Chapter 10
Interaction of Radiation with Atoms

Abstract The preceding chapters have been concerned with the properties of the radiation field alone. In this chapter we turn to the interaction between radiation and matter. This is of course the domain of quantum electrodynamics, however in quantum optics we are usually only concerned with low energy systems of bound electrons which simplifies matters considerably. We will use the occupation number representation for bound many-electron systems to quantize the electronic degrees of freedom, following the approach of Haken [1] and also Cohen-Tannoudji et al. [2].

10.1 Quantization of the Many-Electron System

In the full theory of QED, the interaction between the electromagnetic field and charged matter is described by coupling between the vector potential and the Dirac spinor field. In quantum optics we only need the low energy (non relativistic) limit of this interaction. This is given by the minimal coupling Hamiltonian [3]

$$H = \frac{1}{2m}(\vec{p} - e\vec{A})^2 + eV(\vec{x}) + H_{\text{rad}} \tag{10.1}$$

where $\vec{p}$ is the momentum operator for a particle of charge e moving in a Coulomb potential $V(\vec{x})$. The vector potential is quantised in a box of volume V as

$$\vec{A}(\vec{x},t) = \sum_{n,\nu} \sqrt{\frac{\hbar}{2\varepsilon_o \omega_n V}}\vec{e}_{n,\nu}\left[\mathrm{e}^{\mathrm{i}(\vec{k}_n\cdot\vec{x}-\omega_n t)}a_{n,\nu} + \mathrm{e}^{-\mathrm{i}(\vec{k}_n\cdot\vec{x}-\omega_n t)}a^\dagger_{n,\nu}\right] \tag{10.2}$$

where $\vec{e}_{n,\nu}$ are two orthonormal polarisation vectors $(\nu = 1,2)$ which satisfy $\vec{k}_n \cdot \vec{e}_{n,\nu} = 0$, as required for a transverse field, and the frequency is given by the dispersion relation $\omega_n = c|\vec{k}_n|$. The positive and negative frequency Fourier operators, respectively $a_{n,\nu}$ and $a^\dagger_{m,\nu}$, satisfy

$$[a_{n,\nu}, a_{n',\nu'}] = \delta_{\nu\nu'}\delta_{nn'} \tag{10.3}$$

The last term, $H_{\rm rad}$ is the Hamiltonian of the free radiation field given by

$$H_{\rm rad} = \sum_k \hbar\omega_k a_k^\dagger a_k \tag{10.4}$$

where we have subsumed polarisation and wave vectors labels into the single subscript k.

We now use an occupation number representation in the antisymmetric sector of the many body Hilbert space for the electronic system based on a set of single particle states $|\phi_j\rangle$, with position probability amplitudes, $\phi_j(\vec{x})$, which we take as the bound energy eigenstates of the electronic system without radiation. They could for example be the stationary states of a atom, the quasi bound states of a single Cooper pair on a mesoscopic super-conducting metal island, or the bound exciton states of semiconductor quantum dot. We then define the electronic field operators

$$\hat{\psi}(\vec{x}) = \sum_j c_j \phi_j(\vec{x}) \tag{10.5}$$

where the appropriate commutations relations for the antisymmetric sector are the fermionic forms

$$c_k c_l^\dagger + c_l c_k^\dagger = \delta_{kl} \tag{10.6}$$

$$c_k c_l + c_l c_k = c_k^\dagger c_l^\dagger + c_l^\dagger c_k^\dagger = 0 \tag{10.7}$$

In the occupation number representation the Hamiltonian may be written as the sum of three terms, $H = H_{\rm el} + H_{\rm I} + H_{\rm rad}$ where the electronic part is given by

$$H_{\rm el} = \int {\rm d}^3\vec{x}\hat{\psi}^\dagger(\vec{x}) \left[-\frac{\hbar^2}{2m}\nabla^2 + eV(\vec{x})\right] \hat{\psi}(\vec{x}) = \sum_j E_j c_j^\dagger c_j \tag{10.8}$$

The interaction part may be written as the sum of two terms $H_{\rm I} = H_{\rm I,\,1} + H_{\rm I,\,2}$ where

$$H_{\rm I,\,1} = \int {\rm d}^3\vec{x}\hat{\psi}^\dagger(\vec{x}) \left(-\frac{e}{2m}(\vec{A}(\vec{x}).\vec{p} + \vec{p}.\vec{A}(\vec{x}))\right) \hat{\psi}(\vec{x}) \tag{10.9}$$

$$H_{\rm I,\,2} = \int {\rm d}^3\vec{x}\hat{\psi}^\dagger(\vec{x}) \left(\frac{e^2}{2m}(\vec{A}(\vec{x})^2\right) \hat{\psi}(\vec{x}) \tag{10.10}$$

Unless we are dealing with very intense fields for which multi-photon processes are important, the second term $H_{\rm I,\,2}$ may be neglected.

The dominant interaction energy may then be written as

$$H_{\rm I} = \hbar \sum_{\vec{k},n,m} g_{\vec{k},n,m}(b_{\vec{k}} + b_{\vec{k}}^\dagger) c_n^\dagger c_m \tag{10.11}$$

where the interaction coupling constant is

$$g_{\vec{k},n,m} = -\frac{e}{m}\left(\frac{1}{2\varepsilon_0\hbar\omega_k V}\right)^{1/2}\int \mathrm{d}^3\vec{x}\phi_n^*(\vec{x})\left(\mathrm{e}^{\mathrm{i}\vec{k}.\vec{x}}\vec{p}\right)\phi_m(\vec{x}) \tag{10.12}$$

We now proceed by making the *dipole approximation*. The factor $e^{i\vec{k}\vec{x}}$ varies on a spatial scale determined by the dominant wavelength scale, λ_0, of the field state. At optical frequencies, $\lambda_0 \approx 10^{-6}$ m. However the atomic wave functions, $\phi_n(\vec{x})$ vary on a scale determined by the Bohr radius, $a_0 \approx 10^{-11}$ m. Thus we may remove the oscillatory exponential from the integral and evaluate it at the position of the atom $\vec{x} = \vec{x}_0$. Using the result

$$[\vec{p}^2,\ \vec{x}] = -\mathrm{i}2\hbar\vec{p} \tag{10.13}$$

we can write the interaction in terms of the atomic dipole moments

$$\int \mathrm{d}^3\vec{x}\phi_n^*(\vec{x})\left(\mathrm{e}^{\mathrm{i}\vec{k}.\vec{x}}\vec{p}\right)\phi_m(\vec{x}) = \mathrm{i}\frac{m}{e}\omega_{nm}\mathrm{e}^{\mathrm{i}\vec{k}.\vec{x}_0}\int \mathrm{d}^3\vec{x}\phi_n^*(\vec{x})(\mathrm{e}\vec{k})\phi_m(\vec{x}) \tag{10.14}$$

where $\omega_{nm} = (E_n - E_m)/\hbar$.

In the interaction picture the interaction Hamiltonian becomes explicitly time dependent,

$$\tilde{H}_{\mathrm{I}}(t) = \hbar\sum_{\vec{k},n,m} g_{\vec{k},n,m}(b_{\vec{k}}\mathrm{e}^{-\mathrm{i}\omega(\vec{k})t} + b_{\vec{k}}^\dagger\mathrm{e}^{\mathrm{i}\omega(\vec{k})t})c_n^\dagger c_m\mathrm{e}^{\mathrm{i}\omega_{nm}t} \tag{10.15}$$

where the tilde indicates that we are in the interaction picture. If the field is in state for which the dominant frequency is such that $\omega(\vec{k}_0) \approx \omega_{nm}$, the field is resonant with a particular atomic transition and we may neglect terms rotating at the very high frequency $\omega(\vec{k}) + \omega_{nm}$. This is known as the *rotating wave approximation*. This assumes that the field strength is not too large and further that the state of the field does not vary rapidly on a time scale of ω_{nm}^{-1} i.e. we ignore fields of very fast strong pulses. As a special case we assume the field is resonant (or near-resonant) with a single pair of levels with $E_2 > E_1$. The interaction picture Hamiltonian in the dipole and rotating wave approximation is then given by

$$\tilde{H}_I = \hbar\sum_{\vec{k}} c_1^\dagger c_2 b_{\vec{k}}^\dagger g_{\vec{k}}\mathrm{e}^{-\mathrm{i}(\omega(\vec{k})-\omega_{21})t} + \mathrm{h.c} \tag{10.16}$$

where

$$g_{\vec{k}} = -\mathrm{i}\left(2\hbar\varepsilon_0\omega(\vec{k})V\right)^{-1/2}\omega_{\mathrm{a}}\mu_{21}\mathrm{e}^{\mathrm{i}\vec{k}.\vec{x}_0} \tag{10.17}$$

and

$$\mu_{21} = \langle\phi_n|\mathrm{e}\vec{x}|\phi_m\rangle \tag{10.18}$$

with $\omega_{\mathrm{a}} = \omega_2 - \omega_1$.

It is conventional to describe the operator algebra of a two level system in terms of pseudo-spin representation by noting that the Pauli operators may be defined as

$$\sigma_z = c_2^\dagger c_2 - c_1^\dagger c_1 \tag{10.19}$$

$$\sigma_x = c_2^\dagger c_1 + c_1^\dagger c_2 \tag{10.20}$$

$$\sigma_y = -\mathrm{i}(c_2^\dagger c_1 - c_1^\dagger c_2) \tag{10.21}$$

$$\sigma_+ = \sigma_-^\dagger = c_2^\dagger c_1 \tag{10.22}$$

The operators $s_\alpha = \sigma_\alpha/2$ (with $\alpha = x,y,z$) then obey the $su(2)$ algebra for a spin half system. In terms of these operators we may write the total Hamiltonian for the system of field plus atom in the dipole and rotating wave approximation as

$$H = \sum_{\vec{k}} \hbar\omega(\vec{k}) b_{\vec{k}}^\dagger b_{\vec{k}} + \frac{\hbar\omega_\mathrm{a}}{2}\sigma_z + \hbar\sum_{\vec{k}} g_{\vec{k}} b_{\vec{k}}\sigma_+ + \mathrm{h.c}\,. \tag{10.23}$$

The free Hamiltonian for the two-level electronic system is

$$\mathscr{H}_\mathrm{el} = \frac{\hbar\omega_a}{2}\sigma_z \tag{10.24}$$

Denoting the ground and excited states as $|1\rangle$ and $|2\rangle$ respectively, we see that

$$\mathscr{H}_\mathrm{el}|s\rangle = (-1)^s \frac{\hbar\omega_\mathrm{a}}{2}|s\rangle \quad s = 1,2 \tag{10.25}$$

The action of the raising and lowering operators on the energy eigenstates is: $\sigma_+|1\rangle = |2\rangle$ and $\sigma_-|2\rangle = |1\rangle$, while $\sigma_\pm^2 = 0$. We now relabel the ground state and excited state respectively as $|1\rangle \equiv |g\rangle$, $|2\rangle \equiv |e\rangle$. If the state of the system at time t is ρ, the probability to find the electronic system in the excited state and ground state are, respectively,

$$p_e(t) = \langle 2|\rho|2\rangle = \langle \sigma_+\sigma_-\rangle \tag{10.26}$$

$$p_g(t) = \langle 1|\rho|1\rangle = \langle \sigma_-\sigma_+\rangle \tag{10.27}$$

The *atomic inversion* is defined as the difference between these two probabilities and is given by

$$p_e(t) - p_g(t) = \langle \sigma_z\rangle \tag{10.28}$$

While the *atomic coherences* are defined by

$$\rho_{12} \equiv \langle 1|\rho|2\rangle = \langle \sigma_+\rangle \tag{10.29}$$

with $\rho_{21} = \rho_{12}^*$.

10.2 Interaction of a Single Two-Level Atom with a Single Mode Field

If we further restrict the state of the field to include only a single mode, with frequency ω_0; perhaps using a high Q optical resonator, we arrive at the *Jaynes–Cummings* hamiltonian,

$$H = \hbar\omega_0 b^\dagger b + \frac{\hbar\omega_a}{2}\sigma_z + \hbar(gb\sigma_+ + g^* b^\dagger \sigma_-) \tag{10.30}$$

coupling a single harmonic oscillator degree of freedom to a two-level system, which might well be called the standard model of quantum optics [4]. The coupling constant g can vary from a few kHz to many MHz. An example is provided by the experiment of Aoki et al. [5] in which a cesium atom interacts with the toroidal whispering gallery mode of a micro-resonator as it falls under the action of gravity from a magneto-optical trap. The atomic resonance at is the $6S_{1/2}$; $F = 4 \to 6P_{3/2}$; $F' = 5$ transition in cesium. A coupling constant as large as $g/2\pi = 50\,\text{MHz}$ was achieved.

On resonance, $\omega_a = \omega_c = \omega$, we see that the interaction Hamiltonian $\mathscr{H}_I = \hbar g(b\sigma_+ + b^\dagger\sigma_-)$ (with g chosen as real), commutes with the free Hamiltonian, $\mathscr{H}_0 = \hbar\omega(b^\dagger b + \frac{1}{2}\sigma_z)$, so that the eigenstates of the full Hamiltonian can be written as a linear combination of the degenerate eigenstates of $\mathscr{H}_0$. Defining $|n,s\rangle = |n\rangle_b \otimes |s\rangle$, where $b^\dagger b|n\rangle = n|n\rangle$, the degenerate eigenstates of the free Hamiltonian are $|n,2\rangle,\ |n+1,1\rangle$. Within this degenerate subspace, the state at time t may be written $|\psi_n(t)\rangle = c_{n,2}(t)|n,2\rangle + c_{n+1,2}(t)|n+1,1\rangle$, and the Schröedinger equation in the interaction picture is

$$\begin{pmatrix} \dot{c}_{n,2} \\ \dot{c}_{n+1,1} \end{pmatrix} = -\mathrm{i}\Omega_n \sigma_x \begin{pmatrix} c_{n,2} \\ c_{n+1,1} \end{pmatrix} \tag{10.31}$$

where $\Omega_n = g\sqrt{n+1}$. The eigenvalues of this system of linear equations are $\pm\mathrm{i}\Omega_n$, corresponding to the eigenstates of $\mathscr{H}_\mathrm{I}$

$$|n,\pm\rangle = \frac{1}{\sqrt{2}}(|n,2\rangle \pm |n+1,1\rangle) \tag{10.32}$$

which are often referred to as the *dressed states*. The splitting of the degeneracy is depicted in Fig. 10.1.

Thus the general solution is

$$c_{n,2}(t) = c_{n,2}(0)\cos\Omega_n t - \mathrm{i}c_{n+1,1}(0)\sin\Omega_n t \tag{10.33}$$

$$c_{n+1,1}(t) = c_{n+1,1}(0)\cos\Omega_n t - \mathrm{i}c_{n,2}(0)\sin\Omega_n t \tag{10.34}$$

If the atom is initially in the excited state and the cavity field has exactly n photons, the probability for finding the atom in the *same* state at time $t > 0$ is

$$p_e(t) = |\langle n,2|\psi_n(t)\rangle|^2 = \frac{1}{2}(1+\cos 2\Omega_n t) \tag{10.35}$$

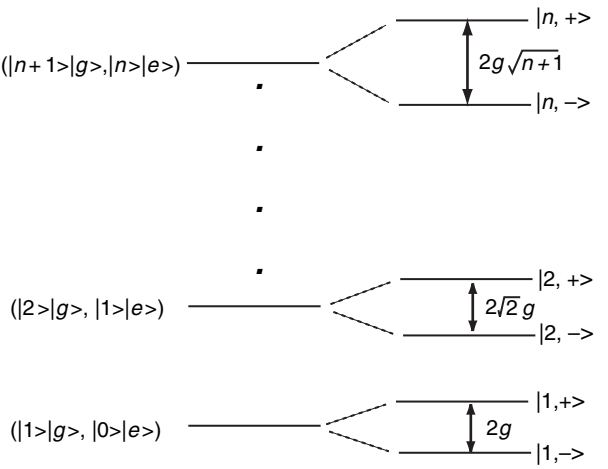

Fig. 10.1 The *dressed states* for the energy eigenstates of the Jaynes–Cummings interaction. On the left are show the degenerate states for zero interaction. When the interaction is turned on the degeneracies are lifted

The excitation oscillates backward and forth between the cavity and the electronic system with frequency Ω_n, the Rabi frequency. Note that for $n = 0$ the separation of these eigenvalues is $2g$, which is known as the *vacuum Rabi splitting*.

If the field is in an arbitrary pure state, $|\phi\rangle = \Sigma_n f_n |n\rangle$ and the atom is initially excited, the probability to find the atom in the excited state at time $t > 0$ may be written

$$p_e(t) = \frac{1}{2}\left[1 + \sum_{n=0}^{\infty} |f_n|^2 \cos(2g\sqrt{n+1}t)\right] \tag{10.36}$$

This is a discrete superposition of harmonic oscillations with incommensurate frequencies. Thus it must exhibit quasiperiodic behaviour. If the initial photon number distribution $|f_n|^2$ has narrow support on n, only a few frequencies are involved and there is a beating between these different frequencies leading to what are known as collapses and revivals. The collapse refers to the decay of oscillations at short times due to beating between the incommensurate frequencies. The revival refers to partial re-phasing of the oscillations at later times. In the case of the field initially in a coherent state, $|\alpha\rangle$, the initial number distribution is Poissonian with standard deviation in number given by the root mean, $\bar{n}^{1/2}|\alpha|$. An approximate evaluation of the sum valid for times such that $gt < \bar{n}^{1/2}$ gives [6]

$$p_e(t) = \frac{1}{2}\left[1 + \mathrm{e}^{-\frac{g^2t^2\bar{n}}{2(n+1)}} \cos(2g\sqrt{\bar{n}+1}t)\right] \tag{10.37}$$

There is an average Rabi oscillation frequency under a Gaussian envelope. The characteristic time for the collapse of the oscillation is thus

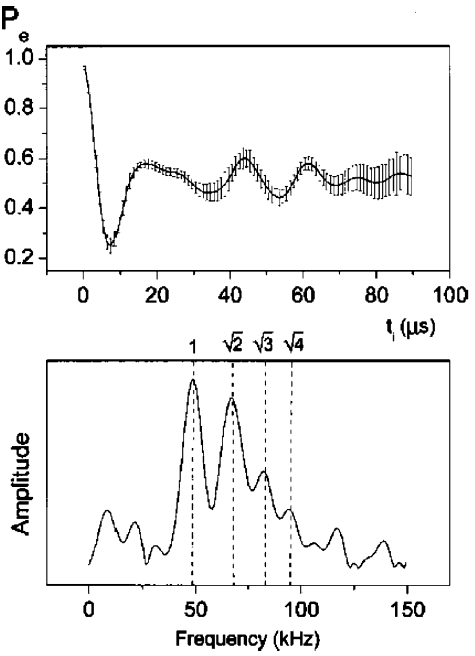

Fig. 10.2 (**a**) The experimental observation of collapse and revival of the oscillations in the occupation of the excited state of a two-level atom interacting with a microwave field initially in a coherent state with mean photon number $\bar{n} = 0.85$. In (**b**) is the Fourier transform of the oscillations with the Rabi frequencies Ω_n, $n = 0,\ 1,\ 2,\ 3$ marked (from [7])

$$t_{\text{col}} \sim \frac{1}{g} \tag{10.38}$$

A more accurate evaluation using the Laplace summation formulae shows that the oscillations first revive at a t time

$$t_{\text{rev}} \sim \frac{2\pi}{g}\bar{n}^{1/2} \tag{10.39}$$

Thus a quasi periodic burst of Rabi oscillations occurs every $\bar{n}$ Rabi periods. The collapse and revival has been seen experimentally using an atom excited to a Ryberg ground state interacting with the microwave field in a superconducting cavity [7]. The results of the experiment are shown in Fig. 10.2.

10.3 Spontaneous Emission from a Two-Level Atom

Spontaneous emission can also be treated using a master equation. In this case the system is a two-level electronic system, with ground state $|g\rangle$ of energy $\hbar\omega_1$ and excited state $|e\rangle$ with energy $\hbar\omega_2$, representing an electric dipole transition, coupled to the many modes of the radiation field in the dipole and rotating wave approximation. The master equation is

$$\frac{\mathrm{d}\rho}{\mathrm{d}t} = -\frac{\mathrm{i}}{\hbar}[H,\rho] + \gamma(\bar{n}+1)\mathscr{D}[\sigma_-]\rho + \gamma\bar{n}\mathscr{D}[\sigma_+]\rho \tag{10.40}$$

where $\bar{n}$ is the thermal occupation of the radiation field mode at the atomic resonance frequency $\omega_\mathrm{a} = \omega_2 - \omega_1$. We have neglected a small term which gives rise to a shift in the atomic transition frequency and which contributes to the Lamb shift. At optical frequencies, $\bar{n} \approx 0$. In the case of a free two-level atom, $H = \frac{\hbar\omega_\mathrm{a}}{2}\sigma_z$ the probability to find the atom in the excited state, $p_e(t) = \langle e|\rho|e\rangle$ satisfies the equation

$$\frac{\mathrm{d}p_e}{\mathrm{d}t} = -\gamma p_e(t) \tag{10.41}$$

with the solution $p_e(t) = p_e(0)\mathrm{e}^{-\gamma t}$, which describes spontaneous emission. The dipole polarisation is proportional to the atomic coherence, $\langle e|\rho|g\rangle = \langle\sigma_-\rangle$ which obeys

$$\frac{\mathrm{d}\langle\sigma_-\rangle}{\mathrm{d}t} = -\left(\mathrm{i}\omega_\mathrm{a} + \frac{\gamma}{2}\right)\langle\sigma_-\rangle \tag{10.42}$$

with the solution

$$\langle\sigma_-(t)\rangle = \langle\sigma_-(0)\rangle\mathrm{e}^{-(\gamma/2+\mathrm{i}\omega_\mathrm{a})t} \tag{10.43}$$

The dipole oscillates at the transition frequency and decays, as it radiates.

The radiated field is related to the input field and the local source through an input/output relation in analogy with the case of a cavity discussed above. The positive frequency components of the field operator takes the form

$$E_\mathrm{o}^{(+}(\vec{x},t) = E_\mathrm{i}^{(+)}(\vec{x},t) - \frac{\omega_a^2}{4\pi\varepsilon_0 c^2 r}\left(\vec{\mu}\times\frac{\vec{x}}{r}\right)\times\frac{\vec{x}}{r}\sigma_-(t-x/c) \tag{10.44}$$

where $r = |\vec{x}|$ is the distance from the source to the point $\vec{x}$ and $\vec{\mu}$ is the atomic dipole moment.

10.4 Phase Decay in a Two-Level System

Spontaneous emission is not the only irreversible process involved in the absorption and emission of light. In an atomic vapour, atomic collisions are also a source of decoherence and cause a decay of the atomic polarisation, $\sigma_x + \mathrm{i}\sigma_y$, without changing the decay of the inversion, σ_z. We can model this process by a coupling between the inversion and a high temperature heat bath,

$$H_\mathrm{col} = \sigma_z\Gamma_\mathrm{c}(t) \tag{10.45}$$

where $\Gamma_\mathrm{c}(t)$ is a bath operator describing the collisions. This Hamiltonian commutes with σ_z and thus does not contribute to the decay of the inversion. It appears like a fluctuating detuning in the Bloch equations and thus will effect the atomic

polarisation. The corresponding master equation, in the interaction picture and including spontaneous emission, is

$$\frac{\mathrm{d}\rho}{\mathrm{d}t} = \frac{\gamma}{2}(2\sigma_-\rho\sigma_+ - \sigma_+\sigma_+\rho - \rho\sigma_+\sigma_+) - \gamma_p[\sigma_z,[\sigma_z[,\rho]] \tag{10.46}$$

The Bloch equations now become

$$\frac{\mathrm{d}\langle\sigma_z\rangle}{\mathrm{d}t} = -\gamma(\langle\sigma_z\rangle + 1) \tag{10.47}$$

$$\frac{\mathrm{d}\langle\sigma_x\rangle}{\mathrm{d}t} = -\left(\frac{\gamma}{2} + \gamma_p\right)\langle\sigma_x\rangle \tag{10.48}$$

$$\frac{\mathrm{d}\langle\sigma_y\rangle}{\mathrm{d}t} = -\left(\frac{\gamma}{2} + \gamma_p\right)\langle\sigma_y\rangle \tag{10.49}$$

In the presence of collisions the decay time for the polarisation, T_2, is no longer given by twice the decay time for the inversion, $T_1 = \gamma^{-1}$, but rather $T_2 < 2T_1$.

10.5 Resonance Fluorescence

If the atom is driven by a classical radiation field, the Hamiltonian becomes (see (10.30) and replace $b \mapsto \beta$)

$$H = \frac{\hbar\omega_\mathrm{a}}{2}\sigma_z + \Omega(\sigma_+ \mathrm{e}^{-\mathrm{i}\omega_\mathrm{L}t} + \sigma_- \mathrm{e}^{\mathrm{i}\omega_\mathrm{L}t}) \tag{10.50}$$

where $\Omega = g\beta$ is the Rabi frequency and ω_L is the carrier frequency of the driving field. The master equation in an interaction picture at the frequency ω_L is

$$\frac{\mathrm{d}\rho}{\mathrm{d}t} = -\mathrm{i}\frac{\Delta\omega}{2}[\sigma_z,\rho] - \mathrm{i}\Omega[\sigma_+ + \sigma_-,\rho] + \gamma\mathscr{D}[\sigma_-]\rho \tag{10.51}$$

where the detuning is $\Delta\omega = \omega_\mathrm{a} - \omega_\mathrm{L}$. The resulting *Bloch equations* for the atomic moments are linear

$$\frac{\mathrm{d}\langle\sigma_-\rangle}{\mathrm{d}t} = -\left(\frac{\gamma}{2} + \mathrm{i}\Delta\omega\right)\langle\sigma_-\rangle + \mathrm{i}\Omega\langle\sigma_z\rangle \tag{10.52}$$

$$\frac{\mathrm{d}\langle\sigma_z\rangle}{\mathrm{d}t} = -\gamma(\langle\sigma_z\rangle + 1) - 2\mathrm{i}\Omega(\langle\sigma_+\rangle - \langle\sigma_-\rangle) \tag{10.53}$$

These inhomogeneous equations can be written as homogeneous equations as

$$\frac{\mathrm{d}}{\mathrm{d}t}(\langle\vec{\sigma}(t)\rangle - \langle\vec{\sigma}\rangle_\mathrm{ss}) = A(\langle\vec{\sigma}(t)\rangle - \langle\vec{\sigma}\rangle_\mathrm{ss}) \tag{10.54}$$

where

$$A = \begin{pmatrix} -\left(\frac{\gamma}{2} - \mathrm{i}\Delta\omega\right) & 0 & -\mathrm{i}\Omega \\ 0 & -\left(\frac{\gamma}{2} + \mathrm{i}\Delta\omega\right) & \mathrm{i}\Omega \\ -2\mathrm{i}\Omega & 2\mathrm{i}\Omega & -\gamma \end{pmatrix} \tag{10.55}$$

with $\langle\vec{\sigma}\rangle = (\langle\sigma_+\rangle, \langle\sigma_-\rangle, \langle\sigma_z\rangle)^T$ and the steady state solutions are

$$\langle\sigma_z\rangle_{\mathrm{ss}} = -\frac{1+\delta^2}{1+\delta^2+Z^2} \tag{10.56}$$

$$\langle\sigma_+\rangle_{\mathrm{ss}} = \frac{\mathrm{i}}{\sqrt{2}} \frac{Z(1+\mathrm{i}\delta)}{1+\delta^2+Z^2} \tag{10.57}$$

with

$$Z = \frac{2\sqrt{2}\Omega}{\gamma}, \quad \delta = \frac{2\Delta\omega}{\gamma} \tag{10.58}$$

The solutions for resonance ($\Delta\omega = 0$), with the atom initially in the ground state, are

$$\langle\sigma_z(t)\rangle = \frac{8\Omega^2}{\gamma^2+8\Omega^2}\left[1 - \mathrm{e}^{-3\gamma t/4}\left(\cosh\kappa t + \frac{3\gamma}{4\kappa}\sinh\kappa t\right)\right] - 1 \tag{10.59}$$

$$\langle\sigma_+(t)\rangle = 2i\Omega\frac{\gamma}{\gamma^2+8\Omega^2}\left[1 - \mathrm{e}^{-3\gamma t/4}\left(\cosh\kappa t + \left(\frac{\kappa}{\gamma} + \frac{3\gamma}{16\kappa}\right)\sinh\kappa t\right)\right] \tag{10.60}$$

where

$$\kappa = \frac{1}{2}\sqrt{\frac{\gamma^2}{4} - 16\Omega^2} \tag{10.61}$$

Clearly there is a threshold at $\Omega = \gamma/8$ below which the solutions monotonically approach the steady state and above which they are oscillating. A similar threshold occurs in the solutions for the two-time correlation function $\langle\sigma_+(t)\sigma_-(t+\tau)\rangle_{t\to\infty}$ which determines the spectrum of the scattered light.

The stationary spectrum, as measured by a monochromatic detector at the point $\vec{x}$ is defined by [9]

$$S(\vec{x},\omega) = \lim_{t\to\infty}\frac{1}{2\pi}\int_{-\infty}^{\infty}\langle E^{(-)}(\vec{x},t)E^{(+)}(\vec{x},t+\tau)\rangle \mathrm{d}\tau \tag{10.62}$$

the Fourier transform of the stationary two-time correlation function $\langle E^{(-)}(t)E^{(+)}(t+\tau)\rangle$ which using (10.44) is given by

$$S(\vec{x},\omega) = \frac{I_0(\vec{x})}{2\pi}\int_{-\infty}^{\infty}\mathrm{d}\tau \mathrm{e}^{-\mathrm{i}\omega\tau}G(\tau) \tag{10.63}$$

where

$$I_0(\vec{x}) = \left| \frac{\omega_0^2}{4\pi\varepsilon_0 c^2 r} \left(\vec{\mu} \times \frac{\vec{x}}{r} \right) \times \frac{\vec{x}}{r} \right|^2 \tag{10.64}$$

and

$$G(\tau) = \lim_{t\to\infty} \langle \sigma_+(t)\sigma_-(t+\tau)\rangle \equiv \langle \sigma_+\sigma_-(\tau)\rangle_{\rm ss} \tag{10.65}$$

with

$$\langle \sigma_+(t)\sigma_-(t+\tau)\rangle = {\rm tr}\left[\sigma_- e^{\mathscr{L}\tau}\rho(t)\sigma_+\right] \tag{10.66}$$

The equation of motion for $G(\tau)$ couples in many other moments. If we define the correlation matrix

$$\mathscr{G}(\tau) = \begin{pmatrix} \langle \sigma_+\sigma_+(\tau)\rangle_{\rm ss} & \langle \sigma_-\sigma_+(\tau)\rangle_{\rm ss} & \langle \sigma_z\sigma_+(\tau)\rangle_{\rm ss} \\ \langle \sigma_+\sigma_-(\tau)\rangle_{\rm ss} & \langle \sigma_-\sigma_-(\tau)\rangle_{\rm ss} & \langle \sigma_z\sigma_-(\tau)\rangle_{\rm ss} \\ \langle \sigma_+\sigma_z(\tau)\rangle_{\rm ss} & \langle \sigma_-\sigma_z(\tau)\rangle_{\rm ss} & \langle \sigma_z\sigma_z(\tau)\rangle_{\rm ss} \end{pmatrix} \tag{10.67}$$

The quantum regression theorem indicates that $\mathscr{G}(\tau)$ as a function of τ obeys the same equations of motion as $\langle \vec{\sigma}(\tau)\rangle - \langle \vec{\sigma}\rangle_{\rm ss}$,

$$\frac{\mathscr{G}(\tau)}{{\rm d}\tau} = A\mathscr{G}(\tau) \tag{10.68}$$

The initial conditions are simplified due to the algebra of the Pauli matrices, for example $\sigma_+\sigma_- = (\sigma_z + 1)/2$ and $\sigma_\pm^2 = 0$, and may thus be written in terms of the stationary solutions in (10.57). On resonance we find that in the Schrödinger picture,

$$\begin{aligned} G(\tau) = & \frac{4\Omega^2}{\gamma^2 + 8\Omega^2} \left[\frac{\gamma^2}{\gamma^2 + 8\Omega^2} e^{-i\omega_a t} + \frac{1}{2} e^{-(\gamma/2 + i\omega_a)\tau} \right. \\ & - \frac{1}{2} \left(\frac{\gamma^2}{\gamma^2 + 8\Omega^2} \frac{3\gamma/4 + \kappa}{\kappa} - \frac{\gamma/2}{\kappa} - \frac{\gamma/4 + \kappa}{2\kappa} \right) \exp\{-(3\gamma/4 - \kappa + i\omega_a)\tau\} \\ & \left. + \frac{1}{2} \left(\frac{\gamma^2}{\gamma^2 + 8\Omega^2} \frac{3\gamma/4 - \kappa}{\kappa} - \frac{\gamma/2}{\kappa} - \frac{\gamma/4 - \kappa}{2\kappa} \right) \exp\{-(3\gamma/4 + \kappa + i\omega_a)\tau\} \right] \end{aligned} \tag{10.69}$$

with $\tau \geq 0$. The corresponding spectrum has a single Lorentzian peak for weak driving fields, $4\Omega << \gamma^2/16$,

$$S(\vec{x}, \omega) = I_0(r) \frac{4\Omega^2}{\gamma^2 + 8\Omega^2} \delta(\omega - \omega_a) \tag{10.70}$$

which corresponds to elastic scattering. For very strong driving fields, $\Omega >> \gamma$ we find that the spectrum acquires three Lorentzian peaks at $\omega = \omega_a$ and $\omega = \omega_a \pm 2\Omega$. The spectrum, including the elastic term, is

$$S(\vec{x},\omega) = \frac{I_0(r)}{2\pi}\left(2\pi\frac{4\Omega^2}{\gamma^2+8\Omega^2}\delta(\omega-\omega_a)+\frac{1}{2}\frac{\gamma/2}{\gamma^2/4+(\omega-\omega_a)^2}\right.$$
$$\left.+\frac{1}{4}\frac{3\gamma/4}{(3\gamma/4)^2+[\omega-(\omega+2\Omega)]^2}+\frac{1}{4}\frac{3\gamma/4}{(3\gamma/4)^2+[\omega-(\omega-2\Omega)]^2}\right) \tag{10.71}$$

This is the *Mollow spectrum* [10].

The light scattered by a two-level atom also exhibits *photon anti-bunching*. Consider the conditional probability that given a photon is counted at time t another photon will be counted a time τ later. This is proportional to the second order correlation function

$$G^{(2)}(t,\tau) = \langle a^\dagger(t)a^\dagger(t+\tau)a(t+\tau)a(t)\rangle \tag{10.72}$$

Usually we are interested in a stationary source so we let $t\to\infty$ and we normalise this by the intensity squared to define

$$g^{(2)}(\tau) = \lim_{t\to\infty}\frac{G^{(2)}(t,\tau)}{\langle a^\dagger(t)a(t)\rangle^2} \tag{10.73}$$

Using the result in (10.44) we can express this directly in terms of correlation functions for the atomic polarisation. As the equations of motion for the atomic variables are linear, the stationary correlation function $\langle\sigma_+(t)\sigma_+(t+\tau)\sigma_-(t+\tau)\sigma_-(t)\rangle_{t\leftarrow\infty}$ is given by the quantum regression theorem. We then find that

$$g^{(2)}(\tau) = 1-e^{-3\gamma t/4}\left(\cosh\kappa\tau+\frac{3\gamma}{4\kappa}\sinh\kappa\tau\right) \tag{10.74}$$

The result $g^{(2)}(\tau=0)=0$ indicates *photon anti-bunching*, as the probability to count a second photon, immediately after a first one has been counted, vanishes. This is a direct result of the emission process of the source. Photons are emitted when an excited atom relaxes back to the ground state. If a photon is counted, the atom is likely to be in ground state and thus a finite time must elapse before it is re-excited and capable of emitting another one. The probability to find the atom in the excited state at time τ given that it starts in the ground state at $\tau=0$ is

$$P_e(\tau) = \frac{4\Omega^2}{\gamma^2+8\Omega^2}\left[1-e^{-3\gamma\tau/4}\left(\cosh\kappa\tau+\frac{3\gamma}{4\kappa}\sinh\kappa\tau\right)\right] \tag{10.75}$$

Comparison with (10.74) indicates this interpretation is correct. This prediction, first made by Carmichael and Walls [8], was one of the earliest examples of how quantum optics would differ from a semiclassical description of light. In Fig. 10.3 we plot $g^{(2)}(\tau)$ for two values of the Rabi frequency.

The first observation of photon antibunching was made by Kimble et al. in 1977 on atomic beams [11]. They saw a positive slope for $g^{(2)}(\tau)$ which is consistent with the predictions of the theory, however fluctuations from atomic numbers in the beam made a detailed comparison with the single atom result impossible. Ion traps

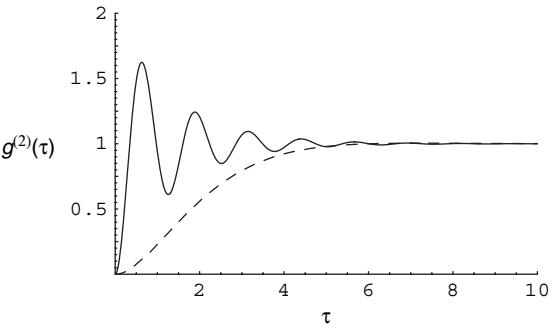

Fig. 10.3 The second order correlation function of the fluorescent light, given by (10.74) versus delay time τ. The *solid line* corresponds to $\Omega = 2.5$, while the *dashed line* corresponds to $\Omega = 0.25$. In both cases $\gamma = 1.0$

(see Chap. 17) provided a means to observe photon antibunching from a single atom [12]. In Fig. 10.4 we show the results of a measurement of the second order correlation function performed on a single trapped mercury ion by Walther's Garching group [13].

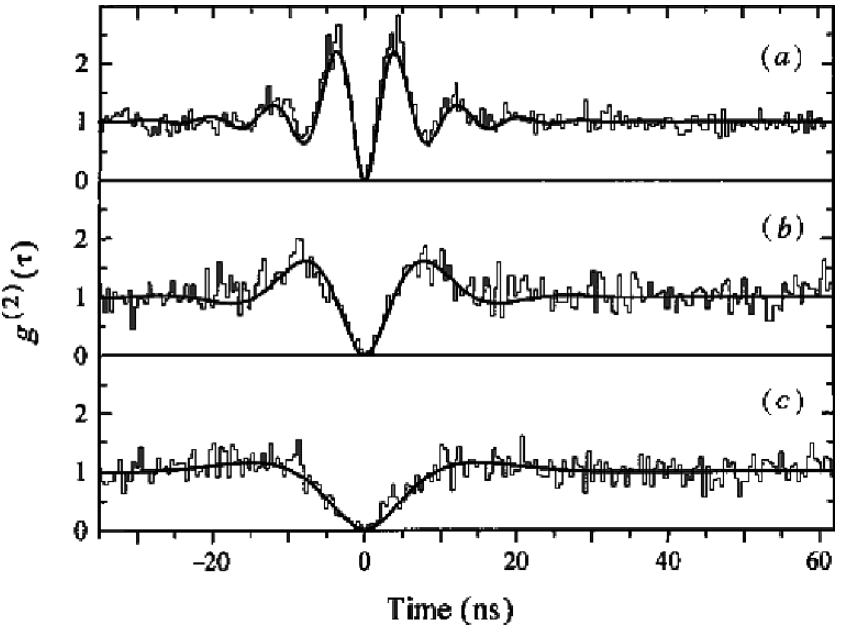

Fig. 10.4 The second order correlation function of the fluorescent light form a single mercury ion in a trap versus delay, τ. (**a**)$\Delta = -2.3\gamma$, $\Omega = 2.8\gamma$. (**b**)$\Delta = -1.1\gamma$, $\Omega = \gamma$, (**c**)$\Delta = -0.5\gamma$, $\Omega = 0.6\gamma$ (from [13])

Exercises

10.1 In the Jaynes–Cummings model, show that if the atom begins in the ground state and the field begins in the state $|\phi\rangle = \Sigma f_n |n\rangle$, the probability to find the atom in the excited state at time $t > 0$ is given by

$$p_e(t) = \sum_{n=1}^{\infty} |f_n|^2 \sin^2(\Omega_{n-1} t) \tag{10.76}$$

10.2 In the Jaynes–Cummings model, show that if the atom begins in the ground state and the field begins in the state $|\phi\rangle = \Sigma f_n |n\rangle$, the state at time $t > 0$ is the entangled state

$$|\psi(t)\rangle = |\phi_g(t)\rangle|g\rangle + |\phi_e(t)\rangle|e\rangle \tag{10.77}$$

where

$$|\phi_g(t)\rangle = \sum_n f_n \cos(\Omega_{n-1} t) \tag{10.78}$$

$$|\phi_e(t)\rangle = \mathrm{i} \sum_n f_n \sin(\Omega_{n-1} t) \tag{10.79}$$

In the case of the field initially in a coherent state, plot the Q-functions for $|\phi_e(t)\rangle$ and $|\phi_e(t)\rangle$ at times equal to half way to the first revival and at the first revival.

10.3 Compute the dressed states, and the corresponding eigenvalues, for the case in which the field mode is detuned from the atomic resonance, $\Delta = \omega_a - \omega_c \neq 0$.

10.4 Define the trace-preserving density operator map on the state of a single two-level system,

$$\rho \mapsto \mathscr{E}(\rho) = \sigma_z \rho \sigma_z \tag{10.80}$$

Sow that this leaves unchanged the diagonal elements of ρ in the eigenstates of σ_z, but changes the phase of the off-diagonal elements by π.

10.5 Calculate the second order correlation function $g^{(2)}(\tau)$ for resonance fluorescence in the presence of atomic dephasing (see (10.46).

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