these measures do not provide the complete information about fluctuations of the field which is measured. For example, for multi-mode fields it is possible for some selected frequencies to exhibit reduced fluctuations or larger fluctuations than that in the total field, which is the integral of the spectral distribution over all frequencies. For this form of fluctuations we shall refer to as spectral component fluctuations and will discuss them in the next section by introducing the concept of phase-dependent noise spectra.

2.4 Homodyne Detection of Quantum Fluctuations

The quadrature components depend on phase. This of course immediately brings up the question how to measure the phase-dependent quadrature components. As we have seen, direct photon counting techniques are not sensitive to the phase of a detected EM field, but only to the field intensity. Therefore, these techniques as a way of detecting phase sensitive quadrature components are impractical. Hence, some alternative techniques are necessary for the measurement of the quadrature components [9, 10]. Typical experimental schemes where phase-dependent fields and their fluctuations are effectively measured are homodyne and heterodyne detection techniques. Homodyning or heterodyning reduces the measurement of a rapidly

oscillating field to the measurement of slowly varying intensities. In the case of optical fields, homodyning or heterodyning is accomplished by adding a strong coherent field to the signal field that is to be measured, and the fluctuations of the superposed field either in photoelectric counting or in photocurrent spectral measurements are detected. The strong coherent field, commonly called the local oscillator, is strictly monochromatic and ideally has a stable amplitude and phase. When the local oscillator frequency is equal to the central frequency of the signal field, we refer to the technique as homodyne detection, and when is different, we refer to as heterodyne detection. The local oscillator provides a controllable fixed phase relative to the signal field that allows to distinguish between the quadrature components of the signal field. The linear superposition of the two fields is achieved using a lossless beam-splitter with the transmissivity very nearly equal unity, so that the signal field is transmitted with a minimal attenuation. As a result, the superposed field emerging from the beam splitter is almost the original signal field together with the much attenuated local oscillator field. What is measured by a detector is the photon count or the photocurrent spectrum of the superposed field as a function of the relative phase difference between the signal field amplitude and the local oscillator amplitude, from which the quadrature components or fluctuations in the quadrature components can be deduced.

A typical homodyne detection scheme is shown in Fig. 2.1. A signal field of an amplitude \hat{E}_s is mixed (beat) at a beam splitter BS with the strong coherent light of a local oscillator field E_{LO} usually derived from an intense, narrow-band laser. In practical realizations of this detection scheme it is necessary to lock the phases of the local oscillator to that of the signal field, and the local oscillator frequency must be equal to the signal field frequency to an accuracy defined by the detection bandwidth. The superposed field E_H (homodyne field) is detected by a detector D placed in one of the outputs of the beam splitter. The resulting photocurrent $i(t)$ is temporarily integrated and analyzed by a photoelectron counter C. In this scheme, the

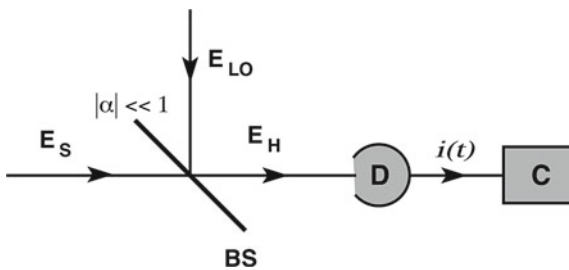


Fig. 2.1 Schematic of a homodyne experiment for detecting phase-dependent quadrature components of the EM field. A signal field E_s is superposed with the local oscillator E_{LO} at a beam splitter BS. The transmissivity of the beam splitter is close to unity, while its reflectivity is very small, $|\alpha| \ll 1$. The superposed field E_H is registered by a detector D and analyzed by a photoelectron counter C. A spectrum analyzer is used instead of the photoelectron counter to detect phase-dependent power spectra of the fluctuations in the selected quadrature of the signal field

beam splitter transmissivity $|\beta|$ is nearly unity while its reflectivity $|\alpha|$ is very small, so that the signal field is transmitted essentially unattenuated. The amplitude of the local oscillator field is kept strong enough that even after a significant attenuation by the beam splitter it still dominates the signal field at the detector.

Let us illustrate the theoretical description of the phase dependence in a homodyne detection scheme, in which a signal field \hat{E}_s is mixed by a beam splitter with a coherent light of a strong local oscillator of the same polarization and angular frequency ω_c , whose phase θ_c can be varied. We will treat the strong local oscillator field classically, even after the much attenuation at the beam splitter, while the signal field amplitude \hat{E}_s will be treated fully quantum-mechanically as an operator. There is only one output field from the beam splitter, the combined (homodyne) field, whose single polarization negative and positive frequency components are given by

$$\begin{aligned}\hat{E}_H^{(-)}(t) &= \alpha E_{LO}^{(-)}(t) + \beta \hat{E}_s^{(-)}(t) , \\ \hat{E}_H^{(+)}(t) &= \alpha^* E_{LO}^{(+)}(t) + \beta^* \hat{E}_s^{(+)}(t) ,\end{aligned}\quad (2.51)$$

where α and β is a complex reflectivity and transmissivity of the beamsplitter satisfying the relations

$$|\alpha|^2 + |\beta|^2 = 1 , \quad \text{and} \quad \beta\alpha^* + \alpha\beta^* = 0 . \quad (2.52)$$

The combined field falls on a photodetector which measures the intensity of the homodyne field, determined by the operator

$$\hat{I}_H(t) = \hat{E}_H^{(-)}(t) \cdot \hat{E}_H^{(+)}(t) . \quad (2.53)$$

If we make use of (2.51) for $\hat{E}_H^{(\pm)}(t)$, which assumes that the photodetector measures a single polarization component, we find that the intensity of the homodyne field becomes

$$\begin{aligned}\hat{I}_H(t) &= \left[|\beta|^2 |\hat{E}_s(t)|^2 + |\alpha|^2 |\mathcal{E}|^2 \right. \\ &\quad \left. + |\alpha\beta| |\mathcal{E}| \left(\hat{E}_\theta(t) \cos \Phi + \hat{E}_{\theta+\pi/2}(t) \sin \Phi \right) \right] ,\end{aligned}\quad (2.54)$$

where $\Phi = \phi + \theta_c - \theta$ is the relative phase difference between the quadrature component and the local oscillator phase θ_c . We have explicitly introduced a constant phase shift ϕ associated with a possible phase change at the beam splitter, $\phi = \arg(\alpha) - \arg(\beta)$, and the phase of the local field has been introduced by writing the positive and negative frequency parts as

$$E_{LO}^{(\pm)}(t) = |\mathcal{E}| e^{\mp i(\omega_c t + \theta_c)} , \quad (2.55)$$

to show that the intensity of the homodyne field can be made to change with the phase of the local oscillator. The quadrature $\hat{E}_\theta(t)$ is in phase with the local oscillator,

while the amplitude $\hat{E}_{\theta+\pi/2}(t)$ is 90° out of phase, and can be obtained from $\hat{E}_\theta(t)$ by incrementing the phase θ_c by $\pi/2$. An examination of (2.54) shows that the dominant signal field contribution to the intensity of the homodyne field arises from the interference term $|\mathcal{E}| \hat{E}_\theta(t)$. This is precisely this interference term that contains quadratures of the signal field and their dependence upon the phase of the local oscillator. Thus, the phase Φ is an important parameter which reflects the phase-dependent signature of the fluctuations of the signal field and determines the measured quadrature component.

Since $|\alpha||\mathcal{E}| \gg |\beta||\langle \hat{E}_s \rangle|$, the first term in (2.54) can be discarded and assuming that the correlation functions involving the local oscillator and the signal field may be factored, we find for the average number of photoelectrons produced by the homodyne field

$$\begin{aligned} \langle m(t, T) \rangle &= 2\varepsilon_0 c \lambda T \langle \hat{I}_H(t) \rangle = \mathcal{U}_T [|\alpha|^2 |\mathcal{E}|^2 \\ &+ |\alpha\beta| |\mathcal{E}| \left(\langle \hat{E}_\theta(t) \rangle \cos \Phi + \langle \hat{E}_{\theta+\pi/2}(t) \rangle \sin \Phi \right)] , \end{aligned} \quad (2.56)$$

where $\mathcal{U}_T = 2\varepsilon_0 c \lambda T$ is the accumulation of the parameters characteristic of the detection process. Equation (2.56) shows that the measured average number of photoelectrons is a weighted combination of the two quadrature components of the signal field. As the phase Φ is varied, the measured quadrature component changes from the in-phase ($\Phi = 0$) to the out-of-phase ($\Phi = \pi/2$) component. We should notice that the detection of a quadrature component is accompanied by a large coherent component to the measured intensity. This is associated with the local oscillator field reflected by the beam splitter and incident on the detector. It cannot be completely eliminated because the reflectivity of the beam splitter can never be zero.

We have just seen how the average intensity of the measured homodyne field provides a measurement of a quadrature component of the signal field. We now turn to the issue of fluctuations of the signal field and will illustrate the concept of measuring the fluctuations of the field amplitude in the homodyne detection scheme. In general, fluctuations of the field amplitude can be found by measuring the variance of the number of photoelectric counts detected in some time interval T . According to (1.40), the variance of the photoelectric counts carries information about the fluctuations of the detected field intensity. If the photoelectric counting experiment is performed on the homodyne field, then the variance of photoelectric counts in a short detection time T is given by

$$\begin{aligned} \langle [\Delta m(t, T)]^2 \rangle &= \mathcal{U}_T \langle \hat{I}_H(t) \rangle + \mathcal{U}_T^2 \left[\langle \hat{E}_H^{(-)}(t) \hat{E}_H^{(-)}(t) \hat{E}_H^{(+)}(t) \hat{E}_H^{(+)}(t) \rangle \right. \\ &\quad \left. - \langle \hat{E}_H^{(-)}(t) \hat{E}_H^{(+)}(t) \rangle^2 \right] . \end{aligned} \quad (2.57)$$

This relation shows explicitly that in general the contribution of the radiation field fluctuations to the photocurrent fluctuations is provided by the fourth-order correlation function of the homodyne field amplitudes.

We may readily relate the variance of photoelectric counts to the normally ordered variance of the quadrature components $\hat{E}_\theta(t)$ and $\hat{E}_{\theta+\pi/2}(t)$ of the signal field. It is done by substituting (2.54) for $\hat{I}_H(t)$ and (2.51) for $\hat{E}_H^{(\pm)}(t)$ and neglecting all terms of order $|\mathcal{E}|$ and less, as small compared with those of the highest order $|\mathcal{E}|^2$, so that we obtain

$$\begin{aligned} \langle [\Delta m(t, T)]^2 \rangle &= \mathcal{U}_T |\alpha|^2 |\mathcal{E}|^2 \\ &+ \frac{1}{2} \mathcal{U}_T^2 |\alpha|^2 |\beta|^2 |\mathcal{E}|^2 \left\{ \langle : [\Delta \hat{E}_\theta(t)]^2 : \rangle (1 + \cos 2\Phi) \right. \\ &+ \langle : [\Delta \hat{E}_{\theta+\pi/2}(t)]^2 : \rangle (1 - \cos 2\Phi) \\ &\left. + 2 \langle : \Delta \hat{E}_\theta(t) \Delta \hat{E}_{\theta+\pi/2}(t) : \rangle \sin 2\Phi \right\}, \end{aligned} \quad (2.58)$$

where $\langle : [\Delta \hat{E}_\theta(t)]^2 : \rangle$ and $\langle : [\Delta \hat{E}_{\theta+\pi/2}(t)]^2 : \rangle$ are the normally ordered variances of the in-phase and out-of-phase quadrature components of the signal field. We see from (2.58) that fluctuations of the quadrature components are manifested as the phase-dependent noise on the homodyne field. For a strong local oscillator, small fluctuations in the signal field are converted into large homodyne field fluctuations.

The two terms appearing on the right-hand side of (2.58) have the following physical interpretation. The first term is the noise term of the local oscillator, equal to the standard quantum limit. This is an unavoidable background noise level in the detection. The second term is the change of the noise due to the interference of the local oscillator with the signal field. This contribution equals the sum of the normally ordered variances for the in-phase and the out-of-phase components together with an interference term involving fluctuation operators of both components. The interference term vanishes for $\Phi = 0$ or $\Phi = \pi/2$, when either the in-phase or out-of-phase component contribution is maximal.

The expression (2.58) applies to the general case of an EM field irrespective of any particular state of the field and for all types of sources of the field. The variance of photoelectric counts can be greater or less than the standard quantum limit and can be optimized with respect to the choice of the phase Φ and the state of the field. When the signal field is in the vacuum state or a coherent state, the normally ordered variance $\langle : [\Delta \hat{E}_\theta(t)]^2 : \rangle = 0$ independent of the phase θ , and then the variance of photoelectric counts is equal to the shot-noise level. When $\langle : [\Delta \hat{E}_\theta(t)]^2 : \rangle < 0$ for some value of the phase θ , the variance is less than the shot-noise level. A negative value of $\langle : [\Delta \hat{E}_\theta(t)]^2 : \rangle$ is nonclassical in the sense that the corresponding (quantum) state of the field cannot be given a diagonal coherent state representation of the density operator ρ of the field. In this case, we say that then the fluctuations of the signal field are squeezed. In other words, turning on the squeezed light lowers the fluctuations of the photoelectric counts. This is of course a reflection of the fact that the field fluctuates less in a squeezed state than in the vacuum state. As we shall see later, information could be carried quite accurately with this field, and this is one of the reasons for our interest in squeezed fields.

Since the variance $\langle [\Delta m(t, T)]^2 \rangle$ depends crucially on the normally ordered variances of the signal field, it follows that the optimum reduction of the variance of photoelectric counts requires us to find the situation where either $\langle : [\Delta \hat{E}_\theta(t)]^2 : \rangle$ or $\langle : [\Delta \hat{E}_{\theta+\pi/2}(t)]^2 : \rangle$ is as negative as possible. In this case the variance $\langle [\Delta m(t, T)]^2 \rangle$ will be reduced below the standard quantum limit by the largest possible amount.

It is interesting to note that the variance in homodyne detection with a strong local oscillator is determined by second-order correlation functions of the signal field operators. In contrast, the variance in direct detection, given by (1.47), is determined by fourth-order correlation functions of the signal operators. In addition, the variance of photoelectric counts depends on the odd correlation functions $\langle \hat{E}^{(-)}(t) \hat{E}^{(-)}(t) \rangle$ and $\langle \hat{E}^{(+)}(t) \hat{E}^{(+)}(t) \rangle$ of the complex field amplitudes of the signal field. These functions, called the *anomalous correlation functions*, are explicit functions of time t , and carry information about two-photon correlations and phase properties of the field [11–15]. Consequently, the anomalous correlation functions are zero for one photon fields such as a coherent or thermal field. We shall have much more to say about these correlation functions, but for the present it is sufficient to note that the anomalous correlation functions are responsible for reduction of the field fluctuations below the quantum limit.

We have already seen that a measurement of the normally ordered variance of the quadrature component involves the homodyning of the total field under study with the local oscillator field. A similar analysis may be carried out for the spectrum of the photon number fluctuations of the homodyne field, which requires the total field to be first frequency filtered and then homodyned with a local oscillator field. As was demonstrated earlier in this chapter, the spectrum can be analyzed by calculating the Fourier transform of the two-time correlation function of photocurrent fluctuations. Let us consider the normally ordered part of the correlation function (1.80) under the time-stationary condition. This is consistent with detection experiments which usually run under stationary conditions, with a steady flow of energy from source to photodetector. The normally ordered two-time correlation function of the homodyne field fluctuations can be written as

$$\begin{aligned} F_{\theta_c}(\tau) &= \lim_{t \rightarrow \infty} \langle \mathcal{T} : \Delta \hat{I}_H(t) \Delta \hat{I}_H(t + \tau) : \rangle \\ &= \lim_{t \rightarrow \infty} \left[\langle \mathcal{T} \hat{E}_H^{(-)}(t) \hat{E}_H^{(-)}(t + \tau) : \hat{E}_H^{(+)}(t) \hat{E}_H^{(+)}(t + \tau) \rangle \right. \\ &\quad \left. - \langle \hat{E}_H^{(-)}(t) \cdot \hat{E}_H^{(+)}(t) \rangle \langle \hat{E}_H^{(-)}(t + \tau) \cdot \hat{E}_H^{(+)}(t + \tau) \rangle \right], \quad (2.59) \end{aligned}$$

where we have put the field operators in the normal order and introduced the subscript θ_c to indicate the dependence of the homodyne field fluctuations on the phase of the local oscillator.

Applying the time ordering, and after substituting for the homodyne field amplitudes from (2.51), the correlation function (2.59) takes the form

$$\begin{aligned}
F_{\theta_c}(\tau) &= |\alpha|^2 |\beta|^2 |\mathcal{E}|^2 F_s(\tau) \\
&= |\alpha|^2 |\beta|^2 |\mathcal{E}|^2 \lim_{t \rightarrow \infty} \left\{ \left[\langle \Delta \hat{E}_s^{(-)}(t) \Delta \hat{E}_s^{(+)}(t + \tau) \rangle e^{i\omega_c \tau} \right. \right. \\
&\quad \left. \left. + \langle \Delta \hat{E}_s^{(-)}(t) \Delta \hat{E}_s^{(-)}(t + \tau) \rangle e^{-i\Phi(t, \tau, \theta_c)} \right] + \text{c.c.} \right\}, \quad (2.60)
\end{aligned}$$

where $\Phi(t, \tau, \theta_c) = 2\omega_c t + \omega_c \tau + 2\phi + 2\theta_c$ and we have designated by $F_s(\tau)$ the correlation functions involving the signal field operators only. As before, an approximation has been made that the amplitude $|\mathcal{E}|$ is large. Consequently, the terms of order $|\mathcal{E}|$ and less have been neglected as small compared with those of order $|\mathcal{E}|^2$.

Evaluation of the correlation function (2.60) is usually carried out in terms of the second-order correlation functions of the quadrature components of the signal field. It is done by substituting from (2.39) for the field amplitudes, and after straightforward calculations we arrive at

$$\begin{aligned}
F_s(\tau) &= \frac{1}{2} \lim_{t \rightarrow \infty} \left\{ \langle \mathcal{T} : \Delta \hat{E}_\theta(t) \Delta \hat{E}_\theta(t + \tau) : \rangle (1 + \cos 2\Phi) \right. \\
&\quad + \langle \mathcal{T} : \Delta \hat{E}_{\theta+\pi/2}(t) \Delta \hat{E}_{\theta+\pi/2}(t + \tau) : \rangle (1 - \cos 2\Phi) \\
&\quad + \left[\langle \mathcal{T} : \Delta \hat{E}_\theta(t) \Delta \hat{E}_{\theta+\pi/2}(t + \tau) : \rangle \right. \\
&\quad \left. \left. + \langle \mathcal{T} : \Delta \hat{E}_{\theta+\pi/2}(t) \Delta \hat{E}_\theta(t + \tau) : \rangle \right] \sin 2\Phi \right\}, \quad (2.61)
\end{aligned}$$

where $\Phi = \phi + \theta_c - \theta$. Note the involvement of the normally ordered and time-ordered correlation functions of the in-phase and the out-of-phase quadrature components together with an interference term.

Once $F_s(\tau)$ has been found, it is only a matter of substitution to derive the stationary spectrum of photocurrent fluctuations. Thus, by substituting (2.61) into (1.81), and replacing $\langle \hat{I}(t) \rangle$ by the dominant term $|\mathcal{E}|^2$, we arrive at the following stationary spectrum of photocurrent fluctuations

$$F(\omega) = |\alpha|^2 |\mathcal{E}|^2 [1 + |\beta|^2 S(\omega, \Phi)], \quad (2.62)$$

where we have introduced the so-called squeezing spectrum associated with the signal field fluctuations

$$S(\omega, \Phi) = 2 \int_0^\infty d\tau F_s(\tau) \cos(\omega\tau). \quad (2.63)$$

Equation (2.62) gives the general expression for the stationary spectrum of photocurrent fluctuations. It relates the measured photocurrent spectral distribution to the squeezing spectrum given in terms of the normally ordered and time-ordered correlation functions for the signal field alone. Thus, the spectrum provides a measure of the contributions of the signal field fluctuations to the photocurrent spectral distribution relative to the vacuum noise level. Just as in the variance of photoelectric counts,

we may identify the first term on the right-hand side of (2.62) with the shot-noise fluctuations of the current. It is white noise, i.e. constant for all frequencies. The second term, proportional to the spectrum of squeezing, depends on frequency and is attributable to the fluctuations of the signal field. Note that the spectrum of squeezing is given by the phase-dependent spectra of quadrature field fluctuations. As the local oscillator phase is varied, the spectrum of squeezing changes from being determined by the quadrature component $\hat{E}_\theta(t)$ to $\hat{E}_{\theta+\pi/2}(t)$ or by any linear combination of the two quadratures. In particular, when the phase is adjusted to satisfy $\Phi = 0$, the squeezing spectrum is determined solely by the in-phase quadrature component

$$S(\omega, 0) = 2 \int_0^\infty d\tau \langle : \Delta \hat{E}_\theta(0) \Delta \hat{E}_\theta(\tau) : \rangle \cos(\omega\tau) , \quad (2.64)$$

and for the choice of $\Phi = \pi/2$, the spectrum simplifies to that determined by the out-of-phase quadrature component

$$S(\omega, \pi/2) = 2 \int_0^\infty d\tau \langle : \Delta \hat{E}_{\theta+\pi/2}(0) \Delta \hat{E}_{\theta+\pi/2}(\tau) : \rangle \cos(\omega\tau) . \quad (2.65)$$

When $S(\omega, \Phi) = 0$, the photocurrent noise is simply the shot noise or vacuum noise seen when the measured signal field is in the vacuum state or in a coherent state. When the signal field is in a squeezed state, the spectrum $S(\omega, \Phi)$ can become negative at some frequencies, resulting in the suppression of the photocurrent noise below the vacuum noise level. The squeezing spectrum has a lower bound of -1 and no upper bound, but the criterion for squeezing as a reduction of the fluctuations below the vacuum level is that the squeezing spectrum must be negative, $S(\omega, \Phi) < 0$. The squeezing will be said to be optimum or perfect at frequency ω when $F(\omega)$ attains its minimum value, $F(\omega) = 0$, with the transmissivity of the beam splitter close to unity, $|\beta|^2 \approx 1$. In this case the squeezing spectrum attains the lower bound $S(\omega, \Phi) = -1$.

A few words should be added at this point about different forms of squeezing. To be specific, for broadband multi-mode fields it is possible that the squeezing spectrum and the normally ordered variance of the total signal field can give different information about squeezing of the field. Namely, if we examine the photocurrent fluctuations $F(\omega)$ for a specific example, we may find that $F(\omega)$ can be smaller than the vacuum noise level for certain ranges of frequencies even though the normally ordered variance of the total signal field is positive. In this case, the field is not squeezed in the full sense. We shall refer to this particular situation as spectral component squeezing. For the case where $F(\omega)$ is smaller than the vacuum noise level for all frequencies, we shall refer to this as homogeneous squeezing. Also, the fluctuations may not be squeezed at some selected frequencies even if $\langle : [\Delta \hat{E}_\theta(t)]^2 : \rangle < 0$, or may exhibit more squeezing than the total field. Nevertheless, in all cases the negative sign of $S(\omega, \Phi)$ indicates the existence of some form of squeezing in the detected field. We shall see that the distinction between squeezing in full sense and squeezing in spectral components is an important one as these two measures give different information about the phase-dependent noise in

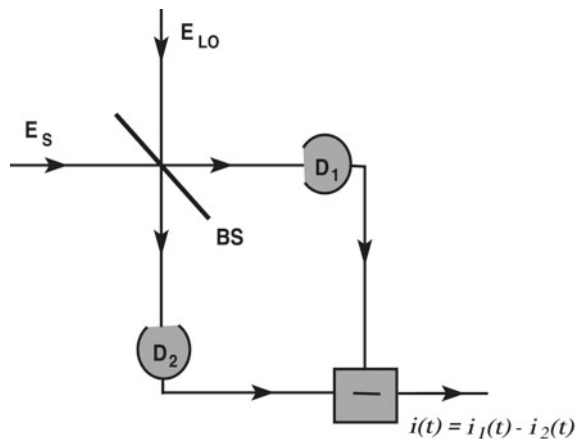
the field. These different forms of squeezing will be further elaborated in the course of specific examples studied in Chaps. 6 and 8.

In summary of this section, we have seen that the photoelectron counting technique in homodyne detection differs in two respects from those of direct photon counting techniques. First, intensity fluctuations in the homodyne detection scheme directly measure the fluctuations in a quadrature component of the signal field. Second, the signal field and its variance depend upon the local oscillator phase, which is an external controllable parameter.

2.5 Balanced Homodyne Detection of Quantum Fluctuations

Experimental measurements of quadrature components and their fluctuations based on the scheme shown in Fig. 2.1 are limited by the contribution of the local oscillator noise. This is because in practice the amplitude of the local oscillator is not truly constant as assumed in an ideal theoretical treatment given in the preceding section. The fluctuations of the local field amplitude lead to the so-called excess noise contribution, which in the ordinary homodyne detection scheme cannot be completely suppressed because the reflectivity of the beam splitter can never be zero. For this reason another scheme, called balanced homodyne detection, has been proposed to measure the phase-dependent quadrature components and their variances [9, 10]. The scheme is illustrated in Fig. 2.2. In contrast to the ordinary homodyne detection scheme, a lossless symmetric 50:50 beam splitter is employed, for which $|\alpha| = |\beta| = 1/\sqrt{2}$, and the superimposed fields are detected with a pair of identical photodetectors D_1 and D_2 located at the two output ports of the beam splitter. Photocurrents from the two detectors are then subtracted electronically and

Fig. 2.2 Schematic diagram of a balanced homodyne experiment for detecting noise limited phase-dependent quadrature components of the EM field and its fluctuations



the difference current $i(t) = i_1(t) - i_2(t)$ and its fluctuations are analyzed. In this way, when the photocurrents are subtracted, the difference current provides measurement of the quadrature components and fluctuations of the signal field alone. The advantage of balanced homodyne detection is that the local oscillator noise can be eliminated completely, leaving only the noise of the signal field. This is explained quantitatively below.

To examine the balanced homodyne detection scheme more closely, let us assume that in addition to a large coherent component \mathcal{E} , the local oscillator amplitude has a certain amount of noise, such that

$$\begin{aligned} E_{LO}^{(+)}(t) &= (\mathcal{E} + \delta\mathcal{E}(t)) e^{-i\omega_c t} , \\ E_{LO}^{(-)}(t) &= (\mathcal{E}^* + \delta\mathcal{E}^*(t)) e^{i\omega_c t} , \end{aligned} \quad (2.66)$$

where $(\mathcal{E} + \delta\mathcal{E}(t)) = |\mathcal{E} + \delta\mathcal{E}(t)| \exp(-i\theta_c)$ is the complex amplitude of the fluctuating local oscillator field with a slowly varying noise $\delta\mathcal{E}(t)$ and a constant phase θ_c not affected by the fluctuations.

The local oscillator noise is specified by a Gaussian statistics with zero mean value $\langle \delta\mathcal{E}(t) \rangle = \langle \delta\mathcal{E}^*(t) \rangle = 0$, and the nonzero second-order correlation

$$\langle \delta\mathcal{E}^*(t) \delta\mathcal{E}(t') \rangle = \mathcal{D} \delta(t - t') , \quad (2.67)$$

where \mathcal{D} represents the noise level of the local oscillator. Without loss in generality, we shall assume that the noise level in the local oscillator is negligible compared with the constant amplitude $|\mathcal{E}|^2$, i.e. $\mathcal{D} \ll |\mathcal{E}|^2$.

In the balanced homodyne detection scheme two photodetectors located at the outputs of the lossless beam splitter measure amplitudes of two homodyne fields

$$\begin{aligned} \hat{E}_1^{(\pm)}(t) &= \frac{1}{\sqrt{2}} \left[\pm i E_{LO}^{(\pm)}(t) + \hat{E}_s^{(\pm)}(t) \right] , \\ \hat{E}_2^{(\pm)}(t) &= \frac{1}{\sqrt{2}} \left[E_{LO}^{(\pm)}(t) \pm i \hat{E}_s^{(\pm)}(t) \right] , \end{aligned} \quad (2.68)$$

where the subscripts 1 and 2 refer to the two outputs of the beam splitter, and the positive and negative parts of the classical local field amplitudes are given by (2.66). Here, the factors “ $\pm i$ ” come from a $\phi = \pi/2$ phase shift between the reflected and transmitted fields at the beam splitter, and we have assumed that the beam splitter has equal amplitude reflectivity and transmissivity coefficients, $|\alpha| = |\beta| = 1/\sqrt{2}$. Because of the lossless mixing of the two fields, the output fields are linear, equally weighted, superpositions of the signal field and the local oscillator amplitudes. The two output fields are separately detected and what is measured is the difference signal between the two outputs from which the quadrature amplitudes and their fluctuations can be deducted.

Let us calculate first the average difference number of photoelectrons produced by the homodyne fields in photodetectors during a short detection time T . We begin with (2.54) and (2.68) to evaluate the intensity of the homodyne fields needed to calculate the number of photoelectrons in each photodetector. As before, in the ordinary homodyne detection we assume that $|\mathcal{E}|$ is much greater than $|\langle \hat{E}_s(t) \rangle|$, so we may ignore the contribution of the term $|\langle \hat{E}_s(t) \rangle|^2$. However, the amplitude $|\mathcal{E}|$ need not be nearly so large as in the ordinary homodyne detection, as $|\alpha|$ is not very small. Thus, if we retain only the leading terms in $|\mathcal{E}|$, we find that the numbers of photoelectrons produced by the two homodyne fields (assuming linearly polarized fields) are

$$\begin{aligned} m_1(t, T) &= \mathcal{U}_T \hat{\mathbf{E}}_1^{(-)}(t) \cdot \hat{\mathbf{E}}_1^{(+)}(t) = \frac{1}{2} \mathcal{U}_T [|\mathcal{E} + \delta\mathcal{E}(t)|^2 \\ &\quad + |\mathcal{E} + \delta\mathcal{E}(t)| (\hat{E}_\theta(t) \cos \Phi + \hat{E}_{\theta+\pi/2}(t) \sin \Phi)] , \\ m_2(t, T) &= \mathcal{U}_T \hat{\mathbf{E}}_2^{(-)}(t) \cdot \hat{\mathbf{E}}_2^{(+)}(t) = \frac{1}{2} \mathcal{U}_T [|\mathcal{E} + \delta\mathcal{E}(t)|^2 \\ &\quad - |\mathcal{E} + \delta\mathcal{E}(t)| (\hat{E}_\theta(t) \cos \Phi + \hat{E}_{\theta+\pi/2}(t) \sin \Phi)] . \end{aligned} \quad (2.69)$$

We see from these expressions that the quadrature components of the signal field are detected against a large background due to the large amplitude and noise of the local oscillator incident on the detectors. This term is certainly of great practical importance since it dominates the interference term, and may be a serious source of limitations in measurement of the quadrature components. Note, however, that this term contributes with the same sign to both numbers of photoelectrons, but the interference term between the local oscillator and the signal field contributes with opposite signs. This suggests that the output photocurrent might be made insensitive to the local oscillator noise if one takes a difference between the two numbers of photoelectrons. Hence, by subtracting the two expressions for the numbers of photoelectrons and taking the expectation value over the initial state of the field, we obtain

$$\begin{aligned} \langle m_-(t, T) \rangle &= \langle m_1(t, T) - m_2(t, T) \rangle \\ &= \mathcal{U}_T |\mathcal{E}| (\langle \hat{E}_\theta(t) \rangle \cos \Phi + \langle \hat{E}_{\theta+\pi/2}(t) \rangle \sin \Phi) . \end{aligned} \quad (2.70)$$

This formula shows that for large enough local oscillator amplitude, the expectation value of the difference number of photoelectrons is free from the contribution of the local oscillator noise, it retains only the interference terms between the amplitude of the local oscillator field and the quadrature components of the signal field. Thus, the cancellation of the local oscillator noise in the difference signal can indeed be achieved and the difficulty with measurement of the quadrature components of the signal field can be avoided. This is a distinct advantage when one wants to measure quadrature components of a weak field.

Consider now the fluctuations in the difference between numbers of photoelectrons in the two output fields. In practice, the fluctuations can be measured by analyzing either the squeezing spectrum or the normally ordered variance of the quadrature

components of the signal field, depending on the experimental scheme adopted. Theoretically, the variance of the difference number of photoelectrons produced by the two homodyne fields may be evaluated using (2.69), from which the fluctuations in the quadrature components of the signal field may be calculated. Assuming the linearly polarized fields, the variance of the difference number of photoelectrons is given by the interference term between the local oscillator intensity $|\mathcal{E}|^2$ and the fluctuations in the quadrature components of the signal field. With the help of the commutation relation (1.13), the variance can be written in terms of normally ordered variances of the signal field quadratures as

$$\langle [\Delta m_-(t, T)]^2 \rangle = \mathcal{U}_T^2 |\mathcal{E}|^2 [1 + F_\theta(t)] , \quad (2.71)$$

where

$$F_\theta(t) = \left\{ \langle : [\Delta \hat{E}_\theta(t)]^2 : \rangle \cos^2 \Phi + \langle : [\Delta \hat{E}_{\theta+\pi/2}(t)]^2 : \rangle \sin^2 \Phi \right. \\ \left. + \langle : \Delta \hat{E}_\theta(t) \Delta \hat{E}_{\theta+\pi/2}(t) : \rangle \sin 2\Phi \right\} . \quad (2.72)$$

Again, we have dropped the term $|\langle \hat{E}_s(t) \rangle|^2$ under the usual assumption that $|\mathcal{E}|$ is very large. We see clearly from (2.71) that even though the local oscillator noise, the variance of the difference number of photoelectrons is free from the local oscillator noise and is a weighted combination of the variance of the fluctuations in the two quadratures of the signal field. Thus, the variance of the difference number of photoelectrons immediately provides information about the noise level in the quadrature components of the signal field.

Of much greater interest is the normally ordered two-time correlation function of the difference current fluctuations. This is because the noise level is often measured as a function of frequency which, according to (1.71), is provided by the spectrum of photocurrent fluctuations. Using (1.81) and by substituting (2.68) for the field amplitudes, we easily find the spectrum of the difference photocurrent fluctuations, which written in terms of the squeezing spectrum of the signal field fluctuations takes the form

$$F_-(\omega) = |\mathcal{E}|^2 [1 + S(\omega, \Phi)] , \quad (2.73)$$

where, once again we have retained only dominant terms in $|\mathcal{E}|$ under the assumption of a strong local oscillator field. Note in particular that the spectrum (2.73) is very similar in form to the expression (2.62) for the ordinary homodyne detection. However, there is an essential difference between these two expressions that the spectrum in the balanced homodyne detection is insensitive to the local oscillator noise, whereas it is altered by the noise in the ordinary homodyne detection.

We may summarize this section as follows. The most notable feature of the balanced homodyne detection scheme is that all contributions due to the large amplitude of the local oscillator and its fluctuations cancel, except for the one arising from the

interference between the local oscillator intensity and variances of the quadrature components of the signal field. For this reason, the balanced homodyne detection scheme provides a better method for measuring quadrature components of the electric field and their fluctuations than the ordinary homodyne detection scheme. It is particularly useful and in fact the most frequently used detection scheme for squeezed light. For details of the experimental observation of squeezed light, we refer the interested reader to some of the excellent review papers [16, 17].

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