

Chapter 2

Single Two-Level Atom

2.1 Overview

We will start our discussion with the most simple system: a single two-level atom interacting with an electromagnetic field. The models used in later Chapters typically treat both the atom and the field classically. However, we will begin here by considering the quantum mechanics of the system, showing that when the electric field is in a sufficiently weak coherent state, the atomic dynamics can be fully described by a classical electric dipole interacting with a classical electric field. Spending some time looking at the quantum model will prove beneficial for understanding the limitations and strengths of the classical model.

The theory presented in this Chapter closely follows the discussions of Zoller and Gardiner which can be found in Chap. 12 of [1] and Chaps. 9 and 14 of [2].

2.2 System Hamiltonian

2.2.1 Two-Level Atom

One of the simplest cases we can consider is that of a single two-level atom, with ground state $|g\rangle$ and excited state $|e\rangle$ (see Fig. 2.1). The energy difference between these two states is $E_e - E_g = \hbar\omega_0$, where $E_{g(e)}$ is the ground (excited) state energy and ω_0 is the resonant transition frequency. Assuming the two states form a closed transition, then the excited state decays solely to the ground state with decay rate $\Gamma_0 \equiv 2\gamma_0$,¹ corresponding to an excited state lifetime of $\tau_0 = 1/\Gamma_0$. We will show in Sect. 2.3.3 that this decay arises due to coupling between the atom and the quantum vacuum field.

¹For convenience, we will use both the decay rate Γ and half-decay rate γ at different points in this Thesis.

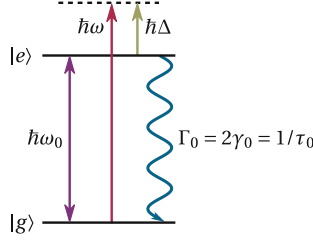


Fig. 2.1 Ground $|g\rangle$ and excited $|e\rangle$ energy levels of a two-level atom. The energy levels are separated by an energy $E = \hbar\omega_0$. A driving field with frequency ω is detuned from the bare atomic transition by $\Delta = \omega - \omega_0$. The excited state decays to the ground state with decay rate $\Gamma_0 = 2\gamma_0 = 1/\tau_0$

In following Sections we will apply a light field to the atom which will couple the two electronic quantum states. In practice, real atoms have many states, some of which may be degenerate. This degeneracy can usually be broken by applying appropriate electric or magnetic fields to the atom (see Sect. 3.2.1), and so assuming that the frequency of the driving field ω is close to the resonant atomic frequency ω_0 (and far detuned from the transitions to all other states), then these other energy levels can be treated as a perturbation, resulting in Stark shifts of the ground and excited state energies (see Chap. 4 of [2]). These shifts are not important for the features we will be considering and so we shall ignore them.

The Hamiltonian describing the bare energies of the two energy levels can be written as

$$H_0 = E_g |g\rangle\langle g| + E_e |e\rangle\langle e|. \quad (2.2.1)$$

Without loss of generality, we can set the ground state energy $E_g = 0$, in which case

$$H_0 = \hbar\omega_0 |e\rangle\langle e|. \quad (2.2.2)$$

2.2.2 Vector Notation

It can be convenient to represent $|g\rangle$ and $|e\rangle$ in vector notation,

$$|g\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \quad |e\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix}. \quad (2.2.3)$$

Note that other texts (e.g., [1–3]) will sometimes use the opposite notation $|g\rangle = (0, 1)^T$ and $|e\rangle = (1, 0)^T$, where T stands for transpose. We can define raising and lowering operators

$$\sigma^+ \equiv |e\rangle\langle g| = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} (1 \ 0) = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix}, \quad \sigma^- \equiv |g\rangle\langle e| = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} (0 \ 1) = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}. \quad (2.2.4)$$

In addition, the Pauli matrices present a convenient basis for describing two-level systems,

$$\sigma^x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \sigma^y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \quad \sigma^z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}. \quad (2.2.5)$$

Using this notation, the bare Hamiltonian in Eq. (2.2.2) can be written as

$$H_0 = \hbar\omega_0 \sigma^+ \sigma^- = \begin{pmatrix} 0 & 0 \\ 0 & \hbar\omega_0 \end{pmatrix}. \quad (2.2.6)$$

2.2.3 Density Matrix

A convenient representation for describing the behaviour of a quantum state with wavefunction $|\Psi\rangle$ is as a density operator, $\rho \equiv |\Psi\rangle\langle\Psi|$.² For the two-level atom, the corresponding density operator (density matrix) in bra-ket and matrix notation is

$$\rho = (a |g\rangle + b |e\rangle) (a^* \langle g| + b^* \langle e|) = \begin{pmatrix} \rho_{gg} & \rho_{ge} \\ \rho_{eg} & \rho_{ee} \end{pmatrix}, \quad (2.2.7)$$

where $\rho_{gg} = |a|^2$ and $\rho_{ee} = |b|^2$ are the ground and excited state *populations* respectively, and $\rho_{ge} = ab^*$ and $\rho_{eg} = ba^*$ are the *coherences*. The requirement that the total population be 1 means that the trace of ρ is $\text{Tr}(\rho) = \rho_{gg} + \rho_{ee} = 1$, and the coherences are related by $\rho_{eg} = \rho_{ge}^*$. The expectation of an operator $\mathbf{A}$ for a state with density matrix ρ can be found by calculating the trace

$$\langle \mathbf{A} \rangle = \text{Tr}(\rho \mathbf{A}). \quad (2.2.8)$$

In addition to a two-level atom, the total system also contains electromagnetic (EM) field modes (Sect. 2.2.5), and so the overall wavefunction $|\Psi_{\text{sys}}\rangle$ describes the field as well as the atomic state. However, in Sect. 2.3.6, we will trace away the field parts of the density matrix, being left with just the atomic density matrix in Eq. (2.2.7).

²A density operator can be used to describe a statistical mixed state of quantum states that cannot be described by one single state vector, $\rho = \sum_i p_i |\Psi_i\rangle\langle\Psi_i|$.

2.2.4 Dipole Operator

We assume the energy levels $|g\rangle$ and $|e\rangle$ are coupled via an electric dipole transition. The quantum operator for the dipole moment is

$$\mathbf{d} \equiv \mathbf{d}_{eg} |e\rangle\langle g| + \mathbf{d}_{ge} |g\rangle\langle e|, \quad (2.2.9)$$

where $\mathbf{d}_{ge} = \langle g|\mathbf{d}|e\rangle = e \langle g|\mathbf{s}|e\rangle = \mathbf{d}_{eg}^*$ is the dipole matrix element, e is the electron charge and $\mathbf{s}$ the electron displacement operator.³ The expectation value of the dipole moment operator is given by

$$\langle \mathbf{d} \rangle = \text{Tr}(\rho \mathbf{d}) = \mathbf{d}_{ge} \rho_{eg} + \mathbf{d}_{eg} \rho_{ge}. \quad (2.2.10)$$

The dipole expectation value is therefore related to the coherence ρ_{eg} . In Sect. 3.2.1 of the next Chapter, we show that the vector polarisation of the dipole transition $\mathbf{d}$ is related to the nature of the ground and excited states. We leave further discussion of this topic to that Section, and for now assume that the transition between the two states results in a dipole polarisation identical to the polarisation of the driving field.

2.2.5 Electric Field Operator

In addition to the atomic states, the system also contains EM field modes $|\Lambda\rangle \equiv |\mathbf{k}, \hat{\mathbf{e}}_{\varepsilon_{\mathbf{k}}}\rangle$. The mode index Λ corresponds to a spatial mode with propagation wavevector $\mathbf{k}$ (in a plane wave basis) and polarisation $\hat{\mathbf{e}}_{\varepsilon_{\mathbf{k}}}$. For a given $\mathbf{k}$, there are two possible polarisations (with index $\varepsilon_{\mathbf{k}} \in \{1, 2\}$), both of which are orthogonal to $\mathbf{k}$ and orthogonal to each other, $(\hat{\mathbf{e}}_{\varepsilon_{\mathbf{k}}})^* \cdot \hat{\mathbf{e}}_{\varepsilon'_{\mathbf{k}}} = \delta_{\varepsilon_{\mathbf{k}}, \varepsilon'_{\mathbf{k}}}$. The wavevector $\mathbf{k} = k \hat{\mathbf{k}}$ has unit vector $\hat{\mathbf{k}}$ and wavenumber $k = |\mathbf{k}|$ with corresponding wavelength $\lambda = 2\pi/k$ and frequency $\omega_k = ck$.

The total electric field operator is (see Eqs. (11.1.43) and (11.1.48) in [1])

$$\mathbf{E}(\mathbf{r}) = i \sum_{\Lambda} \sqrt{\frac{\hbar \omega_k}{2\varepsilon_0}} \left[\mathbf{a}_{\Lambda} \mathbf{u}_{\Lambda}(\mathbf{r}) - \mathbf{a}_{\Lambda}^{\dagger} \mathbf{u}_{\Lambda}^*(\mathbf{r}) \right], \quad (2.2.11)$$

where $\sum_{\Lambda} = \sum_{\mathbf{k}} \sum_{\varepsilon_{\mathbf{k}}}$, the operator $\mathbf{a}^{\dagger}$ ($\mathbf{a}$) creates (annihilates) a photon in mode Λ , ε_0 is the permittivity of free space, $\mathbf{u}_{\Lambda}(\mathbf{r})$ are the set of plane wave mode functions,

³The mass of the positively charged atom core (nucleus plus core electrons) is very much larger than the electron and so is assumed to be unmoved by the electric field.

$$\mathbf{u}_\Lambda(\mathbf{r}) = \frac{1}{\sqrt{V}} \hat{\mathbf{e}}_{\varepsilon_k} e^{i\mathbf{k} \cdot \mathbf{r}}, \quad (2.2.12)$$

and V is the quantisation mode volume. There is also a corresponding magnetic field $\mathbf{B}$ which can be related to the electric field via the vector potential operator $\mathbf{A}$ (see, e.g., Sect. 6.2 of [4]). In Sect. 2.3.5, we will show that if the electric field is in a coherent state, it can be treated as a classical electric field. The energy of the field modes is

$$H_{\text{EM}} = \sum_{\Lambda} \hbar \omega_k \left(a_{\Lambda}^{\dagger} a_{\Lambda} + \frac{1}{2} \right). \quad (2.2.13)$$

2.2.6 Atom–Field Coupling

An electric field couples the atomic ground and excited states, as shown in Fig. 2.1. This induces atomic coherences ρ_{eg} between the ground and excited states which as we saw in Sect. 2.2.4 results in a nonzero electric dipole moment in the atom. The interaction energy between an electric dipole and an electric field is

$$H_{\text{int}} = -\mathbf{d} \cdot \mathbf{E}(\mathbf{r}_0), \quad (2.2.14)$$

where $\mathbf{r}_0$ is the atom position. The size of the atom ($\sim 10^{-10}$ m) is much smaller than the wavelength of the driving light ($\lambda \sim 10^{-6}$ m), and so we can assume that the field is constant over the extent of the atom. This is known as the *dipole approximation* and also allows us to ignore higher order multipolar terms (see Sect. 4.2 of [3]).

Substituting in the electric dipole operator (2.2.9) and electric field operator (2.2.11), the interaction energy becomes

$$H_{\text{int}} = i\hbar\sigma^- \sum_{\Lambda} \left(a_{\Lambda}^{\dagger} \kappa_{\Lambda}^* e^{-i\mathbf{k} \cdot \mathbf{r}_0} - a_{\Lambda} \tilde{\kappa}_{\Lambda}^* e^{i\mathbf{k} \cdot \mathbf{r}_0} \right) - i\hbar\sigma^+ \sum_{\Lambda} \left(a_{\Lambda} \kappa_{\Lambda} e^{i\mathbf{k} \cdot \mathbf{r}_0} - a_{\Lambda}^{\dagger} \tilde{\kappa}_{\Lambda} e^{-i\mathbf{k} \cdot \mathbf{r}_0} \right), \quad (2.2.15)$$

where we have defined the coupling coefficients as⁴

$$\kappa_{\Lambda} \equiv \sqrt{\frac{\omega_k}{2\hbar\varepsilon_0}} \mathbf{d}_{eg} \cdot \mathbf{u}_{\Lambda}(0), \quad \tilde{\kappa}_{\Lambda} \equiv \sqrt{\frac{\omega_k}{2\hbar\varepsilon_0}} \mathbf{d}_{eg} \cdot \mathbf{u}_{\Lambda}^*(0), \quad (2.2.16)$$

⁴The signs of κ_{Λ} and $\tilde{\kappa}_{\Lambda}$ in Eq. (12.1.28) of [1] differ to those here. However, the quantities of interest are $|\kappa_{\Lambda}|^2$ and $|\tilde{\kappa}_{\Lambda}|^2$ and so a sign difference is not important.

2.2.7 Interaction Picture, Rotating Wave Approximation

The dynamics we are interested in are those involving the coupling between the atom and the electric field modes. It is therefore convenient to transform into the *interaction picture*. Until now, we have been working in the *Schrödinger picture*, that is, the frame in which the operators H_S are time-*independent* and the state vectors $|\Psi_S(t)\rangle$ are time-*dependent*. The evolution of the state vectors in the Schrödinger picture is governed by the Schrödinger equation,

$$\frac{d}{dt} |\Psi_S(t)\rangle = -\frac{i}{\hbar} H_{\text{tot},S} |\Psi_S(t)\rangle, \quad (2.2.17)$$

where the Hamiltonians $H_{\text{tot},S} = H_{0,S} + H_{\text{EM},S} + H_{\text{int},S}$ were defined in (2.2.2), (2.2.13) and (2.2.14) respectively. In the *interaction picture*, the operators evolve in time due to the free Hamiltonian (as if there were no atom–field interaction), leaving the state vectors to evolve dependent only on the interaction Hamiltonian H_{int} . The interaction picture operators and state vectors are related to the Schrödinger picture by the following transformation:

$$\begin{aligned} U_I(t) &= \exp \left[-\frac{i}{\hbar} (H_{0,S} + H_{\text{EM},S}) t \right], \\ H_I(t) &= U_I^\dagger(t) H_S U_I(t), \\ |\Psi_I(t)\rangle &= U_I^\dagger(t) |\Psi_S(t)\rangle. \end{aligned} \quad (2.2.18)$$

Since both $H_{0,S}$ and $H_{\text{EM},S}$ are diagonal in atomic and field operators respectively, U_I obeys the general rule for exponentials of diagonal matrices,

$$\exp \begin{pmatrix} X & & & \\ & X & & \\ & & X & \\ & & & \ddots \end{pmatrix} = \begin{pmatrix} e^X & 0 & 0 & \dots \\ 0 & e^X & 0 & \dots \\ 0 & 0 & e^X & \dots \\ \vdots & \vdots & \vdots & \ddots \end{pmatrix}. \quad (2.2.19)$$

The bare atomic Hamiltonian has the form (2.2.2)

$$H_{0,S} = 0 |g\rangle\langle g| + \hbar\omega_0 |e\rangle\langle e|. \quad (2.2.20)$$

Notice here that we have included the additional ground state term, even though we previously set the ground state energy to zero.⁵ When we take the exponential of $H_{0,S}$, the ground state is still important:

⁵The choice of ground state energy is arbitrary, since E_e is defined relative to E_g .

$$\exp\left(-\frac{i}{\hbar}H_{0,S}t\right) = \exp\left[-\frac{i}{\hbar}\left(0|g\rangle\langle g| + \hbar\omega_0|e\rangle\langle e|\right)t\right] = |g\rangle\langle g|e^0 + |e\rangle\langle e|e^{-i\omega_0 t}. \quad (2.2.21)$$

The atomic raising operator in the interaction picture is then⁶

$$\begin{aligned} \sigma_I^+(t) &= U_I^\dagger \sigma_S^+ U_I \\ &= (|g\rangle\langle g| + |e\rangle\langle e|e^{i\omega_0 t}) |e\rangle\langle g| (|g\rangle\langle g| + |e\rangle\langle e|e^{-i\omega_0 t}) \\ &= e^{i\omega_0 t} |e\rangle\langle g| = e^{i\omega_0 t} \sigma_S^+, \end{aligned} \quad (2.2.22)$$

and the lowering operator is $\sigma_I^-(t) = [\sigma_I^+(t)]^\dagger = e^{-i\omega_0 t} \sigma_S^-$. The exponential of the EM field Hamiltonian (2.2.13) has a similar form:

$$\exp\left(-\frac{i}{\hbar}H_{\text{EM},S}t\right) = \exp\left\{-\frac{i}{\hbar}\left[\sum_{\Lambda}\hbar\omega_k\left(\mathbf{a}_{\Lambda}^\dagger\mathbf{a}_{\Lambda} + \frac{1}{2}\right)\right]t\right\}. \quad (2.2.23)$$

It is convenient here to express the creation and annihilation operators in the number state basis,

$$a_{\Lambda}^\dagger = \sum_{n=0}^{\infty} \sqrt{n+1} |\Lambda, n+1\rangle\langle\Lambda, n|, \quad a_{\Lambda} = \sum_{n=1}^{\infty} \sqrt{n} |\Lambda, n-1\rangle\langle\Lambda, n|, \quad (2.2.24)$$

where $|\Lambda, n\rangle$ is the EM field state with n photons in mode Λ . The exponential in Eq. (2.2.23) then becomes

$$\begin{aligned} \exp\left\{-\frac{i}{\hbar}\left[\sum_{\Lambda}\hbar\omega_k\left(\frac{1}{2} + \mathbf{a}_{\Lambda}^\dagger\mathbf{a}_{\Lambda}\right)\right]t\right\} &= \exp\left\{-\frac{i}{\hbar}\left[\sum_{\Lambda}\hbar\omega_k\left(\frac{1}{2} + \sum_{n=0}^{\infty} n |\Lambda, n\rangle\langle\Lambda, n|\right)\right]t\right\} \\ &= \sum_{\Lambda} e^{-i\omega_k t/2} \sum_{n=0}^{\infty} e^{-in\omega_k t} |\Lambda, n\rangle\langle\Lambda, n|. \end{aligned} \quad (2.2.25)$$

An EM field creation operator in the interaction picture is therefore

$$\begin{aligned} \mathbf{a}_{\Lambda,I}^\dagger(t) &= U_I^\dagger(t) \mathbf{a}_{\Lambda,S}^\dagger U_I(t) \\ &= \left(\sum_{\Lambda'} e^{i\omega_{k'} t/2} \sum_{n'=0}^{\infty} e^{in'\omega_{k'} t} |\Lambda', n'\rangle\langle\Lambda', n'|\right) \sum_{n=0}^{\infty} \sqrt{n+1} |\Lambda, n+1\rangle\langle\Lambda, n| \\ &\quad \times \left(\sum_{\Lambda''} e^{-i\omega_{k''} t/2} \sum_{n''=0}^{\infty} e^{-in''\omega_{k''} t} |\Lambda'', n''\rangle\langle\Lambda'', n''|\right) \\ &= \sum_{\Lambda'\Lambda''} e^{i(\omega_{k'} - \omega_{k''})t/2} \sum_{n=0}^{\infty} \sum_{n'=0}^{\infty} \sum_{n''=0}^{\infty} e^{i(n'\omega_{k'} - n''\omega_{k''})t} \sqrt{n+1} |\Lambda', n'\rangle\langle\Lambda'', n''| \end{aligned}$$

⁶The EM field part of the Hamiltonian has no effect on the atomic operator, and vice versa.

$$\begin{aligned}
& \times \delta_{\Lambda', \Lambda} \delta_{\Lambda'', \Lambda} \delta_{n', n+1} \delta_{n, n''} \\
& = e^{i\omega_k t} \sum_{n=0}^{\infty} \sqrt{n+1} |\Lambda, n+1\rangle \langle \Lambda, n| \\
& = e^{i\omega_k t} a_{\Lambda, S}^{\dagger}.
\end{aligned} \tag{2.2.26}$$

The corresponding annihilation operator is $\mathbf{a}_{\Lambda, I}(t) = [\mathbf{a}_{\Lambda, I}^{\dagger}(t)]^{\dagger} = e^{-i\omega_k t} \mathbf{a}_{\Lambda, S}$.

The bare atomic Hamiltonian and field Hamiltonian are therefore unchanged in the interaction picture,

$$\mathbf{H}_{0, S} = \hbar\omega_0 |e\rangle \langle e| = \hbar\omega_0 \sigma_S^+ \sigma_S^- = \hbar\omega_0 \sigma_I^+(t) \sigma_I^-(t) = \mathbf{H}_{0, I}(t), \tag{2.2.27a}$$

$$\mathbf{H}_{\text{EM}, S} = \sum_{\Lambda} \hbar\omega_k \left(a_S^{\dagger} a_S + \frac{1}{2} \right) = \sum_{\Lambda} \hbar\omega_k \left(a_I^{\dagger}(t) a_I(t) + \frac{1}{2} \right) = \mathbf{H}_{\text{EM}, I}(t). \tag{2.2.27b}$$

The interaction Hamiltonian (2.2.15) on the other hand becomes

$$\begin{aligned}
\mathbf{H}_{\text{int}, I}(t) = & i\hbar \sigma_S^- \sum_{\Lambda} \left(\mathbf{a}_{\Lambda, S}^{\dagger} \kappa_{\Lambda}^* e^{i(\omega_k - \omega_0)t - i\mathbf{k} \cdot \mathbf{r}_0} - \mathbf{a}_{\Lambda, S} \tilde{\kappa}_{\Lambda}^* e^{-i(\omega_k + \omega_0)t + i\mathbf{k} \cdot \mathbf{r}_0} \right) \\
& - i\hbar \sigma_S^+ \sum_{\Lambda} \left(\mathbf{a}_{\Lambda, S} \kappa_{\Lambda} e^{-i(\omega_k - \omega_0)t + i\mathbf{k} \cdot \mathbf{r}_0} - \mathbf{a}_{\Lambda, S}^{\dagger} \tilde{\kappa}_{\Lambda} e^{i(\omega_k + \omega_0)t - i\mathbf{k} \cdot \mathbf{r}_0} \right).
\end{aligned} \tag{2.2.28}$$

Notice that we have kept the atomic and field operators in their time-independent Schrödinger picture forms. By convention, we will omit the subscript S to refer to the Schrödinger picture and assume that unless stated explicitly, all operators and state vectors are in the Schrödinger picture.

Using these results, the Schrödinger equation in the interaction picture becomes

$$\begin{aligned}
\frac{d}{dt} |\Psi_I(t)\rangle & = \frac{d}{dt} \mathbf{U}_I^{\dagger}(t) |\Psi_S(t)\rangle \\
& = \left[\frac{d}{dt} \mathbf{U}_I^{\dagger}(t) \right] |\Psi_S(t)\rangle + \mathbf{U}_I^{\dagger}(t) \left[\frac{d}{dt} |\Psi_S(t)\rangle \right] \\
& = \left[\frac{i}{\hbar} (\mathbf{H}_{0, S} + \mathbf{H}_{\text{EM}, S}) \right] \mathbf{U}_I^{\dagger}(t) |\Psi_S(t)\rangle - \frac{i}{\hbar} \mathbf{U}_I^{\dagger}(t) (\mathbf{H}_{0, S} + \mathbf{H}_{\text{EM}, S} + \mathbf{H}_{\text{int}, S}) |\Psi_S(t)\rangle \\
& = -\frac{i}{\hbar} \left[\mathbf{U}_I^{\dagger}(t) \mathbf{H}_{\text{int}, S} \mathbf{U}_I(t) \right] \left[\mathbf{U}_I^{\dagger}(t) |\Psi_S(t)\rangle \right] \\
& = -\frac{i}{\hbar} \mathbf{H}_{\text{int}, I}(t) |\Psi_I(t)\rangle.
\end{aligned} \tag{2.2.29}$$

This is also known as the *Schwinger–Tomonaga equation*. As already mentioned, the evolution of $|\Psi_I(t)\rangle$ therefore only depends on the interaction Hamiltonian.

2.2.8 Rotating Wave Approximation

There are two different contributions to the atom–field interaction that we need to consider. Firstly, the atom interacts with an external driving field, which we will assume to be both monochromatic and close to resonance ($\omega_k \simeq \omega_0$). Secondly, the atom also interacts with the quantum vacuum field, spontaneously and randomly emitting and absorbing photons into and from all possible field modes. However, we can restrict the sum over modes Λ in (2.2.28) to a small range around ω_0 , $|\omega_k - \omega_0| < \vartheta$, where ϑ is the *coupling bandwidth* (see Sect. 9.1 of [2]).⁷ This will allow us to set appropriate time scales for the Born–Markov approximations discussed later in Sect. 2.3.3.

With these two considerations, it becomes clear that the $\omega_k + \omega_0$ terms in Eq. (2.2.28) will oscillate very much faster than any of the other time scales involved, and these terms can thus be dropped from H_{int} ,

$$H_{\text{int},I}(t) \simeq i\hbar \sum_{|\omega_k - \omega_0| < \vartheta} \left(\sigma^- a_{\Lambda}^{\dagger} \kappa_{\Lambda}^* e^{i(\omega_k - \omega_0)t - i\mathbf{k} \cdot \mathbf{r}_0} - \sigma^+ a_{\Lambda} \kappa_{\Lambda} e^{-i(\omega_k - \omega_0)t + i\mathbf{k} \cdot \mathbf{r}_0} \right). \quad (2.2.30)$$

The two remaining terms correspond physically to the atom emitting a photon into mode Λ ($\sigma^- a_{\Lambda}^{\dagger}$) and the atom absorbing a photon from mode Λ ($\sigma^+ a_{\Lambda}$). The two terms that were discarded corresponded to the atom decaying and the field gaining a photon simultaneously and vice versa, which is clearly much less likely for the case of resonant driving. There are many situations in which these non-resonant terms are important [5–7], although that is beyond the scope of this Thesis.

2.3 Dissipation: Quantum Stochastic Schrödinger Equation and the Master Equation

2.3.1 Overview

We now have a Schrödinger equation (2.2.29) which describes the dynamics of the system in the interaction picture. If it were possible to follow the evolution of the EM field states, the evolution of the system would be coherent. However, in tracing over the EM field, the atomic evolution becomes noisy resulting in dissipation, i.e. loss of energy and information into the EM field from the atomic system. In this Section, we will employ one of the stochastic methods of modelling this dissipation: the *quantum stochastic Schrödinger equation* (again, following the discussion from Chaps. 9 and 14 of [1]). From this we will derive a master equation which will describe the full dissipative system dynamics.

⁷It is still assumed that we sum over the two polarisations as well.

2.3.2 Noise Operator

The atom–field interaction Hamiltonian in the interaction picture and the rotating wave approximation (2.2.30) can be written as

$$H_{\text{int},I}(t) = i\hbar [\mathbf{f}^\dagger(t)\sigma^- - \sigma^+\mathbf{f}(t)], \quad (2.3.1)$$

where $\mathbf{f}(t)$ is a time-dependent noise operator

$$\mathbf{f}(t) \equiv \sum_{|\omega_k - \omega_0| < \vartheta} \kappa_\Lambda \mathbf{a}_\Lambda e^{-i(\omega_k - \omega_0)t + i\mathbf{k} \cdot \mathbf{r}_0}. \quad (2.3.2)$$

These noise operators have the commutation relation

$$[\mathbf{f}(t), \mathbf{f}^\dagger(t')] = \gamma(t - t'), \quad (2.3.3)$$

where

$$\begin{aligned} \gamma(\tau) &= \sum_{|\omega_k - \omega_0| < \vartheta} |\kappa_\Lambda|^2 e^{-i(\omega_k - \omega_0)\tau}, \\ &\rightarrow \int_{\omega_0 - \vartheta}^{\omega_0 + \vartheta} d\omega_k g(\omega_k) |\kappa(\omega_k)|^2 e^{-i(\omega_k - \omega_0)\tau}, \end{aligned} \quad (2.3.4)$$

where we have assumed the modes are a continuum with a density of states $g(\omega_k)$. We have also made use of the bosonic commutation relation $[\mathbf{a}_\Lambda, \mathbf{a}_{\Lambda'}^\dagger] = \delta_{\Lambda, \Lambda'}$. One of the requirements of the choice of coupling bandwidth ϑ is that $g(\omega_k)|\kappa(\omega_k)|$ varies smoothly over the range $(\omega_0 \pm \vartheta)$.

2.3.3 Born–Markov Approximation

The interaction picture Schrödinger equation (2.2.29) is

$$\frac{d}{dt} |\Psi_I(t)\rangle = [\sigma^- \mathbf{f}^\dagger(t) - \sigma^+ \mathbf{f}(t)] |\Psi_I(t)\rangle, \quad (2.3.5)$$

where we have assumed that the initial state is a product of the atomic and field states $|\Psi_I(0)\rangle = |\Psi_{0,I}(0)\rangle \otimes |\Psi_{\text{EM},I}(0)\rangle$ and so the atomic ($\sigma^\pm$) and field ($f, f^\dagger$) operators commute. Integrating Eq. (2.3.5) from $t = 0$ to t gives⁸

⁸If $dy(t)/dt = x(t)y(t)$ then $\int_0^t \dot{y}(t_1) dt_1 = \int_0^t x(t_1)y(t_1) dt_1 = \int_0^t x(t_1)y(0) dt_1 + \int_0^t x(t_1) \left(\int_0^{t_1} \dot{y}(t_2) dt_2 \right) dt_1$.

$$\begin{aligned}
|\Psi_I(t)\rangle = & \left\{ 1 + \int_0^t dt_1 [\sigma^- \mathbf{f}^\dagger(t_1) - \sigma^+ \mathbf{f}(t_1)] \right\} |\Psi_I(0)\rangle \\
& + \int_0^t dt_1 [\sigma^- \mathbf{f}^\dagger(t_1) - \sigma^+ \mathbf{f}(t_1)] \int_0^{t_1} dt_2 [\sigma^- \mathbf{f}^\dagger(t_2) - \sigma^+ \mathbf{f}(t_2)] |\Psi_I(t_2)\rangle.
\end{aligned} \tag{2.3.6}$$

Let us consider a time interval $0 < t_2 < \Delta t$. We can replace $|\Psi_I(t_2)\rangle \simeq |\Psi_I(0)\rangle$ in the second line of (2.3.6) if we assume that interaction is sufficiently weak. This is known as the *Born approximation*. If we also assume that the field is initially in the vacuum state such that $\mathbf{f} |\Psi_I(0)\rangle = 0$ and use the commutation relation from (2.3.3), then the state vector after the time interval Δt is

$$\begin{aligned}
|\Psi_I(\Delta t)\rangle = & \left[1 - \sigma^+ \sigma^- \int_0^{\Delta t} dt_1 \int_0^{t_1} dt_2 \gamma(t_1 - t_2) \right] |\Psi_I(0)\rangle \\
& + \sigma^- \int_0^{\Delta t} dt_1 \mathbf{f}^\dagger(t_1) |\Psi_I(0)\rangle \\
& + \sigma^- \sigma^- \int_0^{\Delta t} dt_1 \int_0^{t_1} dt_2 \mathbf{f}^\dagger(t_1) \mathbf{f}^\dagger(t_2) |\Psi_I(0)\rangle.
\end{aligned} \tag{2.3.7}$$

The three lines in Eq. (2.3.7) correspond respectively to states with zero, one and two photons. For sufficiently small Δt , the probability of the two-photon state is negligible (order Δt^2) and so only the first two lines need be considered.

If we further assume that $\Delta t \gg 1/\vartheta$, then we can approximate

$$\gamma(t_1 - t_2) \simeq 2 \left(\frac{1}{2} \Gamma + i\delta\omega \right) \delta(t_1 - t_2), \tag{2.3.8}$$

where $\delta(t_1 - t_2)$ is the Dirac delta function, and the damping constant Γ and line-shift $\delta\omega$ are given by

$$\Gamma = 2\pi g(\omega_0) |\kappa(\omega_0)|^2, \tag{2.3.9a}$$

$$\delta\omega = -iP \int_{\omega_0 - \vartheta}^{\omega_0 + \vartheta} d\omega_k \frac{g(\omega_k) |\kappa(\omega_k)|^2}{\omega_k - \omega_0}, \tag{2.3.9b}$$

where P is the principal value integral (Eq. 12.2.21 of [1] and Eq. C.2.7 in Appendix C). This is known as the *Markov approximation* and has the physical significance that the free system dynamics happen on a much faster time scale (ω_0, ω_k) than the evolution in the interaction picture ($\Gamma, \delta\omega$). The double integral in the first line of Eq. (2.3.7) then becomes

$$\int_0^{\Delta t} dt_1 \int_0^{t_1} dt_2 \gamma(t_1 - t_2) \simeq \left(\frac{1}{2} \Gamma + i\delta\omega \right) \Delta t. \tag{2.3.10}$$

Substituting in κ from (2.3.7) and integrating over the quantisation volume V , the damping constant Γ becomes (C.2.6)

$$\Gamma = \Gamma_0 = \frac{|\mathbf{d}_{eg}|^2 k_0^3}{3\pi \epsilon_0 \hbar}. \quad (2.3.11)$$

This is just the natural decay rate of the single two-level atom Γ_0 introduced in Sect. 2.2.1. The line-shift $\delta\omega$ can also be determined, although this requires careful renormalisation of $(\omega_k - \omega_0)$ terms in the denominator. In full quantum electrodynamics, this line-shift is the *Lamb shift*. From now on we shall assume that $\delta\omega$ is included within the definition of ω_0 and so can be ignored.

2.3.4 Quantum Stochastic Schrödinger Equation

The one-photon term in (2.3.7) can be redefined as a *quantum Ito increment* (see Sect. 4.1 of [1] and Sects. 9.3 and 21.2.2 of [2])

$$\Delta \mathbf{B}(t) \equiv \frac{1}{\sqrt{\Gamma_0}} \int_t^{t+\Delta t} \mathbf{f}(t_1) dt_1. \quad (2.3.12)$$

We cannot let $\Delta t \rightarrow 0$ (since $\Delta t \gg \vartheta$), although in the limit that the interaction strength $(\Gamma_0, \delta\omega)$ is very weak, (2.3.7) can be written as a differential equation

$$d|\Psi_I(t)\rangle = \left[-\frac{\Gamma_0}{2} \sigma^+ \sigma^- dt + \sqrt{\Gamma_0} \sigma^- d\mathbf{B}^\dagger(t) \right] |\Psi_I(t)\rangle. \quad (2.3.13)$$

This is known as the *quantum stochastic Schrödinger equation*.

Ito stochastic differential equations were originally used to describe classical Brownian motion of particles in thermal systems. Collisions between particles in these systems result in each particle experiencing a random force acting on its motion. What we are considering here is the quantum analog of this, where the dynamics of the internal atomic quantum state are being influenced by a random noise term due to quantum vacuum fluctuations. The classical Ito term describes the differential change in particle velocity; the quantum Ito term describes the equivalent differential change in the state wavefunction. It is assumed that the noise terms in first order average to zero, and so using the calculus resulting from the Ito formalism (Sect. 2.3.6), the expectations resulting from the state dynamics can be expressed in a way that removes these fluctuating noise terms. What this means is that whilst the state vector itself evolves noisily, the expectation of the state vector evolves smoothly and deterministically. More details can be found in Chaps. 3 and 4 of [1].

As an aside, one alternative method for solving the quantum stochastic Schrödinger equation directly is via the method of quantum jumps [8]. In this method, the wavefunction of the (atomic) system is allowed to evolve coherently with no dissipation. At random time intervals however, a decay event is assumed to occur, corresponding to the detection of a spontaneous photon. When this happens, the atomic wavefunction is instantaneously projected onto the ground state, and then once more allowed to evolve coherently until another decay event occurs. The time between decay events is a random variable with some characteristic lifetime. This process is repeated over many realisations, and the average of these realisations then converges to the full dissipative dynamics of the system. One advantage of this type of approach over a master equation approach, which we shall discuss in Sect. 2.3.6, is that the wavefunction has the square root of the dimension of the density matrix, which for large systems can greatly reduce the computational complexity.

2.3.5 Classical Driving Field

In Sect. 9.5 of [2] the authors consider the case where the EM field modes are initially each in a coherent state. A coherent state $|\beta_\Lambda\rangle$ is the eigenstate of the field annihilation operator,

$$a_\Lambda |\beta_\Lambda\rangle = \beta_\Lambda |\beta_\Lambda\rangle. \quad (2.3.14)$$

The noise operator acting on a coherent state $|\beta\rangle = \bigotimes_\Lambda |\beta_\Lambda\rangle$, where $\bigotimes_\Lambda$ is the tensor product over all field modes, is then

$$f(t) |\beta\rangle = F(t) |\beta\rangle, \quad (2.3.15)$$

where

$$F(t) = \sum_{|\omega_k - \omega_0| < \vartheta} \kappa_\Lambda \beta_\Lambda e^{-i(\omega_k - \omega_0)t + i\mathbf{k} \cdot \mathbf{r}_0}, \quad (2.3.16)$$

is a complex-valued function of t (and $\mathbf{r}_0$). When we use this new state vector (the coherent field in addition to the vacuum field), we obtain a similar expression for $|\Psi_I(\Delta t)\rangle$ (2.3.7), with terms corresponding to zero, one and two photons,

$$\begin{aligned} |\Psi_I(\Delta t)\rangle = & \left[1 - \sigma^+ \sigma^- \int_0^{\Delta t} dt_1 \int_0^{t_1} dt_2 \gamma(t_1 - t_2) \right] |\Psi_I(0)\rangle \\ & + \int_0^{\Delta t} dt_1 [\sigma^- f^\dagger(t_1) + \sigma^- F^*(t_1) - \sigma^+ F(t_1)] |\Psi_I(0)\rangle \\ & + (\text{two-photon terms}). \end{aligned} \quad (2.3.17)$$

The zero-photon term has remained unchanged and the two-photon term can again be neglected since the probability scales with Δt^2 .

The modified quantum stochastic Schrödinger equation (2.3.13) has the form

$$d|\Psi_I(t)\rangle = \left[-\frac{\Gamma_0}{2} \sigma^+ \sigma^- dt + \left(F^*(t) \sigma^- - F(t) \sigma^+ \right) dt + \sqrt{\Gamma_0} \sigma^- dB^\dagger(t) \right] |\Psi_I(t)\rangle, \quad (2.3.18)$$

where we have subtracted the coherent part of the noise operator from the quantum Ito increment,

$$\Delta B(t) \equiv \frac{1}{\sqrt{\Gamma_0}} \int_t^{t+\Delta t} \left[f(t_1) - F(t_1) \right] dt_1. \quad (2.3.19)$$

Comparing the first two terms in (2.3.18), we can see that the coherent state adds an additional Hamiltonian-like term, which has the form

$$\begin{aligned} F^*(t) \sigma^- - F(t) \sigma^+ &= \sum_{|\omega_k - \omega_0| < \vartheta} \left(\kappa_\Lambda^* \beta_\Lambda^* e^{i(\omega_k - \omega_0)t - i\mathbf{k} \cdot \mathbf{r}_0} \sigma^- - \kappa_\Lambda \beta_\Lambda e^{-i(\omega_k - \omega_0)t + i\mathbf{k} \cdot \mathbf{r}_0} \sigma^+ \right) \\ &= -\frac{i}{\hbar} \left[-\mathbf{d} \cdot \mathcal{E}_0(\mathbf{r}_0) \right], \end{aligned} \quad (2.3.20)$$

where $\mathbf{d} = \mathbf{d}_{ge} \sigma^- e^{-i\omega_0 t} + \mathbf{d}_{eg} \sigma^+ e^{i\omega_0 t}$ is the dipole operator and $\mathcal{E}_0$ is a classical electric field:

$$\begin{aligned} \mathcal{E}_0(\mathbf{r}, t) &= \mathbf{E}_0(\mathbf{r}, t) e^{-i\omega_k t} + \mathbf{E}_0^*(\mathbf{r}, t) e^{i\omega_k t} \\ &= 2 \operatorname{Re}[\mathbf{E}_0(\mathbf{r}, t)] \cos \omega_k t + 2 \operatorname{Im}[\mathbf{E}_0(\mathbf{r}, t)] \sin \omega_k t. \end{aligned} \quad (2.3.21)$$

We assume that the temporal profile function $\mathbf{E}_0(t)$ varies slowly on the optical time scale, $|\dot{\mathbf{E}}_0(t)| \ll \omega_k |\mathbf{E}_0(t)|$. In most of this Thesis we will just consider constant driving field amplitudes $\mathbf{E}_0(\mathbf{r}, t) = \mathbf{E}_0(\mathbf{r})$. The sum over modes has reduced to just a single mode with frequency ω_k (monochromatic) and polarisation $\hat{\mathbf{e}}$.

This is an important result: a coherent field state behaves the same as a classical electric field, with interaction Hamiltonian of the same form as H_{int} (2.2.14).

2.3.6 Master Equation

In its current form, the quantum stochastic Schrödinger equation

$$d|\Psi_I(t)\rangle = \left[-\frac{i}{\hbar} H_{\text{eff}, I} dt + \sqrt{\Gamma_0} \sigma^- dB^\dagger(t) \right] |\Psi_I(t)\rangle, \quad (2.3.22)$$

with effective (non-Hermitian) Hamiltonian

$$H_{\text{eff},I} = -i\hbar \frac{\Gamma_0}{2} \sigma^+ \sigma^- - \mathbf{d}_{eg} \cdot \mathbf{E}_0(\mathbf{r}_0, t) e^{-i(\omega_k - \omega_0)t} \sigma^+ - \mathbf{d}_{ge} \cdot \mathbf{E}_0^*(\mathbf{r}_0, t) e^{i(\omega_k - \omega_0)t} \sigma^-, \quad (2.3.23)$$

is inconvenient to work with given the presence of the quantum Ito term $d\mathbf{B}$. The advantage of the quantum Ito representation lies in the algebra rules that govern it (Eq. 9.3.21 in [2]):

$$\begin{aligned} d\mathbf{B}(t) d\mathbf{B}^\dagger(t) &= dt, \\ d\mathbf{B}^\dagger(t) d\mathbf{B}(t) &= d\mathbf{B}(t)^2 = d\mathbf{B}^\dagger(t)^2 = 0, \\ d\mathbf{B}(t)dt &= dt d\mathbf{B}(t) = d\mathbf{B}^\dagger(t)dt = dt d\mathbf{B}^\dagger(t) = dt^2 = 0. \end{aligned} \quad (2.3.24)$$

The calculus of two quantum Ito terms $\mathbf{B}$ and $\mathbf{C}$ also contains additional terms compared with the conventional calculus,

$$d(\mathbf{B}\mathbf{C}) = (d\mathbf{B})\mathbf{C} + \mathbf{B}(d\mathbf{C}) + (d\mathbf{B})(d\mathbf{C}). \quad (2.3.25)$$

A solution to (2.3.22) can be found using a method like that described in Sect. 2.3.4, involving many numerical stochastic realisations. It can, however, be convenient to consider the density matrix $\rho(t) \equiv |\Psi(t)\rangle\langle\Psi(t)|$ defined in (Sect. 2.2.3), as opposed to just the wavefunction $|\Psi(t)\rangle$ (for example, if considering a mixed state). Converting the quantum stochastic Schrödinger equation into a master equation will allow us to obtain equations of motion for the individual elements in the density matrix, rather than relying on numerical stochastic realisations. For particular limits, we will even be able to obtain analytic solutions to these equations of motion.

We are not concerned with the behaviour of the field part of the wavefunction and so we can extract just the atomic part of the density matrix by calculating the *reduced atomic density matrix*,

$$\rho_0(t) \equiv \text{Tr}_{\text{EM}} \left(|\Psi(t)\rangle\langle\Psi(t)| \right), \quad (2.3.26)$$

where Tr_{EM} is a trace over the field parts of the density matrix. Transforming back into the *Schrödinger picture*⁹ and using the quantum Ito calculus rules in (2.3.24) and (2.3.25), the nonzero terms left when considering a differential element of $d\rho_0(t)$ give us the single atom master equation [2],

$$\dot{\rho}_0(t) = -\frac{i}{\hbar} [\mathbf{H}_{\text{sys},S}, \rho_0(t)] + \frac{\Gamma_0}{2} (2\sigma^- \rho(t) \sigma^+ - \sigma^+ \sigma^- \rho(t) - \rho(t) \sigma^+ \sigma^-), \quad (2.3.27)$$

⁹We do not remove the oscillation of the EM field as this is no longer operator-valued.

where $[a, b] \equiv ab - ba$ is a commutator and the coherent dynamics are included in $\mathbf{H}_{\text{sys}}$,

$$\mathbf{H}_{\text{sys},S} = \hbar\omega_0 \sigma^+ \sigma^- - \mathbf{d}_{eg} \cdot \mathbf{E}_0(\mathbf{r}_0, t) e^{-i\omega_k t} \sigma^+ - \mathbf{d}_{ge} \cdot \mathbf{E}_0^*(\mathbf{r}_0, t) e^{i\omega_k t} \sigma^-. \quad (2.3.28)$$

The dissipation is now included in the three terms outside the commutator. The terms $(-\sigma^+ \sigma^- \rho - \rho \sigma^+ \sigma^-)$ come from the imaginary part of $\mathbf{H}_{\text{eff}}$. The quantum Ito term $(\sigma^- \rho \sigma^+)$ is known as the *recycling term* because it recycles population, conserving the trace of ρ .

The master equation in (2.3.27) can be expressed in terms of the well known *optical Bloch equations* for a single two-level atom,

$$\dot{\rho}_{ee} = -\dot{\rho}_{gg} = -\rho_{ee}\Gamma_0 - \frac{i}{\hbar} (\mathbf{d}_{ge} \cdot \mathbf{E}_0^* e^{i\omega_k t} \rho_{eg} - \mathbf{d}_{eg} \cdot \mathbf{E}_0 e^{-i\omega_k t} \rho_{ge}), \quad (2.3.29a)$$

$$\dot{\rho}_{eg} = \dot{\rho}_{ge}^* = -\left(i\omega_0 + \frac{\Gamma_0}{2}\right) \rho_{eg} - \frac{i}{\hbar} (\rho_{ee} - \rho_{gg}) \mathbf{d}_{eg} \cdot \mathbf{E}_0 e^{-i\omega_k t}. \quad (2.3.29b)$$

These can be calculated from the many-atom optical Bloch equations (D.5.4) and (D.5.6) in Appendix D by ignoring the many-atom terms and just considering a single excited energy level.

2.4 Dipole Moment

2.4.1 Equations of Motion

We have written the oscillating (classical) electric field in the form (2.3.21)

$$\begin{aligned} \mathcal{E}_0(\mathbf{r}, t) &= \mathbf{E}_0(\mathbf{r}, t) e^{-i\omega_k t} + \mathbf{E}_0^*(\mathbf{r}, t) e^{i\omega_k t} \\ &= 2 \mathbf{E}_0^{\Re}(\mathbf{r}, t) \cos \omega_k t + 2 \mathbf{E}_0^{\Im}(\mathbf{r}, t) \sin \omega_k t. \end{aligned} \quad (2.4.1)$$

We have constructed this in such a way that the real and imaginary parts of $\mathbf{E}_0 = \mathbf{E}_0^{\Re} + i\mathbf{E}_0^{\Im}$ correspond to the two different quadratures of the oscillating total field,

oscillating $\pi/2$ out of phase with respect to each other.¹⁰ $\mathbf{E}_0$ itself is not oscillating but rather is in a rotating frame with frequency ω_k .

We can express the dipole operator (in the Schrödinger picture) in a similar manner,

$$\begin{aligned}\langle \mathbf{d} \rangle &= \text{Tr}(\rho \mathbf{d}) = \mathbf{d}_{ge} \rho_{eg} + \mathbf{d}_{eg} \rho_{ge} \\ &= (\mathbf{d}_{ge} \rho_{eg} e^{i\omega_k t}) e^{-i\omega_k t} + (\mathbf{d}_{eg} \rho_{ge} e^{-i\omega_k t}) e^{i\omega_k t} \\ &\equiv \mathbf{d} e^{-i\omega_k t} + \mathbf{d}^* e^{i\omega_k t} \\ &= 2 \mathbf{d}^{\Re} \cos \omega_k t + 2 \mathbf{d}^{\Im} \sin \omega_k t,\end{aligned}\tag{2.4.2}$$

where $\mathbf{d} \equiv \mathbf{d}_{ge} \rho_{eg} e^{i\omega_k t}$. As with the electric field, this is therefore a way of representing a real dipole moment $\langle \mathbf{d} \rangle$, which oscillates with the same frequency as the driving field ω_k , as a complex vector $\mathbf{d}$ whose real and imaginary parts correspond to the two different quadratures of oscillation. For example, if $\mathbf{E}_0^{\Im} = 0$, i.e. $\mathcal{E}_0$ oscillates with $\cos \omega_k t$, then the real and imaginary parts of $\mathbf{d}$ correspond to the parts of $\langle \mathbf{d} \rangle$ oscillating in phase and out of phase respectively with the driving field.

Using the optical Bloch equations (2.3.29), the equation of motion for the dipole moment $\mathbf{d}$ is

$$\begin{aligned}\frac{d}{dt} \mathbf{d} &= \frac{d}{dt} (\mathbf{d}_{ge} \rho_{eg} e^{i\omega_k t}) \\ &= i\omega_k (\mathbf{d}_{ge} \rho_{eg} e^{i\omega_k t}) + \mathbf{d}_{ge} \frac{d\rho_{eg}}{dt} e^{i\omega_k t} \\ &= i\omega_k \mathbf{d} - \left(i\omega_0 + \frac{\Gamma_0}{2} \right) \mathbf{d} - \mathbf{d}_{ge} \frac{i}{\hbar} (\rho_{ee} - \rho_{gg}) \mathbf{d}_{eg} \cdot \mathbf{E}_0 \\ &= \left(i\Delta - \frac{\Gamma_0}{2} \right) \mathbf{d} - i \frac{|\mathbf{d}_{ge}|^2}{\hbar} (\rho_{ee} - \rho_{gg}) (\hat{\mathbf{d}}_{ge} \cdot \hat{\mathbf{e}}_0) E_0 \hat{\mathbf{d}}_{ge},\end{aligned}\tag{2.4.3}$$

where $\Delta = \omega_k - \omega_0$ is the detuning of the driving field from the atomic transition frequency, and $\hat{\mathbf{d}}_{eg}$ and $\hat{\mathbf{e}}_0$ are the polarisation unit vectors for the dipole ($\mathbf{d}_{eg} = d_{eg} \hat{\mathbf{d}}_{eg}$) and field [$\mathbf{E}_0(\mathbf{r}, t) = E_0(\mathbf{r}, t) \hat{\mathbf{e}}_0$] respectively. Assuming that $\hat{\mathbf{d}}_{ge} = \hat{\mathbf{e}}_0$, then $\dot{\mathbf{d}}$ becomes

$$\dot{\mathbf{d}} = \left(i\Delta - \frac{\Gamma_0}{2} \right) \mathbf{d} - i \frac{|\mathbf{d}_{ge}|^2}{\hbar} (\rho_{ee} - \rho_{gg}) \mathbf{E}_0,\tag{2.4.4}$$

where the equation of motion for $\dot{\rho}_{ee}$ is

¹⁰ $\sin(\omega t) = \cos(\omega t - \pi/2)$.

$$\dot{\rho}_{ee} = -\rho_{ee}\Gamma_0 - 2\frac{i}{\hbar}\text{Im}(\mathbf{d} \cdot \mathbf{E}_0^*), \quad (2.4.5)$$

and $\rho_{gg}(t) = 1 - \rho_{ee}(t)$.

2.4.2 Atomic Polarisability and the Weak-Driving Steady State

If we assume weak driving (see Sect. 2.4.3), then we can set $(\rho_{ee} - \rho_{gg}) \simeq -1$, resulting in just one equation of motion,

$$\dot{\mathbf{d}} \simeq \left(i\Delta - \frac{\Gamma_0}{2}\right)\mathbf{d} + i\frac{|\mathbf{d}_{ge}|^2}{\hbar}\mathbf{E}_0. \quad (2.4.6)$$

This has a linear steady-state solution

$$\mathbf{d} = \alpha \mathbf{E}_0, \quad (2.4.7)$$

where we have introduced the atomic polarisability,

$$\alpha = -\alpha_0 \frac{\gamma_0}{\Delta + i\gamma_0}, \quad (2.4.8)$$

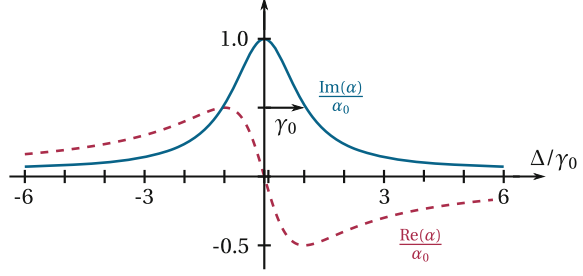
where [using Eq. (2.3.11)]

$$\alpha_0 \equiv \frac{|\mathbf{d}_{ge}|^2}{\hbar} = \frac{6\pi\epsilon_0}{k_0^3}, \quad \text{and} \quad \gamma_0 = \frac{\Gamma_0}{2} = \frac{|\mathbf{d}_{eg}|^2 k_0^3}{6\pi\epsilon_0\hbar}. \quad (2.4.9)$$

This complex polarisability α describes the resonance behaviour of the atomic dipole to the driving field. In Fig. 2.2 we plot the real and imaginary parts of α ,

$$\frac{\alpha}{\alpha_0} = -\frac{\gamma_0\Delta}{\Delta^2 + \gamma_0^2} + i\frac{\gamma_0^2}{\Delta^2 + \gamma_0^2}. \quad (2.4.10)$$

Fig. 2.2 Real (red dashed) and imaginary (blue solid) parts of the single-atom polarisability, α (2.4.10), normalised by α_0 . The blue solid line is a Lorentzian with halfwidth γ_0



The imaginary part (blue line) is a Lorentzian lineshape with halfwidth-at-half-maximum (HWHM) γ_0 and centered on $\Delta = 0$ (i.e. it is a maximum when the driving frequency ω_k is resonant with the natural frequency of the atomic transition ω_0).

2.4.3 Transient and Steady-State Solutions

Let us now consider how quickly the system settles into the steady-state solution. Substituting α back into the equations of motion (2.4.4) and (2.4.5), we get

$$\dot{d} = \left(i\Delta - \frac{\Gamma_0}{2} \right) d - i(\rho_{ee} - \rho_{gg}) \frac{\Gamma_0}{2} \alpha_0 E_0, \quad (2.4.11a)$$

$$\dot{\rho}_{ee} = -\Gamma_0 \rho_{ee} - \Gamma_0 \operatorname{Im} \left(\frac{d^* \alpha_0 E_0}{|\mathbf{d}_{ge}|^2} \right), \quad (2.4.11b)$$

where $\mathbf{d} = d\hat{\mathbf{e}}_0$. Looking at these two equations, the characteristic time scale is the natural decay rate Γ_0 , the characteristic dipole moment magnitude is $|\mathbf{d}_{ge}|$, and the characteristic electric field magnitude is $|\mathbf{d}_{ge}|/\alpha_0$. We can therefore define new dimensionless variables,

$$\tilde{t} \equiv \Gamma_0 t = \frac{t}{\tau_0}, \quad \tilde{\Delta} \equiv \frac{2\Delta}{\Gamma_0}, \quad \tilde{d} \equiv \frac{d}{|\mathbf{d}_{ge}|}, \quad \tilde{E}_0 \equiv \frac{\alpha_0 E_0}{|\mathbf{d}_{ge}|}, \quad (2.4.12)$$

resulting in the dimensionless equations of motion

$$\frac{d}{d\tilde{t}} \tilde{d} = \left(i\tilde{\Delta} - 1 \right) \frac{\tilde{d}}{2} - i \left(\rho_{ee} - \frac{1}{2} \right) \tilde{E}_0, \quad (2.4.13a)$$

$$\frac{d}{d\tilde{t}} \rho_{ee} = -\rho_{ee} - \operatorname{Im} \left(\tilde{d}^* \tilde{E}_0 \right). \quad (2.4.13b)$$

Note also that the normalised dipole-moment is equivalent to ρ_{eg} in the rotating frame, $\tilde{d} = \bar{\rho}_{eg} e^{i\omega_k t}$.

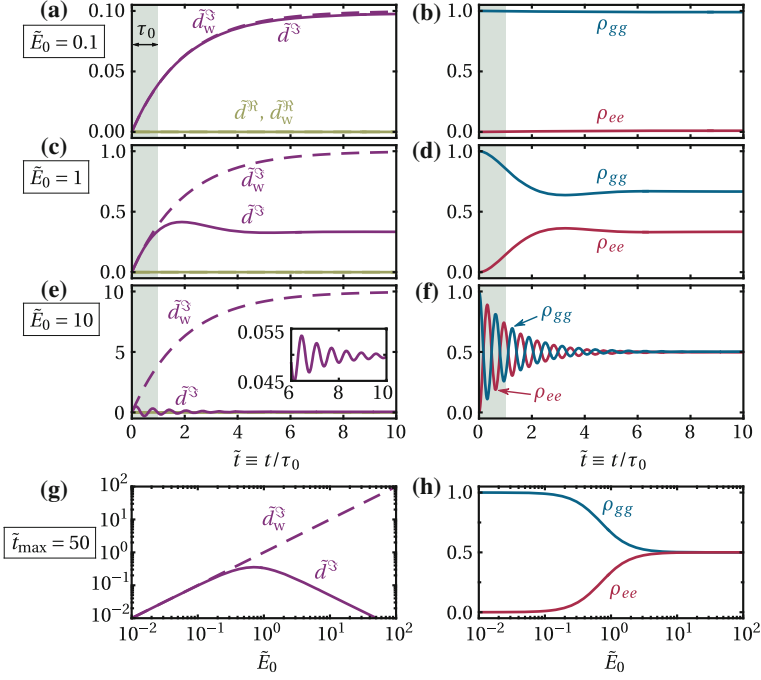


Fig. 2.3 Solutions to the single-atom two-level optical Bloch equations with dimensionless parameters (2.4.13). **a, c, e** The time evolution of the real (*olive*) and imaginary (*purple*) dipole moments $\tilde{d} \equiv d/|\mathbf{d}_{ge}|$. The *dashed lines* are the solutions in the weak-driving approximation (setting $\rho_{gg} = 1$). **b, d, f** The evolution of the ground (*blue*) and excited (*red*) state populations. The driving fields $\tilde{E}_0 \equiv E_0\alpha_0/|\mathbf{d}_{ge}|$ are 0.1 (a,b), 1 (c,d) and 10 (e,f) and are resonant with the atomic frequency ($\Delta = 0$). The steady-state solutions for the dipole moments and populations after $\tilde{t} = 50$ are plotted in **(g)** and **(h)** respectively, as a function of driving field strength. For $\tilde{E}_0 \ll 1$, the weak-driving model is a good approximation for the full behaviour. For $\tilde{E}_0 \gtrsim 1$ however, the full behaviour deviates from the weak-driving approximation

In Fig. 2.3a–f we plot the time evolution of $\tilde{d}$ and ρ_{gg} for different driving field strengths, $\tilde{E}_0 = 0.1, 1, 10$. For all three driving strengths, after around 10 natural lifetimes ($\tilde{t} \equiv t/\tau_0 \simeq 10$), the evolution has settled into a steady state. This steady-state solution is plotted in Fig. 2.3g,h as a function of driving strength. For weak driving ($\tilde{E}_0 \ll 1$), the full solution can be well approximated by assuming that $\rho_{gg} = 1$, as we did for Eq. (2.4.7). Once the driving strength increases past $\tilde{E}_0 \simeq 1$ however, this is no longer a good approximation as there is now appreciable population in the excited state. For strong driving, the linear response of $\tilde{d}$ to $\tilde{E}_0$ breaks down and the nonlinearities in (2.4.13) significantly attenuate the dipole moment.

In most of this Thesis, we will use the weak-driving approximation as this allows us to calculate a linear steady-state solution with minimal computational effort. What we have seen in this Section is that this approximation is valid provided that

$$E_{0,\text{weak}} \ll \frac{|\mathbf{d}_{ge}|}{\alpha}. \quad (2.4.14)$$

2.5 Classical Dipole—Simple Harmonic Oscillator

In Sect. 2.3.5, we showed that when the field is in a coherent state, we can treat it as a classical electric field. In this Section, we will show (as has already been shown in, e.g., [9, 10]) that the behaviour of the weakly-driven electric dipole in Sect. 2.4.2 is identical to that of a damped driven simple harmonic oscillator.

As before, let us assume that the driving field has the form

$$\mathcal{E}_0(\mathbf{r}, t) = \mathbf{E}_0(\mathbf{r}) e^{-i\omega t} + \mathbf{E}_0^*(\mathbf{r}) e^{i\omega t} = 2\mathbf{E}_0^{\Re}(\mathbf{r}) \cos \omega t + 2\mathbf{E}_0^{\Im}(\mathbf{r}) \sin \omega t. \quad (2.5.1)$$

We are assuming that the only time dependence in $\mathcal{E}_0$ is the rotation with frequency ω . The equation of motion for the displacement $\mathbf{s}(t) = s(t)\hat{\mathbf{e}}_0$ of an electron in this external electric field is

$$\frac{d^2 s^2}{dt^2} + \Gamma \frac{ds}{dt} + \omega_0^2 s = \frac{2e}{m} [E_0^{\Re}(\mathbf{r}_0) \cos \omega t + E_0^{\Im}(\mathbf{r}_0) \sin \omega t], \quad (2.5.2)$$

where motion is damped by Γ , the natural frequency of oscillation is ω_0 , e is the electron charge and m is the electron mass. Again we assume the *dipole approximation*, i.e. the maximum electron displacement is very much smaller than the field wavelength.

2.5.1 Steady-State Solution

Let us first look for a solution of the form

$$s(t) = \bar{s} e^{-i\omega t} + \bar{s}^* e^{i\omega t} = 2\bar{s}^{\Re} \cos \omega t + 2\bar{s}^{\Im} \sin \omega t. \quad (2.5.3)$$

By substituting (2.5.3) into (2.5.2) and comparing coefficients of $\cos$ and $\sin$, we arrive at the solutions (see Appendix B.1)

$$\bar{s}^{\Re} = \frac{e}{m} \frac{(\omega_0^2 - \omega^2) E_0^{\Re} - \omega \Gamma E_0^{\Im}}{(\omega_0^2 - \omega^2)^2 + \omega^2 \Gamma^2}, \quad (2.5.4a)$$

$$\bar{s}^{\Im} = \frac{e}{m} \frac{(\omega_0^2 - \omega^2) E_0^{\Im} + \omega \Gamma E_0^{\Re}}{(\omega_0^2 - \omega^2)^2 + \omega^2 \Gamma^2}, \quad (2.5.4b)$$

$$\Rightarrow \quad \bar{s} = \bar{s}^{\Re} + i \bar{s}^{\Im} = \frac{e}{m} \frac{(\omega_0^2 - \omega^2) + i \omega \Gamma}{(\omega_0^2 - \omega^2)^2 + \omega^2 \Gamma^2} E_0. \quad (2.5.4c)$$

Again assuming that $|\omega_0 - \omega| \ll (\omega_0 + \omega)$ (*rotating wave approximation*), we can approximate

$$(\omega_0^2 - \omega^2) = (\omega_0 + \omega)(\omega_0 - \omega) \simeq 2\omega_0(\omega_0 - \omega), \quad (2.5.5)$$

resulting in a steady-state dipole moment (in the rotating frame) $d = e\bar{s}$,

$$\begin{aligned} d &\simeq \frac{e^2}{m} \frac{2\omega_0(\omega_0 - \omega) + i\omega_0\Gamma}{4\omega_0^2(\omega_0 - \omega)^2 + \omega_0^2\Gamma^2} E_0 \\ &= \frac{e^2}{2m\omega_0} \frac{-\Delta + i(\Gamma/2)}{\Delta^2 + (\Gamma/2)^2} E_0 \\ &= -\alpha_0 \frac{\gamma}{\Delta^2 + \gamma^2} E_0. \end{aligned} \quad (2.5.6)$$

This has the same form as Eq. (2.4.7) where the steady-state solution to the dipole moment is $d = \alpha E_0$. Comparing the two, we can see that the dissipation rate Γ is equivalent to the natural decay rate of the atomic excited state Γ_0 (2.4.9),

$$\Gamma \equiv 2\gamma = \frac{\mathcal{D}_0^2 k_0^3}{3\pi \varepsilon_0 \hbar}, \quad \alpha_0 = \frac{6\pi \varepsilon_0}{k_0^3}, \quad (2.5.7)$$

where we have chosen the characteristic dipole element $\mathcal{D}_0 = ea_0$ with characteristic length $a_0 = \sqrt{\hbar/m\omega_0}$.

2.5.2 Transient Solution

In addition to this steady-state solution, Eq. (2.5.2) also has a transient solution (Appendix B.1),

$$s_t(t) = s(0) e^{-\gamma t} \cos\left(\sqrt{\omega_0^2 - \gamma^2} t\right), \quad (2.5.8)$$

where $s(0)$ is the initial state of $s(t)$ (if we assume the driving field is switched on at $t = 0$). The amplitude of these oscillations decays away with lifetime γ , after which the steady-state solution (2.5.4c) dominates.

Chapter Summary

- The interaction between a 2-level atom and an EM field can be described using the master equation
- A weak EM field in a coherent state, such as from a laser, can be treated as an oscillating classical EM field
- For a weak driving field, the atomic electric dipole has a steady-state solution
- This steady-state solution can also be derived from the behaviour of a classical driven damped simple harmonic oscillator

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Bettles, R.

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