CHAPTER 5

Atom–field interaction – semiclassical theory

One of the simplest nontrivial problems involving the atom–field interaction is the coupling of a two-level atom with a single mode of the electromagnetic field. A two-level atom description is valid if the two atomic levels involved are resonant or nearly resonant with the driving field, while all other levels are highly detuned. Under certain realistic approximations, it is possible to reduce this problem to a form which can be solved exactly; allowing essential features of the atom–field interaction to be extracted.

In this chapter we present a semiclassical theory of the interaction of a single two-level atom with a single mode of the field in which the atom is treated as a quantum two-level system and the field is treated classically. A fully quantum mechanical theory will be presented in Chapter 6.

A two-level atom is formally analogous to a spin-1/2 system with two possible states. In the dipole approximation, when the field wavelength is larger than the atomic size, the atom–field interaction problem is mathematically equivalent to a spin-1/2 particle interacting with a time-dependent magnetic field. Just as the spin-1/2 system undergoes the so-called Rabi oscillations between the spin-up and spin-down states under the action of an oscillating magnetic field, the two-level atom also undergoes *optical* Rabi oscillations under the action of the driving electromagnetic field. These oscillations are damped if the atomic levels decay. An understanding of this simple model of the atom–field interaction enables us to consider more complicated problems involving an ensemble of atoms interacting with the field. Perhaps the most important

example of such a problem is the laser, which we discuss later in this chapter.*

5.1 Atom–field interaction Hamiltonian

An electron of charge e and mass m interacting with an external electromagnetic field is described by a minimal-coupling Hamiltonian

$$\mathscr{H} = \frac{1}{2m}[\mathbf{p} - e\mathbf{A}(\mathbf{r},t)]^2 + eU(\mathbf{r},t) + V(r), \tag{5.1.1}$$

where $\mathbf{p}$ is the canonical momentum operator, $\mathbf{A}(\mathbf{r},t)$ and $U(\mathbf{r},t)$ are the vector and scalar potentials of the external field, respectively, and $V(r)$ is an electrostatic potential that is normally the atomic binding potential. In this section, we first derive this Hamiltonian from a gauge invariance point of view, before reducing it to a simple form suitable for describing the interaction of a two-level atom with the radiation field.

5.1.1 *Local gauge (phase) invariance and minimal-coupling Hamiltonian*

The motion of a free electron is described by the Schrödinger equation

$$\frac{-\hbar^2}{2m}\nabla^2\psi = i\hbar\frac{\partial\psi}{\partial t}, \tag{5.1.2}$$

such that

$$P(\mathbf{r},t) = |\psi(\mathbf{r},t)|^2 \tag{5.1.3}$$

gives the probability density of finding an electron at position $\mathbf{r}$ and time t. In Eq. (5.1.2), if $\psi(\mathbf{r},t)$ is a solution so is $\psi_1(\mathbf{r},t) = \psi(\mathbf{r},t)\exp(i\chi)$ where χ is an arbitrary constant phase. The probability density $P(\mathbf{r},t)$ would also remain unaffected by an arbitrary choice of χ. Thus the choice of the phase of the wave function $\psi(\mathbf{r},t)$ is completely arbitrary, and two functions differing only by a constant phase factor represent the same physical state.

The situation is different, however, if the phase is allowed to vary *locally*, i.e. to be a function of space and time variables, i.e.,

$$\psi(\mathbf{r},t) \rightarrow \psi(\mathbf{r},t)e^{i\chi(\mathbf{r},t)}. \tag{5.1.4}$$

* The semiclassical theory of laser behavior as developed by the schools of Lamb and Haken (see Lamb [1963,1964] and Haken [1964]) are the pioneering treatments of the problem. Lamb begins from the coupled Maxwell–Schrödinger equations, while Haken and co-workers take a semiclassical (factorized) limit of quantum fields.

The probability $P(\mathbf{r},t)$ remains unaffected by this transformation, but the Schrödinger equation (5.1.2) is no longer satisfied. If we want to satisfy *local* gauge (phase) invariance, then the Schrödinger equation must be modified by adding new terms to Eq. (5.1.2)

$$\left\{-\frac{\hbar^2}{2m}\left[\nabla - i\frac{e}{\hbar}\mathbf{A}(\mathbf{r},t)\right]^2 + eU(\mathbf{r},t)\right\}\psi = i\hbar\frac{\partial\psi}{\partial t}, \tag{5.1.5}$$

where $\mathbf{A}(\mathbf{r},t)$ and $U(\mathbf{r},t)$ are functions which must be inserted into (5.1.2) if we want to be able to make the transformation (5.1.4), and are given by

$$\mathbf{A}(\mathbf{r},t) \to \mathbf{A}(\mathbf{r},t) + \frac{\hbar}{e}\nabla\chi(\mathbf{r},t), \tag{5.1.6}$$

$$U(\mathbf{r},t) \to U(\mathbf{r},t) - \frac{\hbar}{e}\frac{\partial\chi}{\partial t}(\mathbf{r},t). \tag{5.1.7}$$

The functions $\mathbf{A}(\mathbf{r},t)$ and $U(\mathbf{r},t)$ are identified as the vector and scalar potentials of the electromagnetic field, respectively. These are the gauge-dependent potentials. The gauge-independent quantities are the electric and magnetic fields

$$\mathbf{E} = -\nabla U - \frac{\partial\mathbf{A}}{\partial t}, \tag{5.1.8}$$

$$\mathbf{B} = \nabla\times\mathbf{A}. \tag{5.1.9}$$

Equation (5.1.5), which is the logical extension of Eq. (5.1.2) due to the requirement of local gauge (phase) invariance, has the form

$$\mathscr{H}\psi = i\hbar\partial\psi/\partial t, \tag{5.1.10}$$

with $\mathscr{H}$ being the minimal-coupling Hamiltonian (recall $\mathbf{p} = -i\hbar\nabla$) described in Eq. (5.1.1). The Schrödinger equation (5.1.5) represents the interaction of an electron with a given electromagnetic field. The electrons are described by the wave function $\psi(\mathbf{r},t)$ whereas the field is described by the vector and scalar potentials $\mathbf{A}$ and U, respectively.

It is interesting to note that the Hamiltonian (5.1.1) has been 'derived' from a gauge invariance argument and is expressed in terms of the gauge-dependent quantities $\mathbf{A}(\mathbf{r},t)$ and $U(\mathbf{r},t)$. The vector and scalar potentials have therefore a larger physical significance than is usually attributed to them. They are not merely introduced for the sake of mathematical simplicity in problems dealing with 'observable' electric and magnetic fields. Instead, they arise naturally in any gauge (phase) invariance argument as shown above.

We also note that the Schrödinger equation plus the concept of local gauge invariance has led us to the introduction of the electromagnetic field. In this way, we can and do argue that the 'photon' (in our

derivation, the classical field limit of the same) has been 'derived' from the Schrödinger equation plus the local gauge invariance arguments.

We have here a taste of one of the most fundamental concepts in modern physics, namely, that of the gauge field theory. Gauge theory, in the hands of Steven Weinberg and Abdus Salam, led to the unification of the weak and the electromagnetic interactions.

5.1.2 Dipole approximation and $\mathbf{r}\cdot\mathbf{E}$ Hamiltonian

We now examine the problem of an electron bound by a potential $V(r)$ to a force center (nucleus) located at $\mathbf{r}_0$. The minimal-coupling Hamiltonian (5.1.1) for an interaction between an atom and the radiation field can be reduced to a simple form by using the dipole approximation. The entire atom is immersed in a plane electromagnetic wave described by a vector potential $\mathbf{A}(\mathbf{r}_0+\mathbf{r},t)$. This vector potential may be written in the dipole approximation, $\mathbf{k}\cdot\mathbf{r} \ll 1$, as

$$\begin{aligned}\mathbf{A}(\mathbf{r}_0+\mathbf{r},t) &= \mathbf{A}(t)\exp[i\mathbf{k}\cdot(\mathbf{r}_0+\mathbf{r})]\\ &= \mathbf{A}(t)\exp(i\mathbf{k}\cdot\mathbf{r}_0)(1+i\mathbf{k}\cdot\mathbf{r}+\ldots)\\ &\simeq \mathbf{A}(t)\exp(i\mathbf{k}\cdot\mathbf{r}_0).\end{aligned} \tag{5.1.11}$$

The Schrödinger equation for this problem (in the dipole approximation) is given by Eq. (5.1.5) with $\mathbf{A}(\mathbf{r},t) \equiv \mathbf{A}(\mathbf{r}_0,t)$, i.e.,

$$\left\{-\frac{\hbar^2}{2m}\left[\nabla - \frac{ie}{\hbar}\mathbf{A}(\mathbf{r}_0,t)\right]^2 + V(r)\right\}\psi(\mathbf{r},t) = i\hbar\frac{\partial\psi(\mathbf{r},t)}{\partial t}, \tag{5.1.12}$$

where we have added a binding potential $V(r)$. We note that in Eq. (5.1.12), and elsewhere in this book, we are working in the radiation gauge, in which

$$U(\mathbf{r},t) = 0, \tag{5.1.13}$$

and

$$\nabla\cdot\mathbf{A} = 0. \tag{5.1.14}$$

We have added the term $V(r)$ in the Hamiltonian which arises from the electrostatic potential that binds the electron to the nucleus.

We proceed to simplify Eq. (5.1.12) by defining a new wave function $\phi(\mathbf{r},t)$ as

$$\psi(\mathbf{r},t) = \exp\left[\frac{ie}{\hbar}\mathbf{A}(\mathbf{r}_0,t)\cdot\mathbf{r}\right]\phi(\mathbf{r},t). \tag{5.1.15}$$

Inserting Eq. (5.1.15) into Eq. (5.1.12), we find

$$i\hbar\left[\frac{ie}{\hbar}\dot{\mathbf{A}}\cdot\mathbf{r}\phi(\mathbf{r},t)+\dot{\phi}(\mathbf{r},t)\right]\exp\left(\frac{ie}{\hbar}\mathbf{A}\cdot\mathbf{r}\right)$$
$$=\exp\left(\frac{ie}{\hbar}\mathbf{A}\cdot\mathbf{r}\right)\left[\frac{p^2}{2m}+V(r)\right]\phi(\mathbf{r},t). \tag{5.1.16}$$

This equation, after the cancellation of the exponential factor and some rearrangement, takes the simple form

$$i\hbar\dot{\phi}(\mathbf{r},t)=[\mathscr{H}_0-e\mathbf{r}\cdot\mathbf{E}(\mathbf{r}_0,t)]\phi(\mathbf{r},t), \tag{5.1.17}$$

where

$$\mathscr{H}_0=\frac{p^2}{2m}+V(r), \tag{5.1.18}$$

is the unperturbed Hamiltonian of the electron and we use $\mathbf{E}=-\dot{\mathbf{A}}$. Notice that the total Hamiltonian

$$\mathscr{H}=\mathscr{H}_0+\mathscr{H}_1 \tag{5.1.19}$$

with

$$\mathscr{H}_1=-e\mathbf{r}\cdot\mathbf{E}(\mathbf{r}_0,t), \tag{5.1.20}$$

is given in terms of the gauge-independent field $\mathbf{E}$. We shall use this Hamiltonian in our subsequent studies of atom–field interaction. Note also that this Hamiltonian has been obtained from the radiation gauge Hamiltonian (5.1.12) by applying the gauge transformation $\chi(\mathbf{r},t)=-e\mathbf{A}(\mathbf{r}_0,t)\cdot\mathbf{r}/\hbar$.

5.1.3 $\mathbf{p}\cdot\mathbf{A}$ *Hamiltonian*

In many textbooks one finds the atom–field Hamiltonian expressed in terms of the canonical momentum $\mathbf{p}$ and the vector potential $\mathbf{A}$ instead of the simple gauge invariant expression (5.1.17). This has resulted in considerable confusion, and we therefore consider the problem in some detail. We again choose a radiation gauge in which $U(\mathbf{r},t)=0$ and $\nabla\cdot\mathbf{A}=0$. The condition $\nabla\cdot\mathbf{A}=0$ implies, in quantum mechanics, that $[\mathbf{p},\mathbf{A}]=0$. The total Hamiltonian (5.1.1) can, therefore, be written as

$$\mathscr{H}'=\mathscr{H}_0+\mathscr{H}_2, \tag{5.1.21}$$

where $\mathscr{H}_0$ is given by Eq. (5.1.18) and, in the dipole approximation (5.1.11),

$$\mathscr{H}_2=-\frac{e}{m}\mathbf{p}\cdot\mathbf{A}(\mathbf{r}_0,t)+\frac{e^2}{2m}A^2(\mathbf{r}_0,t), \tag{5.1.22}$$

and the Schrödinger equation reads

$$\left[\mathscr{H}_0-\frac{e}{m}\mathbf{p}\cdot\mathbf{A}(\mathbf{r}_0,t)+\frac{e^2}{2m}A^2(\mathbf{r}_0,t)\right]\psi(\mathbf{r},t)=i\hbar\frac{\partial}{\partial t}\psi(\mathbf{r},t). \tag{5.1.23}$$

The A^2 term in Eq. (5.1.23) is ususally small and can be ignored. The wave function $\psi(\mathbf{r},t)$ then obeys the equation of motion

$$i\hbar\frac{\partial}{\partial t}\psi(\mathbf{r},t)=\left[\mathscr{H}_0-\frac{e}{m}\mathbf{p}\cdot\mathbf{A}(\mathbf{r}_0,t)\right]\psi(\mathbf{r},t), \tag{5.1.24}$$

corresponding to a Hamiltonian

$$\mathscr{H}'=\mathscr{H}_0-\frac{e}{m}\mathbf{p}\cdot\mathbf{A}(\mathbf{r}_0,t), \tag{5.1.25}$$

and

$$\mathscr{H}_2=-\frac{e}{m}\mathbf{p}\cdot\mathbf{A}(\mathbf{r}_0,t). \tag{5.1.26}$$

The two different Hamiltonians $\mathscr{H}_1$ and $\mathscr{H}_2$ given in Eqs. (5.1.20) and (5.1.26), respectively, seem to give different physical results since the matrix elements of these Hamiltonians, calculated between the eigenstates of the *unperturbed* Hamiltonian $\mathscr{H}_0$, given by Eq. (5.1.18), are not the same. In order to show this explicitly, we consider a linearly polarized monochromatic plane-wave field interacting with an atom placed at $\mathbf{r}_0=0$. The electric field then takes the form

$$\mathbf{E}(0,t)=\mathscr{E}\cos\nu t, \tag{5.1.27}$$

and the corresponding vector potential in the radiation gauge is

$$\mathbf{A}(0,t)=-\frac{1}{\nu}\mathscr{E}\sin\nu t. \tag{5.1.28}$$

Consider now the time-independent amplitudes associated with $\mathscr{H}_1$ and $\mathscr{H}_2$,

$$W_1=-e\mathbf{r}\cdot\mathscr{E}, \tag{5.1.29a}$$
$$W_2=\frac{e}{m\nu}\mathbf{p}\cdot\mathscr{E}. \tag{5.1.29b}$$

We may relate W_1 and W_2 by noting that

$$\mathbf{p}=m\mathbf{v}=-m\left(\frac{i}{\hbar}\right)[\mathbf{r},\mathscr{H}_0]. \tag{5.1.30}$$

We then find for the matrix elements of W_1 and W_2, calculated between an initial eigenstate $|i\rangle$ of $\mathscr{H}_0$ (with $\mathscr{H}_0|i\rangle=\hbar\omega_i|i\rangle$) and a

final eigenstate $|f\rangle$ (with $\mathscr{H}_0|f\rangle = \hbar\omega_f|f\rangle$), the ratio

$$\left|\frac{\langle f|W_2|i\rangle}{\langle f|W_1|i\rangle}\right| = \left|-\frac{(e/mv)\langle f|\mathbf{p}|i\rangle\cdot\mathscr{E}}{e\langle f|\mathbf{r}|i\rangle\cdot\mathscr{E}}\right| = \frac{\omega}{v}, \tag{5.1.31}$$

where $\omega = \omega_f - \omega_i$ is the transition frequency. Hence, the matrix elements of the two interaction Hamiltonians $\mathscr{H}_1$ and $\mathscr{H}_2$ differ by the ratio of the transition frequency over the field frequency. As was first pointed out by Lamb, this makes a difference in measurable quantities like transition rates. We present a resolution of this in Appendix 5.A.

5.2 Interaction of a single two-level atom with a single-mode field

5.2.1 Probability amplitude method

Consider the interaction of a single-mode radiation field of frequency v with a two-level atom (Fig. 5.1). Let $|a\rangle$ and $|b\rangle$ represent the upper and lower level states of the atom, i.e., they are eigenstates of the unperturbed part of the Hamiltonian $\mathscr{H}_0$ with the eigenvalues $\hbar\omega_a$ and $\hbar\omega_b$, respectively. The wave function of a two-level atom can be written in the form

$$|\psi(t)\rangle = C_a(t)|a\rangle + C_b(t)|b\rangle, \tag{5.2.1}$$

where C_a and C_b are the probability amplitudes of finding the atom in states $|a\rangle$ and $|b\rangle$, respectively. The corresponding Schrödinger equation is

$$|\dot{\psi}(t)\rangle = -\frac{i}{\hbar}\mathscr{H}|\psi(t)\rangle, \tag{5.2.2}$$

with

$$\mathscr{H} = \mathscr{H}_0 + \mathscr{H}_1, \tag{5.2.3}$$

where $\mathscr{H}_0$ and $\mathscr{H}_1$ represent the unperturbed and interaction parts of the Hamiltonian, respectively. By using the completeness relation $|a\rangle\langle a| + |b\rangle\langle b| = \mathbf{1}$, we can write $\mathscr{H}_0$ as

$$\begin{aligned}\mathscr{H}_0 &= (|a\rangle\langle a| + |b\rangle\langle b|)\mathscr{H}_0(|a\rangle\langle a| + |b\rangle\langle b|)\\ &= \hbar\omega_a|a\rangle\langle a| + \hbar\omega_b|b\rangle\langle b|,\end{aligned} \tag{5.2.4}$$

where we use $\mathscr{H}_0|a\rangle = \hbar\omega_a|a\rangle$ and $\mathscr{H}_0|b\rangle = \hbar\omega_b|b\rangle$. Similarly, the part of the Hamiltonian $\mathscr{H}_1$ that represents the interaction of the atom

with the radiation field can be written as

$$\begin{aligned}\mathscr{H}_1 &= -exE(t)\\ &= -e(|a\rangle\langle a| + |b\rangle\langle b|)x(|a\rangle\langle a| + |b\rangle\langle b|)E(z,t)\\ &= -(\wp_{ab}|a\rangle\langle b| + \wp_{ba}|b\rangle\langle a|)E(t),\end{aligned} \tag{5.2.5}$$

where $\wp_{ab} = \wp_{ba}^* = e\langle a|x|b\rangle$ is the matrix element of the electric dipole moment and $E(t)$ is the field at the atom. Here, we assume that the electric field is linearly polarized along the x-axis. In the dipole approximation, the field can be expressed as

$$E(t) = \mathscr{E}\cos vt, \tag{5.2.6}$$

where $\mathscr{E}$ is the amplitude and $v = ck$ is the frequency of the field. The equations of motion for the amplitudes C_a and C_b may be written as

$$\dot{C}_a = -i\omega_a C_a + i\Omega_R e^{-i\phi}\cos(vt)C_b, \tag{5.2.7}$$

$$\dot{C}_b = -i\omega_b C_b + i\Omega_R e^{i\phi}\cos(vt)C_a, \tag{5.2.8}$$

where the Rabi frequency Ω_R is defined as

$$\Omega_R = \frac{|\wp_{ba}|\mathscr{E}}{\hbar}, \tag{5.2.9}$$

and ϕ is the phase of the dipole matrix element $\wp_{ba} = |\wp_{ba}|\exp(i\phi)$. In order to solve for C_a and C_b, we first write the equations of motion for the slowly varying amplitudes:

$$c_a = C_a e^{i\omega_a t}, \tag{5.2.10}$$

$$c_b = C_b e^{i\omega_b t}. \tag{5.2.11}$$

It then follows from Eqs. (5.2.7) and (5.2.8) that

$$\dot{c}_a = i\frac{\Omega_R}{2}e^{-i\phi}c_b e^{i(\omega - v)t}, \tag{5.2.12}$$

$$\dot{c}_b = i\frac{\Omega_R}{2}e^{i\phi}c_a e^{-i(\omega - v)t}, \tag{5.2.13}$$

where $\omega = \omega_a - \omega_b$ is the atomic transition frequency. In deriving Eqs. (5.2.12) and (5.2.13), we have ignored counter-rotating terms proportional to $\exp[\pm i(\omega + v)t]$ on the right-hand side in the *rotating-wave approximation*. This is generally a very good approximation. Furthermore, in some cases the counter-rotating terms never appear (as seen later in section 5.2.3) and Eqs. (5.2.12) and (5.2.13) are exact.

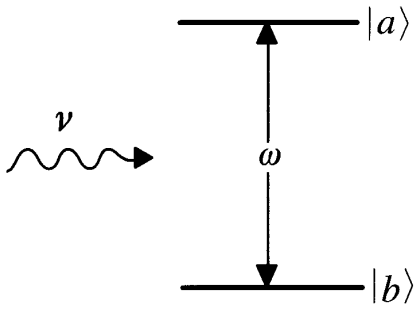


Fig. 5.1
Interaction of a two-level atom with a single-mode field.

The solutions for c_a and c_b can be written as

$$c_a(t) = \left(a_1 e^{i\Omega t/2} + a_2 e^{-i\Omega t/2}\right) e^{i\Delta t/2}, \tag{5.2.14}$$

$$c_b(t) = \left(b_1 e^{i\Omega t/2} + b_2 e^{-i\Omega t/2}\right) e^{-i\Delta t/2}, \tag{5.2.15}$$

where $\Delta = \omega - \nu$,

$$\Omega = \sqrt{\Omega_R^2 + (\omega - \nu)^2}, \tag{5.2.16}$$

and a_1, a_2, b_1, and b_2 are constants of integration which are determined from the initial conditions:

$$a_1 = \frac{1}{2\Omega}\left[(\Omega - \Delta)c_a(0) + \Omega_R e^{-i\phi} c_b(0)\right], \tag{5.2.17}$$

$$a_2 = \frac{1}{2\Omega}\left[(\Omega + \Delta)c_a(0) - \Omega_R e^{-i\phi} c_b(0)\right], \tag{5.2.18}$$

$$b_1 = \frac{1}{2\Omega}\left[(\Omega + \Delta)c_b(0) + \Omega_R e^{i\phi} c_a(0)\right], \tag{5.2.19}$$

$$b_2 = \frac{1}{2\Omega}\left[(\Omega - \Delta)c_b(0) - \Omega_R e^{i\phi} c_a(0)\right]. \tag{5.2.20}$$

We then have

$$c_a(t) = \left\{c_a(0)\left[\cos\left(\frac{\Omega t}{2}\right) - \frac{i\Delta}{\Omega}\sin\left(\frac{\Omega t}{2}\right)\right] + i\frac{\Omega_R}{\Omega} e^{-i\phi} c_b(0) \sin\left(\frac{\Omega t}{2}\right)\right\} e^{i\Delta t/2}, \tag{5.2.21}$$

$$c_b(t) = \left\{c_b(0)\left[\cos\left(\frac{\Omega t}{2}\right) + \frac{i\Delta}{\Omega}\sin\left(\frac{\Omega t}{2}\right)\right] + i\frac{\Omega_R}{\Omega} e^{i\phi} c_a(0) \sin\left(\frac{\Omega t}{2}\right)\right\} e^{-i\Delta t/2}. \tag{5.2.22}$$

It is not difficult to verify that

$$|c_a(t)|^2 + |c_b(t)|^2 = 1, \tag{5.2.23}$$

which is a simple statement of the conservation of probability since the atom is in state $|a\rangle$ or $|b\rangle$.

If we assume that the atom is initially in the state $|a\rangle$ then $c_a(0) = 1, c_b(0) = 0$. The probabilities of the atom being in states $|a\rangle$ and $|b\rangle$ at time t are then given by $|c_a(t)|^2$ and $|c_b(t)|^2$. The inversion is given by

$$W(t) = |c_a(t)|^2 - |c_b(t)|^2 = \left(\frac{\Delta^2 - \Omega_R^2}{\Omega^2}\right)\sin^2\left(\frac{\Omega t}{2}\right) + \cos^2\left(\frac{\Omega t}{2}\right). \tag{5.2.24}$$

Under the action of the incident field, a dipole moment is induced between the two atomic levels. This induced dipole moment is given by the expectation value of the dipole moment operator

$$P(t) = e\langle\psi(t)|r|\psi(t)\rangle = C_a^* C_b \wp_{ab} + \text{c.c.} = c_a^* c_b \wp_{ab} e^{i\omega t} + \text{c.c.} \tag{5.2.25}$$

On substituting Eqs. (5.2.21) and (5.2.22) into Eq. (5.2.25), we obtain, for an atom initially in the upper level,

$$P(t) = 2\text{Re}\left\{\frac{i\Omega_R}{\Omega}\wp_{ab}\left[\cos\left(\frac{\Omega t}{2}\right) + \frac{i\Delta}{\Omega}\sin\left(\frac{\Omega t}{2}\right)\right]\sin\left(\frac{\Omega t}{2}\right) e^{i\phi} e^{i\nu t}\right\}. \tag{5.2.26}$$

The dipole moment therefore oscillates with the frequency of the incident field.

In the special case when the atom is at resonance with the incident field ($\Delta = 0$), we get $\Omega = \Omega_R$ and

$$W(t) = \cos(\Omega_R t). \tag{5.2.27}$$

The inversion oscillates between -1 and 1 at a frequency Ω_R (see Fig. 5.2).

In 1937, Rabi considered the problem of a spin-1/2 magnetic dipole undergoing precessions in a magnetic field and obtained an expression for the probability that a spin-1/2 atom incident on a Stern–Gerlach apparatus would be flipped from the $\binom{1}{0}$ or $\binom{0}{1}$ state to the $\binom{0}{1}$ or $\binom{1}{0}$ state, respectively, by an applied radio-frequency magnetic field. In the present problem, the atom undergoes a Rabi 'flopping' between the *upper and lower levels under the action of the electromagnetic field in* complete analogy with the spin-1/2 system.

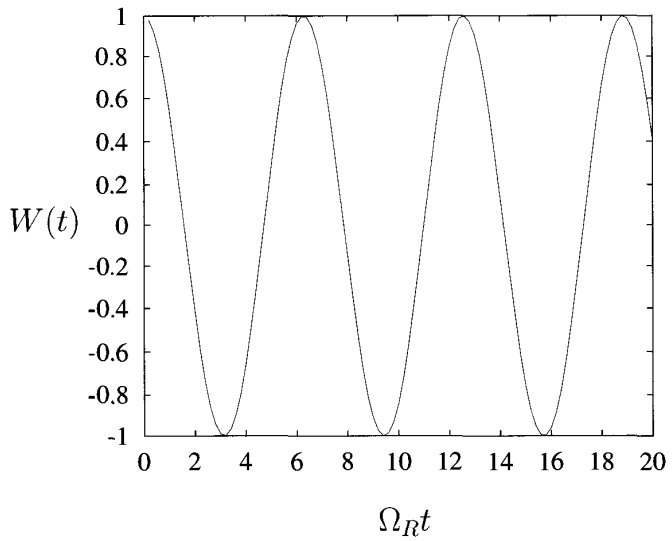


Fig. 5.2
Oscillations of the population inversion $W(t)$ as a function of time.

5.2.2 Interaction picture

Consider the Schrödinger equation

$$\frac{\partial}{\partial t}|\psi(t)\rangle = -\frac{i}{\hbar}\mathscr{H}|\psi(t)\rangle. \tag{5.2.28}$$

This equation can be integrated formally to give

$$|\psi(t)\rangle = U(t)|\psi(0)\rangle, \tag{5.2.29}$$

where the unitary time-evolution operator is defined by

$$\dot{U}(t) = -\frac{i}{\hbar}\mathscr{H}U(t), \tag{5.2.30}$$

and $U(0) = 1$.

A useful approach to the atom–field interaction problem exists in the *interaction picture* in which we assign to the state vector the time dependence due only to the interaction energy. This is accomplished by defining the state vector $|\psi_I\rangle$ in the interaction picture via

$$|\psi_I(t)\rangle = U_0^\dagger(t)|\psi(t)\rangle, \tag{5.2.31}$$

where

$$U_0(t) = \exp\left(-\frac{i}{\hbar}\mathscr{H}_0 t\right). \tag{5.2.32}$$

It then follows that

$$\frac{\partial}{\partial t}|\psi_I(t)\rangle = \left[\frac{\partial}{\partial t}U_0^\dagger(t)\right]|\psi(t)\rangle + U_0^\dagger(t)\frac{\partial}{\partial t}|\psi(t)\rangle, \tag{5.2.33}$$

and hence, from Eqs. (5.2.28), (5.2.31), and (5.2.32), we obtain

$$\frac{\partial}{\partial t}|\psi_I(t)\rangle = -\frac{i}{\hbar}\mathscr{V}(t)|\psi_I(t)\rangle. \tag{5.2.34}$$

Here

$$\mathscr{V}(t) = U_0^\dagger(t)\mathscr{H}_1 U_0(t), \tag{5.2.35}$$

is the interaction picture Hamiltonian. An operator O in the Schrödinger picture will accordingly transform as

$$O_I(t) = U_0^\dagger(t)OU_0(t). \tag{5.2.36}$$

This can be seen from the expectation value

$$\begin{aligned}\langle O\rangle &= \langle\psi(t)|O|\psi(t)\rangle \\ &= \langle\psi_I(t)|U_0^\dagger(t)OU_0(t)|\psi_I(t)\rangle \\ &= \langle\psi_I(t)|O_I(t)|\psi_I(t)\rangle.\end{aligned} \tag{5.2.37}$$

A formal solution of Eq. (5.2.34) is

$$|\psi_I(t)\rangle = U_I(t)|\psi_I(0)\rangle, \tag{5.2.38}$$

where

$$U_I(t) = \mathscr{T}\exp\left[-\frac{i}{\hbar}\int_0^t \mathscr{V}(\tau)d\tau\right] \tag{5.2.39}$$

is the time-evolution operator in the interaction picture, and $\mathscr{T}$ is the time-ordering operator, which is a shorthand notation for

$$\begin{aligned}&\mathscr{T}\exp\left[-\frac{i}{\hbar}\int_0^t \mathscr{V}(\tau)d\tau\right] \\ &= 1-\frac{i}{\hbar}\int_0^t dt_1\mathscr{V}(t_1)+\left(-\frac{i}{\hbar}\right)^2\int_0^t dt_1\int_0^{t_1} dt_2\mathscr{V}(t_1)\mathscr{V}(t_2)+\ldots\end{aligned} \tag{5.2.40}$$

In order to demonstrate the usefulness of the above formalism, we consider the interaction of a two-level atom with a monochromatic field of frequency ν. The Hamiltonian for this problem is given by Eqs. (5.2.3), (5.2.4), and (5.2.5). It follows, on using

$$\mathscr{H}_0^n = (\hbar\omega_a)^n|a\rangle\langle a| + (\hbar\omega_b)^n|b\rangle\langle b|, \tag{5.2.41}$$

that

$$U_0(t) = \exp\left(-\frac{i}{\hbar}\mathscr{H}_0 t\right)$$
$$= \exp(-i\omega_a t)|a\rangle\langle a| + \exp(-i\omega_b t)|b\rangle\langle b|. \tag{5.2.42}$$

For an atom at $z = 0$, the interaction picture Hamiltonian is, therefore, given by

$$\mathscr{V}(t) = -\hbar\Omega_R U_0^\dagger(t)(e^{-i\phi}|a\rangle\langle b| + e^{i\phi}|b\rangle\langle a|)U_0(t)\cos\nu t$$
$$= -\frac{\hbar\Omega_R}{2}[e^{-i\phi}|a\rangle\langle b|e^{i\Delta t} + e^{i\phi}|b\rangle\langle a|e^{-i\Delta t}$$
$$+e^{-i\phi}|a\rangle\langle b|e^{i(\omega+\nu)t} + e^{i\phi}|b\rangle\langle a|e^{-i(\omega+\nu)t}], \tag{5.2.43}$$

where $\Delta = \omega - \nu$. The terms proportional to $\exp[\pm i(\omega + \nu)t]$ vary very rapidly and their average over a time scale larger than $1/\nu$ is zero. These terms can therefore be neglected in the *rotating-wave approximation*. The simplified interaction picture Hamiltonian is

$$\mathscr{V}(t) = -\frac{\hbar\Omega_R}{2}\left(e^{-i\phi}|a\rangle\langle b| + e^{i\phi}|b\rangle\langle a|\right), \tag{5.2.44}$$

where we assume resonance, $\Delta = 0$. The time-evolution operator in the interaction picture $U_I(t)$ can be obtained simply from Eq. (5.2.39) by using

$$\mathscr{V}^{2n}(t) = \left(\frac{\hbar\Omega_R}{2}\right)^{2n}(|a\rangle\langle a| + |b\rangle\langle b|)^n,$$
$$\mathscr{V}^{2n+1}(t) = -\left(\frac{\hbar\Omega_R}{2}\right)^{2n+1}\left(e^{-i\phi}|a\rangle\langle b| + e^{i\phi}|b\rangle\langle a|\right). \tag{5.2.45}$$

The resulting expression for $U_I(t)$ is

$$U_I(t) = \cos\left(\frac{\Omega_R t}{2}\right)(|a\rangle\langle a| + |b\rangle\langle b|) + i\sin\left(\frac{\Omega_R t}{2}\right)\left(e^{-i\phi}|a\rangle\langle b|\right.$$
$$\left.+ e^{i\phi}|b\rangle\langle a|\right). \tag{5.2.46}$$

If the atom is initially in the excited state ($|\psi(0)\rangle \equiv |a\rangle$),

$$|\psi(t)\rangle = U_I(t)|a\rangle$$
$$= \cos\left(\frac{\Omega_R t}{2}\right)|a\rangle + i\sin\left(\frac{\Omega_R t}{2}\right)e^{i\phi}|b\rangle, \tag{5.2.47}$$

and we obtain the probability amplitudes

$$c_a(t) = \langle a|\psi\rangle = \cos\left(\frac{\Omega_R t}{2}\right), \tag{5.2.48a}$$

$$c_b(t) = \langle b|\psi\rangle = i\sin\left(\frac{\Omega_R t}{2}\right)e^{i\phi}, \tag{5.2.48b}$$

in agreement with Eqs. (5.2.21) and (5.2.22).

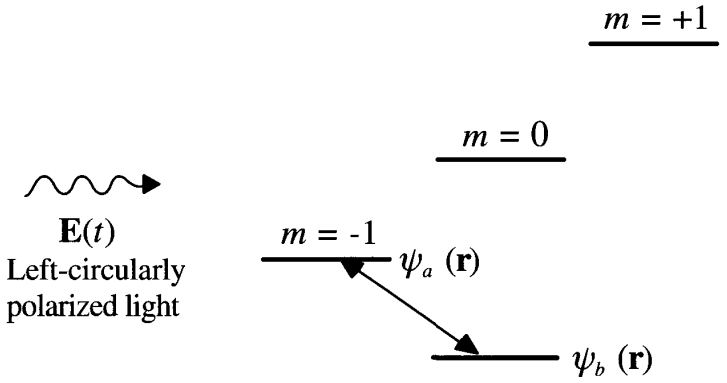


Fig. 5.3
Figure illustrating an incident electric field interacting with a hydrogen (Rydberg) atom such that the energy difference $\epsilon_{m=1} - \epsilon_b$ is much larger than $\epsilon_{m=-1} - \epsilon_b$.

5.2.3 Beyond the rotating-wave approximation

In quantum optics, the so-called rotating-wave approximation, as discussed in connection with Eq. (5.2.13), is frequently encountered. Of course, it is a very good approximation and amounts to keeping only energy-conserving terms in the Hamiltonian.

Moreover, as we show here, there are situations in which it is "exact", i.e., for all practical purposes the 'counter-rotating terms' never show up. Consider the case of a hydrogen atom in a strong magnetic field interacting with a monochromatic field as shown in Fig. 5.3. If the levels are sharp and well separated, we may focus on only the two levels, see Problem 5.7, for which

$$\psi_a(\mathbf{r}) = \frac{1}{\sqrt{64\pi a_0^3}}\frac{1}{a_0}(x - iy)\exp(-r/2a_0), \tag{5.2.49a}$$

$$\psi_b(\mathbf{r}) = \frac{1}{\sqrt{\pi a_0^3}}\exp(-r/a_0), \tag{5.2.49b}$$

where a_0 is the Bohr radius.

Using the dipole approximation (see Section 5.1.2) and placing the atom at the origin so that $\mathbf{R} = 0$, we have the interaction picture Hamiltonian

$$\mathscr{V} = -e\mathbf{r}(t)\cdot\mathbf{E}(t), \tag{5.2.50a}$$

where

$$\mathbf{r}(t) = e^{i\mathscr{H}_0 t}\mathbf{r}e^{-i\mathscr{H}_0 t}, \tag{5.2.50b}$$

and therefore

$$\mathscr{V}_{ab}(t) = -e\mathbf{r}_{ab}(t)\cdot\mathbf{E}(t) = -e\mathbf{r}_{ab}\cdot\mathbf{E}(t)e^{i\omega t}, \tag{5.2.51a}$$

$$\mathscr{V}_{ba}(t) = -e\mathbf{r}_{ba}(t)\cdot\mathbf{E}(t) = -e\mathbf{r}_{ba}\cdot\mathbf{E}(t)e^{-i\omega t}, \tag{5.2.51b}$$

where ω is the atomic frequency.

Now, for the case of linear polarization in which

$$\mathbf{E}(t) = \hat{x}\mathscr{E}\cos\nu t, \tag{5.2.52}$$

Eqs. (5.2.51a, 5.2.51b) and (5.2.52) imply

$$\begin{aligned}\mathscr{V}_{ab}(t) &= -ex_{ab}\mathscr{E}\cos\nu t e^{i\omega t}\\ &= -ex_{ab}\frac{\mathscr{E}}{2}\left[e^{i(\nu+\omega)t} + e^{-i(\nu-\omega)t}\right]\\ &\cong -ex_{ab}\frac{\mathscr{E}}{2}e^{-i(\nu-\omega)t},\end{aligned} \tag{5.2.53}$$

and likewise

$$\begin{aligned}\mathscr{V}_{ba}(t) &= -ex_{ba}\mathscr{E}\cos\nu t e^{-i\omega t}\\ &= -ex_{ba}\frac{\mathscr{E}}{2}\left[e^{i(\nu-\omega)t} + e^{-i(\nu+\omega)t}\right]\\ &\cong -ex_{ba}\frac{\mathscr{E}}{2}e^{i(\nu-\omega)t}.\end{aligned} \tag{5.2.54}$$

Thus we make the rotating-wave approximation in neglecting counter terms that go like $\exp[\pm i(\omega+\nu)t]$.

Now consider the case of left-circular polarization (LCP), which connects $\psi_a(r)$ and $\psi_b(r)$, as given by Eqs. (5.2.49). The electric field is given by

$$\mathbf{E}(t) = \hat{x}\mathscr{E}\cos\nu t - \hat{y}\mathscr{E}\sin\nu t. \tag{5.2.55}$$

Equations (5.2.53) and (5.2.54) now take the form

$$\mathscr{V}_{ab}(t) = -e\mathscr{E}\left(x_{ab}\cos\nu t + y_{ab}\sin\nu t\right)e^{i\omega t} \tag{5.2.56a}$$

$$\mathscr{V}_{ba}(t) = -e\mathscr{E}\left(x_{ba}\cos\nu t + y_{ba}\sin\nu t\right)e^{-i\omega t} \tag{5.2.56b}$$

where, in view of Eqs. (5.2.49a) and (5.2.49b), we can write

$$ex_{ab} = \int \psi_a^*(\mathbf{r})x\psi_b(\mathbf{r})dr = \wp, \tag{5.2.57a}$$

$$ey_{ab} = \int \psi_a^*(\mathbf{r})y\psi_b(\mathbf{r})dr = -i\wp, \tag{5.2.57b}$$

and similarly, $ex_{ba} = \wp$ and $ey_{ba} = i\wp$. Therefore Eqs. (5.2.56a) and (5.2.56b) take the simple form

$$\mathscr{V}_{ab}(t) = -\wp\mathscr{E}\left(\cos\nu t - i\sin\nu t\right)e^{i\omega t} = -\wp\mathscr{E}e^{-i(\nu-\omega)t} \tag{5.2.58a}$$

$$\mathscr{V}_{ba}(t) = -\wp\mathscr{E}\left(\cos\nu t + i\sin\nu t\right)e^{-i\omega t} = -\wp\mathscr{E}e^{i(\nu-\omega)t}, \tag{5.2.58b}$$

and the counter-rotating terms never appear.

Finally, we note that although there are no counter terms of the form $e^{i(\nu+\omega)t}$ associated with the LCP light inducing $\Delta m = -1$ transitions, there are counter terms associated with LCP and transitions to a state $n = 2, l = 1, m_l = +1$, i.e., $\Delta m = +1$. Such transitions are usually said to vanish due to angular momentum selection rules. Here they are seen to 'vanish' since they go as counter rotating terms. That is, they are allowed in the sense of an atom making a transition to an excited state with the emission of a photon. Such terms can be much smaller than the usual counter-rotating terms; see Problem 5.7.

5.3 Density matrix for a two-level atom*

For a given physical system, there exists a state vector $|\psi\rangle$ which contains all possible information about the system. If we want to extract a piece of the system's information, we must calculate the expectation value of the corresponding operator O,

$$\langle O\rangle_{\mathrm{QM}} = \langle\psi|O|\psi\rangle. \tag{5.3.1}$$

In many situations we may not know $|\psi\rangle$; we may only know the probability P_ψ that the system is in the state $|\psi\rangle$. For such a situation, we not only need to take the quantum mechanical average but also the ensemble average over many identical systems that have been similarly prepared. Instead of Eq. (5.3.1), we now have (see Section 3.1)

$$\langle\langle O\rangle_{\mathrm{QM}}\rangle_{\mathrm{ensemble}} = \mathrm{Tr}(O\rho), \tag{5.3.2}$$

where the density operator ρ is defined by

$$\rho = \sum_\psi P_\psi|\psi\rangle\langle\psi|. \tag{5.3.3}$$

It can be seen that

$$\mathrm{Tr}(O\rho) = \mathrm{Tr}(\rho O). \tag{5.3.4}$$

In the particular case where all P_ψ are zero except the one for a state $|\psi_0\rangle$, then

$$\rho = |\psi_0\rangle\langle\psi_0|, \tag{5.3.5}$$

and the state is called a pure state. It follows from the conservation of probability that $\mathrm{Tr}(\rho) = 1$. Also, for a pure state,

$$\mathrm{Tr}(\rho^2) = 1. \tag{5.3.6}$$

* The more complete picture of stimulated emission as developed by Lamb and Scully [1971] is found in Chapter III of Sargent, Scully and Lamb [1974].

5.3.1 Equation of motion for the density matrix

We can obtain the equation of motion for the density matrix from the Schrödinger equation,

$$|\dot{\psi}\rangle = -\frac{i}{\hbar}\mathscr{H}|\psi\rangle. \tag{5.3.7}$$

Taking the time derivative of ρ (Eq. (5.3.3)) we have

$$\dot{\rho} = \sum_{\psi} P_{\psi}(|\dot{\psi}\rangle\langle\psi| + |\psi\rangle\langle\dot{\psi}|), \tag{5.3.8}$$

where P_{ψ} is time independent. Using Eq. (5.3.7) to replace $|\dot{\psi}\rangle$ and $\langle\dot{\psi}|$ in Eq. (5.3.8) we get

$$\dot{\rho} = -\frac{i}{\hbar}[\mathscr{H}, \rho]. \tag{5.3.9}$$

Equation (5.3.9) is often called the Liouville or Von Neumann equation of motion for the density matrix. It is more general than the Schrödinger equation since it uses the density operator instead of a specific state vector and can therefore give statistical as well as quantum mechanical information.

In Eq. (5.3.9), we have not included the decay of the atomic levels due to spontaneous emission. The excited atomic levels can also decay because of collisions and other phenomena. The finite lifetime of the atomic levels can be described very well by adding phenomenological decay terms to the density operator equation (5.3.9) (see also Problem 5.2).

The decay rates can be incorporated in Eq. (5.3.9) by a relaxation matrix Γ, which is defined by the equation

$$\langle n|\Gamma|m\rangle = \gamma_n \delta_{nm}. \tag{5.3.10}$$

With this addition, the density matrix equation of motion becomes

$$\dot{\rho} = -\frac{i}{\hbar}[\mathscr{H}, \rho] - \frac{1}{2}\{\Gamma, \rho\}, \tag{5.3.11}$$

where $\{\Gamma, \rho\} = \Gamma\rho + \rho\Gamma$. In general, the relaxation processes are more complicated.

The ijth matrix element of Eq. (5.3.11) is

$$\dot{\rho}_{ij} = -\frac{i}{\hbar}\sum_k (\mathscr{H}_{ik}\rho_{kj} - \rho_{ik}\mathscr{H}_{kj}) - \frac{1}{2}\sum_k (\Gamma_{ik}\rho_{kj} + \rho_{ik}\Gamma_{kj}). \tag{5.3.12}$$

This formula is useful in the treatment of many-level systems.

5.3.2 Two-level atom

We now consider the two-level atomic system again where the state of the system is a linear combination of states $|a\rangle$ and $|b\rangle$, i.e., $|\psi\rangle = C_a|a\rangle + C_b|b\rangle$. Then the density matrix operator can be written as

$$\begin{aligned}\rho = |\psi\rangle\langle\psi| &= [C_a(t)|a\rangle + C_b(t)|b\rangle]\ [C_a^*(t)\langle a| + C_b^*(t)\langle b|] \\ &= |C_a|^2|a\rangle\langle a| + C_aC_b^*|a\rangle\langle b| + C_bC_a^*|b\rangle\langle a| + |C_b|^2|b\rangle\langle b|.\end{aligned} \tag{5.3.13}$$

Taking the matrix elements, we get

$$\rho_{aa} = \langle a|\rho|a\rangle = |C_a(t)|^2, \tag{5.3.14}$$

$$\rho_{ab} = \langle a|\rho|b\rangle = C_a(t)C_b^*(t), \tag{5.3.15}$$

$$\rho_{ba} = \rho_{ab}^*, \tag{5.3.16}$$

$$\rho_{bb} = \langle b|\rho|b\rangle = |C_b(t)|^2. \tag{5.3.17}$$

The matrix form of the density operator is

$$\rho = \begin{pmatrix} \rho_{aa} & \rho_{ab} \\ \rho_{ba} & \rho_{bb} \end{pmatrix}. \tag{5.3.18}$$

It is obvious that ρ_{aa} and ρ_{bb} are the probabilities of being in the upper and lower states, respectively. For the meaning of the off-diagonal elements we need to remember that the atomic polarization, see Eq. (5.2.25), of the two-level atom (at z) is

$$P(z,t) = C_aC_b^*\wp_{ba} + \text{c.c.} = \rho_{ab}(z,t)\wp_{ba} + \text{c.c.} \tag{5.3.19}$$

So we see that the off-diagonal elements determine the atomic polarization.

We could have found this form for ρ more directly from Eq. (5.3.5) by remembering that in spinor notation

$$|\psi\rangle = \begin{pmatrix} C_a \\ C_b \end{pmatrix}; \qquad \langle\psi| = (C_a^*\ C_b^*). \tag{5.3.20}$$

Then by matrix multiplication

$$\rho = \begin{pmatrix} C_a \\ C_b \end{pmatrix}(C_a^*\ C_b^*) = \begin{pmatrix} |C_a|^2 & C_aC_b^* \\ C_bC_a^* & |C_b|^2 \end{pmatrix}. \tag{5.3.21}$$

We can derive the equations of motion for the density matrix elements from Eq. (5.3.12) with the Hamiltonian given by Eqs. (5.2.4)

and (5.2.5). The resulting equations are

$$\dot{\rho}_{aa} = -\gamma_a \rho_{aa} + \frac{i}{\hbar}[\wp_{ab} E \rho_{ba} - \text{c.c.}], \tag{5.3.22}$$

$$\dot{\rho}_{bb} = -\gamma_b \rho_{bb} - \frac{i}{\hbar}[\wp_{ab} E \rho_{ba} - \text{c.c.}], \tag{5.3.23}$$

$$\dot{\rho}_{ab} = -(i\omega + \gamma_{ab})\rho_{ab} - \frac{i}{\hbar}\wp_{ab} E(\rho_{aa} - \rho_{bb}), \tag{5.3.24}$$

where $\gamma_{ab} = (\gamma_a + \gamma_b)/2$ with γ_a and γ_b defined by Eq. (5.3.10) and $E(t)$ is given by Eq. (5.2.6). In the rotating-wave approximation, $\cos(\nu t)$ is replaced by $\exp(-i\nu t)/2$ in Eqs. (5.3.22)–(5.3.24).

5.3.3 *Inclusion of elastic collisions between atoms*

The physical interpretation of the elements of the density matrix allows us to include in these equations terms associated with certain processes. One such process is the elastic collision between atoms in a gas.

In particular, during an atom–atom collision the energy levels experience random Stark shifts without a change of state and the decay rate for ρ_{ab} is increased without much change in γ_a and γ_b. The change in the decay rate of ρ_{ab} may be computed in a simple way as follows.

We assume that the random Stark shifts are included in Eq. (5.3.24) by adding a random shift $\delta\omega(t)$ to the energy difference ω. Ignoring the atom–field interactions for simplicity, we can write the equation of motion for the density matrix element ρ_{ab} as

$$\dot{\rho}_{ab} = -[i\omega + i\delta\omega(t) + \gamma_{ab}]\rho_{ab}. \tag{5.3.25}$$

Integrating Eq. (5.3.25) formally, we have

$$\rho_{ab}(t) = \exp\left[-(i\omega + \gamma_{ab})t - i\int_0^t dt'\,\delta\omega(t')\right]\rho_{ab}(0). \tag{5.3.26}$$

We now perform an ensemble average of (5.3.26) over the random variations in $\delta\omega(t)$. This average affects only the $\delta\omega(t)$ factor, so that we find $\langle \exp[-i\int_0^t dt'\,\delta\omega(t')]\rangle$.

The function $\delta\omega$ is as often positive as negative. Hence the ensemble average $\langle\delta\omega(t)\rangle$ is zero. Furthermore, as the variations in $\delta\omega(t)$ are usually rapid compared to other changes which occur in times like $1/\gamma_{ab}$, we take

$$\langle\delta\omega(t)\delta\omega(t')\rangle = 2\gamma_{\text{ph}}\delta(t - t'), \tag{5.3.27}$$

where γ_{ph} is a constant. We also assume that $\delta\omega(t)$ is described by a Gaussian random process, so that the well-known moment theorem

of Gaussian processes is valid. Under these conditions we obtain

$$\left\langle \exp\left[-i\int_0^t dt'\,\delta\omega(t')\right]\right\rangle = \exp(-\gamma_{\text{ph}}t), \tag{5.3.28}$$

which gives for the average of (5.3.26)

$$\rho_{ab}(t) = \exp[-(i\omega + \gamma_{ab} + \gamma_{\text{ph}})t]\rho_{ab}(0). \tag{5.3.29}$$

It follows, on differentiating this equation and including the interaction term, that we have the modified equation of motion for ρ_{ab}:

$$\dot{\rho}_{ab} = -(i\omega + \gamma)\rho_{ab} - \frac{i}{\hbar}\wp_{ab} E(z,t)(\rho_{aa} - \rho_{bb}), \tag{5.3.30}$$

where $\gamma = \gamma_{ab} + \gamma_{\text{ph}}$ is the new decay rate. Equation (5.3.30) is an average equation with respect to collisions.

5.4 Maxwell–Schrödinger equations

The interaction of a single atom with the single-mode field, which was discussed in the previous sections, represents a simple, idealized system. In many problems of interest in quantum optics, one is interested in the interaction of the radiation field with a large number of atoms. The prime example of such a system is a single-mode laser where atoms pumped into the excited level interact with the electromagnetic field inside a cavity . Other examples include coherent pulse propagation and optical bistability.

In this section, we develop a mathematical framework to treat such problems based on a self-consistent set of equations for the matter and the field. This set of equations and its extensions enable us to deal with many semiclassical problems where the atoms are treated quantum mechanically and the field is treated classically.

In the present semiclassical atom–field interaction, the classical field induces electric dipole moments in the medium according to the laws of quantum mechanics. The density matrix is used to facilitate the statistical summations involved in obtaining the macroscopic polarization of the medium for the individual dipole moments. The semiclassical approach, though remarkably good for many problems of interest in the study of a given system, is however inadequate to provide information about the quantum statistical features of light. These aspects will be presented in later chapters where the radiation field will be treated quantum mechanically.

5.4.1 Population matrix and its equation of motion

We consider the interaction of an electromagnetic field with a medium which consists of two-level homogeneously broadened atoms. The individual atoms are described by the density operator (see Eqs. (5.3.14)–(5.3.17))

$$\rho(z,t,t_0)=\sum_{\alpha,\beta}\rho_{\alpha\beta}(z,t,t_0)|\alpha\rangle\langle\beta|, \tag{5.4.1}$$

where $\alpha,\beta=a,b$ and $\rho_{\alpha\beta}(z,t,t_0)$ are the density matrix elements for an individual atom at time t and position z, which starts interacting with the field at an initial time t_0. The initial time t_0 can be random. The single-atom density matrix elements $\rho_{\alpha\beta}(z,t,t_0)$ obey the equations of motion (5.3.22), (5.3.23), and (5.3.30). If the state of the atom at the time of injection is described by

$$\rho(z,t_0,t_0)=\sum_{\alpha,\beta}\rho^{(0)}_{\alpha\beta}|\alpha\rangle\langle\beta|, \tag{5.4.2}$$

then

$$\rho_{\alpha\beta}(z,t_0,t_0)=\rho^{(0)}_{\alpha\beta}. \tag{5.4.3}$$

The effect of all the atoms which are pumped at the rate $r_a(z,t_0)$ atoms per second per unit volume is obtained by summing over initial times. The resulting *population matrix* is defined as

$$\begin{aligned}\rho(z,t)&=\int_{-\infty}^{t}dt_0 r_a(z,t_0)\rho(z,t,t_0)\\&=\sum_{\alpha,\beta}\int_{-\infty}^{t}dt_0 r_a(z,t_0)\rho_{\alpha\beta}(z,t,t_0)|\alpha\rangle\langle\beta|.\end{aligned} \tag{5.4.4}$$

Generally the excitation $r_a(z,t_0)$ varies slowly and can be taken to be a constant. The macroscopic polarization of the medium, $P(z,t)$, will be produced by an ensemble of atoms that arrive at z at time t, regardless of their time of excitation, i.e.,

$$\begin{aligned}P(z,t)&=\int_{-\infty}^{t}dt_0 r_a(z,t_0)\mathrm{Tr}[\hat{\wp}\rho(z,t,t_0)]\\&=\sum_{\alpha,\beta}\int_{-\infty}^{t}dt_0 r_a(z,t_0)\rho_{\alpha\beta}(z,t,t_0)\wp_{\beta\alpha},\end{aligned} \tag{5.4.5}$$

where $\hat{\wp}$ is the dipole moment operator and, in the second line, we have substituted for $\rho(z,t,t_0)$ from Eq. (5.4.1). For a two-level atom, with $\wp_{ab}=\wp_{ba}=\wp$, we obtain

$$P(z,t)=\wp[\rho_{ab}(z,t)+\text{c.c.}]. \tag{5.4.6}$$

Thus the off-diagonal elements of the population matrix determine the macroscopic polarization.

The equations of motion for the elements of the population matrix $\rho(z,t)$ can be obtained by taking the time derivative of Eq. (5.4.4) and using Eqs. (5.3.22), (5.3.23), and (5.3.30). For example, if the atoms are incoherently excited to levels $|a\rangle$ and $|b\rangle$ at a constant rate r_a ($\rho^{(0)}_{ab}=\rho^{(0)}_{ba}=0$), we obtain

$$\dot{\rho}_{aa}=\lambda_a-\gamma_a\rho_{aa}+\frac{i}{\hbar}(\wp E\rho_{ba}-\text{c.c.}), \tag{5.4.7}$$

$$\dot{\rho}_{bb}=\lambda_b-\gamma_b\rho_{bb}-\frac{i}{\hbar}(\wp E\rho_{ba}-\text{c.c.}), \tag{5.4.8}$$

$$\dot{\rho}_{ab}=-(i\omega+\gamma)\rho_{ab}-\frac{i}{\hbar}\wp E(\rho_{aa}-\rho_{bb}), \tag{5.4.9}$$

where $\lambda_a=r_a\rho^{(0)}_{aa}$ and $\lambda_b=r_a\rho^{(0)}_{bb}$. These equations for the two-level atomic medium are coupled to the field E. The condition of self-consistency requires that the equation of motion for the field E is driven by the atomic population matrix elements. In the following section, we derive such an equation for a single-mode running wave.

5.4.2 Maxwell's equations for slowly varying field functions

The electromagnetic field radiation is described by Maxwell's equations:

$$\nabla\cdot\mathbf{D}=0,\qquad \nabla\times\mathbf{E}=-\frac{\partial\mathbf{B}}{\partial t}, \tag{5.4.10}$$

$$\nabla\cdot\mathbf{B}=0,\qquad \nabla\times\mathbf{H}=\mathbf{J}+\frac{\partial\mathbf{D}}{\partial t}, \tag{5.4.11}$$

where

$$\mathbf{D}=\epsilon_0\mathbf{E}+\mathbf{P},\quad \mathbf{B}=\mu_0\mathbf{H},\quad \mathbf{J}=\sigma\mathbf{E}. \tag{5.4.12}$$

Here $\mathbf{P}$ is the macroscopic polarization of the medium. In order to avoid a complicated boundary-value problem, we assume the presence of a medium with conductivity σ. This conductivity is intended to take into account phenomenologically the linear losses due to any absorbing background medium, and also those losses due to diffraction and mirror transmission. Combining the curl equations, taking the appropriate time derivatives, and using Eq. (5.4.12), we get the wave equation

$$\nabla\times(\nabla\times\mathbf{E})+\mu_0\sigma\frac{\partial\mathbf{E}}{\partial t}+\mu_0\epsilon_0\frac{\partial^2\mathbf{E}}{\partial t^2}=-\mu_0\frac{\partial^2\mathbf{P}}{\partial t^2}. \tag{5.4.13}$$

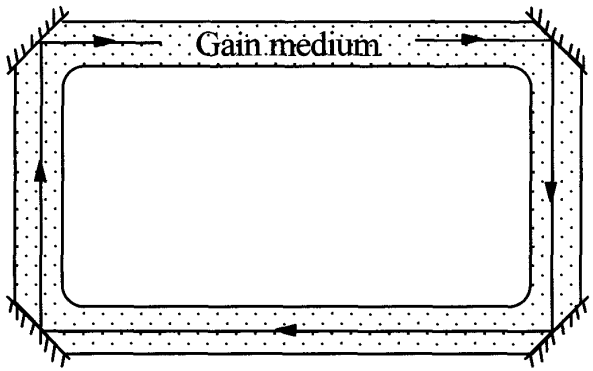


Fig. 5.4
Schematic diagram of a laser in a unidirectional ring configuration.

The polarization $\mathbf{P}(\mathbf{r},t)$ thus acts as a source term in the equation for the radiation field. We have in mind a situation in which the radiation field interacts with two-level atoms inside a unidirectional ring cavity as shown in Fig. 5.4. Usually both running waves exist inside the cavity. The unidirectional situation is achieved by the insertion of a device with high loss for one running wave. The variations in the field intensity transverse to the laser axis are typically slowly varying on the scale of the optical wavelength. Hence, we neglect the x- and y-dependence of $\mathbf{E}$, i.e.,

$$\mathbf{E}(\mathbf{r},t) = E(z,t)\hat{\mathbf{x}}. \tag{5.4.14}$$

Equation (5.4.13) then reduces to

$$-\frac{\partial^2 E}{\partial z^2} + \mu_0\sigma\frac{\partial E}{\partial t} + \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} = -\mu_0\frac{\partial^2 P}{\partial t^2}. \tag{5.4.15}$$

The field of frequency ν is represented as a running wave

$$E(z,t) = \frac{1}{2}\mathscr{E}(z,t)e^{-i[\nu t - kz + \phi(z,t)]} + \text{c.c.}, \tag{5.4.16}$$

where $\mathscr{E}(z,t)$ and $\phi(z,t)$ are slowly varying functions of position and time with $k = \nu/c$. For the problem of laser oscillation, $k = \nu_c/c$ where ν_c is the cavity frequency. In general, $\mathscr{E}(z,t)$ is a complex function; however, in the present and in the next section, we assume it to be real.

If the field is written as in Eq. (5.4.16) then the response of the medium, neglecting higher harmonics, is given by the polarization

$$P(z,t) = \frac{1}{2}\mathscr{P}(z,t)e^{-i[\nu t - kz + \phi(z,t)]} + \text{c.c.}, \tag{5.4.17}$$

where $\mathscr{P}(z,t)$ is a slowly varying function of position and time.

The slowly varying complex polarization $\mathscr{P}(z,t)$ is given in terms of the population matrix by identification of the positive frequency parts in Eqs. (5.4.6) and (5.4.17):

$$\mathscr{P}(z,t) = 2\wp\rho_{ab}e^{i[\nu t - kz + \phi(z,t)]}. \tag{5.4.18}$$

The expressions for $E(z,t)$ and $P(z,t)$ are substituted from Eqs. (5.4.16) and (5.4.17) in Eq. (5.4.15), and the following approximations are made

$$\frac{\partial\mathscr{E}}{\partial t} \ll \nu\mathscr{E}, \quad \frac{\partial\mathscr{E}}{\partial z} \ll k\mathscr{E}, \quad \frac{\partial\phi}{\partial t} \ll \nu, \quad \frac{\partial\phi}{\partial z} \ll k, \tag{5.4.19}$$

$$\frac{\partial\mathscr{P}}{\partial t} \ll \nu\mathscr{P}, \quad \frac{\partial\mathscr{P}}{\partial z} \ll k\mathscr{P}. \tag{5.4.20}$$

These slowly varying amplitude and phase approximations are justified when $\mathscr{E}$, ϕ, and $\mathscr{P}$ do not change appreciably in an optical frequency period. By noting that Eq. (5.4.15) can be rewritten as

$$\left(\frac{\partial}{\partial z} + \frac{1}{c}\frac{\partial}{\partial t}\right)\left(-\frac{\partial}{\partial z} + \frac{1}{c}\frac{\partial}{\partial t}\right)E = -\mu_0\sigma\frac{\partial E}{\partial t} - \mu_0\frac{\partial^2 P}{\partial t^2}, \tag{5.4.21}$$

and

$$\left(-\frac{\partial}{\partial z} + \frac{1}{c}\frac{\partial}{\partial t}\right)E \cong -2ikE, \tag{5.4.22}$$

we obtain

$$\frac{\partial\mathscr{E}}{\partial z} + \frac{1}{c}\frac{\partial\mathscr{E}}{\partial t} = -\kappa\mathscr{E} - \frac{1}{2\epsilon_0}k\,\text{Im}\mathscr{P}, \tag{5.4.23}$$

$$\frac{\partial\phi}{\partial z} + \frac{1}{c}\frac{\partial\phi}{\partial t} = k - \frac{\nu}{c} - \frac{1}{2\epsilon_0}k\mathscr{E}^{-1}\text{Re}\mathscr{P}, \tag{5.4.24}$$

where $\kappa = \sigma/2\epsilon_0 c$ is the linear loss coefficient.

Equations (5.4.7)–(5.4.9), (5.4.23), and (5.4.24) form a self-consistent set of equations. This set of equations is the starting point of the study of many systems involving the interaction of the radiation field with an ensemble of atoms. The generalization of this set of equations to a multi-level atomic system and a multi-mode field is straightforward.

As an important example of the applications of this set of equations, we present the semiclassical theory of the laser in the next section.

5.5 Semiclassical laser theory

In this section, we first outline the basic principle of laser operation and then present a theory of the laser as developed principally by Lamb and co-workers. The threshold condition for a laser and the evolution equation of the electromagnetic field is also derived.

5.5.1 Basic principle

In 1954, Gordon, Zeiger, and Townes showed that coherent electromagnetic radiation can be generated in the radio frequency range by the so-called maser (microwave amplification by stimulated emission of radiation). The first maser action was observed in ammonia.

The maser principle was extended by Schawlow and Townes, and also by Prokhorov, to the optical domain, thus obtaining a laser (light amplification by stimulated emission of radiation). A laser consists of a set of atoms interacting with an electromagnetic field inside a cavity. The cavity supports only a specific set of modes corresponding to a discrete sequence of eigenfrequencies. The active atoms, i.e., the ones that are pumped to the upper level of the laser transition, are in resonance with one of the eigenfrequencies of the cavity. A resonant electromagnetic field gives rise to stimulated emission, and the atoms transfer their excitation energy to the radiation field. The emitted radiation is still at resonance. If the upper level is sufficiently populated, this radiation gives rise to further transitions in other atoms. In this way all the excitation energy of the atoms is transferred to a single mode of the radiation field.

The first pulsed laser operation was demonstrated by Maiman in ruby. The first continuous wave (cw) laser, a He–Ne gas laser, was built by Javan. Since then, a large variety of systems have been demonstrated to exhibit lasing action; generating coherent light over a frequency domain ranging from infrared to ultraviolet. These include dye lasers, chemical lasers, and semiconductor lasers. A new class of lasers which uses electrons in a periodic magnetic field (called free-electron lasers) has also been developed.

From our discussion of the laser principle, it is clear that a laser theory should incorporate three basic elements, an active medium (two-level atoms with population inversion), pumping to the upper lasing level, and the radiation losses due to the cavity. A systematic semiclassical theory of the laser was developed by Lamb.

5.5.2 Lamb's semiclassical theory

We consider the semiclassical laser theory for the simple case of a linearly polarized electric field in a unidirectional ring cavity and two-level homogeneously broadened, active atoms.

The time dependence of the electric field $\mathscr{E}(z,t)$ can be separated from the spatial part by expanding the field in the normal modes

of the cavity. In a ring cavity only certain discrete modes achieve appreciable magnitude, namely, those with the frequencies

$$\nu_m = \frac{m\pi c}{S} = k_m c, \tag{5.5.1}$$

where S is the circumference of the ring, m is a large integer (typically of the order 10^6), and k_m is the corresponding wave number. Here, we consider a single mode with unidirectional (running-wave) mode functions $U(z) = \exp(ikz)$ (Fig. 5.4).

The equations of motion for the field amplitude (5.4.23) and phase (5.4.24) for the present problem reduce to

$$\frac{\partial \mathscr{E}}{\partial t} = -\frac{1}{2}\mathscr{C}\mathscr{E} - \frac{1}{2}\left(\frac{\nu}{\epsilon_0}\right)\mathrm{Im}\mathscr{P}, \tag{5.5.2a}$$

$$\frac{\partial \phi}{\partial t} = (\nu_c - \nu) - \frac{1}{2}\left(\frac{\nu}{\epsilon_0}\right)\mathscr{E}^{-1}\mathrm{Re}\mathscr{P}, \tag{5.5.2b}$$

where ν_c is the cavity frequency and $\gamma = (\gamma_a + \gamma_b)/2$. In Eq. (5.5.2a), κ has been replaced by $\mathscr{C}/2c$ where $\mathscr{C} = \nu_c/Q$ (where Q is the quality factor of the cavity) to account for the field losses through the mirrors of the cavity. The driving polarization $\mathscr{P}$ (Eq. (5.4.18)) is determined by Eq. (5.4.9) which yields

$$\mathscr{P}(z,t) = \frac{-i\wp^2}{\hbar}\int_{-\infty}^{t} \exp[-\gamma(t-t') - i(\omega-\nu)(t-t')] \times \mathscr{E}(t')[\rho_{aa}(t') - \rho_{bb}(t')]dt'. \tag{5.5.3}$$

The integral (5.5.3) can be simply performed, provided the amplitude $\mathscr{E}(t')$ and the population difference $\rho_{aa}-\rho_{bb}$ do not change appreciably in the time $1/\gamma$, for then these terms can be factored outside the integral. This solution leads to rate equations for the atomic populations. These approximations are exact in steady state ($\dot{\mathscr{P}} = 0$). This gives

$$\mathrm{Im}\mathscr{P}(t) = \frac{-\wp^2}{\hbar}\mathscr{E}(t)\gamma\frac{\rho_{aa}(t) - \rho_{bb}(t)}{\gamma^2 + (\omega-\nu)^2}, \tag{5.5.4a}$$

$$\mathrm{Re}\mathscr{P}(t) = \frac{-\wp^2}{\hbar}\mathscr{E}(t)(\omega-\nu)\frac{\rho_{aa}(t) - \rho_{bb}(t)}{\gamma^2 + (\omega-\nu)^2}. \tag{5.5.4b}$$

On substituting Eqs. (5.5.4) into the equations of motion for ρ_{aa} and ρ_{bb} ((5.4.7) and (5.4.8)), we obtain the rate equations

$$\dot{\rho}_{aa} = \lambda_a - \gamma_a\rho_{aa} - R(\rho_{aa} - \rho_{bb}), \tag{5.5.5a}$$

$$\dot{\rho}_{bb} = \lambda_b - \gamma_b\rho_{bb} + R(\rho_{aa} - \rho_{bb}), \tag{5.5.5b}$$

where the rate constant is

$$R = \frac{1}{2}\left(\frac{\wp\mathscr{E}}{\hbar}\right)^2 \frac{\gamma}{\gamma^2 + (\omega-\nu)^2}. \tag{5.5.6}$$

It is evident that the rate constant R, which determines the rate at which the population difference varies in time, depends primarily on the rate at which the total field intensity varies. Hence, the rate-equation approximation consists of the assumption that the electric field envelope varies slowly in atomic lifetimes. We can determine the population difference in the steady state from Eqs. (5.5.5). This difference can be substituted in turn back into the equations for Im$\mathscr{P}$ and Re$\mathscr{P}$, thus determining the polarization components.

In the steady state ($\dot{\rho}_{aa} = \dot{\rho}_{bb} = 0$), Eqs. (5.5.5) yield

$$\rho_{aa} - \rho_{bb} = \frac{N_0}{1 + R/R_s}, \tag{5.5.7}$$

where $N_0 = \lambda_a \gamma_a^{-1} - \lambda_b \gamma_b^{-1}$ and $R_s = \gamma_a \gamma_b / 2\gamma$. The population difference is therefore given by N_0, which appears in the absence of the field, divided by a factor which increases as the intensity of the electric field increases.

Combining Eq. (5.5.7) with Eqs. (5.5.4) and (5.5.2) we obtain the amplitude and frequency determining equations

$$\dot{\mathscr{E}} = -\frac{\mathscr{C}}{2}\mathscr{E} + \frac{\mathscr{A}\mathscr{E}}{2\left[1 + \frac{\mathscr{B}}{\mathscr{A}}\left(\frac{\epsilon_0 V}{2\hbar\nu}\right)\mathscr{E}^2\right]}, \tag{5.5.8}$$

$$\nu + \dot{\phi} = \nu_c + \frac{(\omega - \nu)\mathscr{A}}{2\gamma\left[1 + \frac{\mathscr{B}}{\mathscr{A}}\left(\frac{\epsilon_0 V}{2\hbar\nu}\right)\mathscr{E}^2\right]}, \tag{5.5.9}$$

where V is the volume of the cavity and

$$\mathscr{A} = \left(\frac{\wp^2 \nu \gamma}{\epsilon_0 \hbar}\right) \frac{N_0}{\gamma^2 + (\omega - \nu)^2}, \tag{5.5.10a}$$

$$\mathscr{B} = \left(\frac{4\wp^2}{\hbar^2}\right)\left(\frac{\gamma^2}{\gamma_a \gamma_b}\right) \frac{\mathscr{A}}{\gamma^2 + (\omega - \nu)^2}\left(\frac{\hbar\nu}{2\epsilon_0 V}\right). \tag{5.5.10b}$$

Here $\mathscr{A}$ is the linear gain parameter and $\mathscr{B}$ is the saturation parameter.

We now define a dimensionless intensity

$$n = \frac{\epsilon_0 \mathscr{E}^2 V}{2\hbar\nu}, \tag{5.5.11}$$

which corresponds to the 'number of photons' in the laser. (Here, $\epsilon_0 \mathscr{E}^2 V/2$ is the total energy in the laser beam and $\hbar\nu$ is the energy associated with a single photon.) This statement will be sharpened when we study the quantum theory of the laser in Chapter 11. It follows from Eqs. (5.5.8) and (5.5.9) that

$$\dot{n} = -\mathscr{C}n + \frac{\mathscr{A}n}{1 + \frac{\mathscr{B}}{\mathscr{A}}n}, \tag{5.5.12}$$

$$\nu + \dot{\phi} = \nu_c + \frac{(\omega - \nu)\mathscr{A}}{2\gamma\left(1 + \frac{\mathscr{B}}{\mathscr{A}}n\right)}. \tag{5.5.13}$$

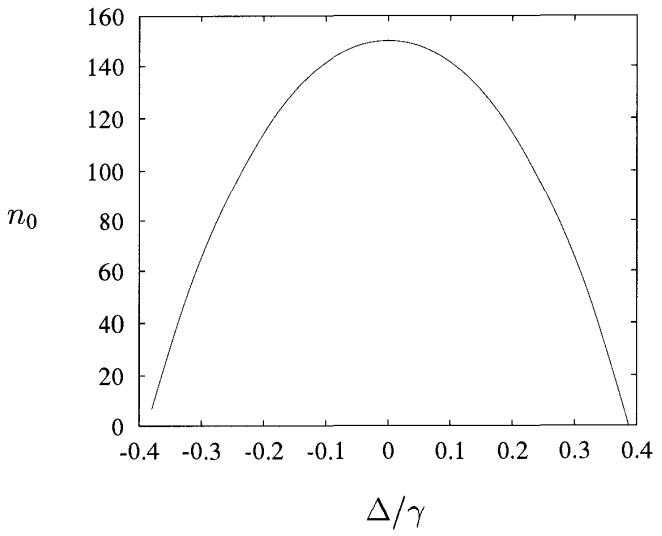


Fig. 5.5
A plot of the steady-state intensity n_0 versus the detuning δ/γ. Here $\mathscr{C} = 1\mu\text{sec}^{-1}$, $\wp^2 \nu N_0/\epsilon_0 \hbar\gamma = 1.15\mu\text{sec}^{-1}$, and $2\wp^4 \nu^2 N_0/\epsilon_0^2 \hbar^2 \gamma_a \gamma_b \gamma V = 10^{-3}\mu\text{sec}^{-1}$.

For small excitations ($\mathscr{B}n/\mathscr{A} \ll 1$), a perturbation theory is obtained by expanding the denominator in Eqs. (5.5.12) and (5.5.13), resulting in

$$\dot{n} = (\mathscr{A} - \mathscr{C})n - \mathscr{B}n^2, \tag{5.5.14}$$

$$\nu + \dot{\phi} = \nu_c + \left(\frac{\omega - \nu}{2\gamma}\right)(\mathscr{A} - \mathscr{B}n). \tag{5.5.15}$$

Equations (5.5.14) and (5.5.15) are the basic equations for the laser. As shown below, they yield the laser threshold condition, the steady-state and transient intensity of the laser, and the frequency pulling due to the presence of the gain medium.

It is easily seen from Eq. (5.5.14) that, in steady state ($\dot{n} = 0$), $n = 0$ unless $\mathscr{A} > \mathscr{C}$. When $\mathscr{A} > \mathscr{C}$, the steady-state intensity is given by

$$n_0 \equiv n = \frac{\mathscr{A} - \mathscr{C}}{\mathscr{B}}. \tag{5.5.16}$$

Thus, the laser threshold condition is $\mathscr{A} = \mathscr{C}$, i.e., when the gain is equal to the cavity losses.

In Fig. 5.5, the steady-state intensity is plotted against the detuning $\Delta = \omega - \nu$. According to Eq. (5.5.15), the oscillation frequency ν itself depends on the intensity. A good approximation, however, results from taking $\nu = \nu_c$ in the calculation of the various coefficients.

The frequency determining Eq. (5.5.15) predicts a pulling of the oscillation frequency from the passive cavity frequency towards line

center. Specifically, in steady state ($\dot{\phi} = 0$)

$$\nu = \frac{\nu_c + \mathscr{S}\omega}{1 + \mathscr{S}}, \tag{5.5.17}$$

where the stabilization factor

$$\mathscr{S} = \frac{\mathscr{A} - \mathscr{B}n_0}{2\gamma} = \frac{\mathscr{C}}{2\gamma}. \tag{5.5.18}$$

Equation (5.5.17) can be interpreted as a center-of-mass equation in which the oscillation frequency ν assumes the average value of ν_c and ω with weights 1 and $\mathscr{S}$, respectively. In the typical case, $\mathscr{C} \ll 2\gamma$ and therefore $\nu \cong \nu_c$, but ν is pulled closer to the atomic frequency ω. This is called mode pulling.

5.6 A physical picture of stimulated emission and absorption

In order to better appreciate the physics behind stimulated emission and absorption, let us consider an atom at the point $z = 0$ interacting with the field $E(z,t) = \mathscr{E}(z,t)\cos(\nu t - kz)$. As before, the amplitudes C_a and C_b are determined by Eqs. (5.2.7) and (5.2.8), and the slowly varying amplitudes $c_a = C_a e^{i\omega_a t}$ and $c_b = C_b e^{i\omega_b t}$ are determined by Eqs. (5.2.12) and (5.2.13), respectively. For simplicity, we assume exact resonance $\Delta = \omega - \nu = 0$. Then the solution (5.2.21)–(5.2.22) becomes

$$c_a(t) = \left[c_a(0)\cos\left(\frac{\Omega_R t}{2}\right) + ic_b(0)\sin\left(\frac{\Omega_R t}{2}\right)\right], \tag{5.6.1a}$$

$$c_b(t) = \left[c_b(0)\cos\left(\frac{\Omega_R t}{2}\right) + ic_a(0)\sin\left(\frac{\Omega_R t}{2}\right)\right], \tag{5.6.1b}$$

where we have assumed a real dipole matrix element $\wp_{ab} = \wp_{ba} \equiv \wp$. Now, to the lowest order, we may trivially calculate $\rho_{ab} = c_a c_b^* e^{-i\omega t}$ for the cases of atom in the excited state and the ground state.

For the first case (stimulated emission), in which $c_a(0) = 1$ and $c_b(0) = 0$, we find to lowest order for an atom which passes through the laser cavity in a time τ

$$c_a(\tau) \cong 1, \tag{5.6.2a}$$

$$c_b(\tau) \cong i\frac{\Omega_R \tau}{2}, \tag{5.6.2b}$$

and the polarization is then (see Eq. (5.4.18))

$$\begin{aligned}\mathscr{P} &= 2\wp\rho_{ab}e^{i\nu\tau} \\ &\cong -i\wp\Omega_R\tau.\end{aligned} \tag{5.6.3}$$

For the case of absorption, initially $c_a(0) = 0$, $c_b(0) = 1$, to the lowest order one gets

$$c_a(\tau) \cong i\frac{\Omega_R \tau}{2}, \tag{5.6.4a}$$

$$c_b(\tau) \cong 1, \tag{5.6.4b}$$

and

$$\mathscr{P} \cong i\wp\Omega_R\tau. \tag{5.6.5}$$

Now, using Eq. (5.4.23), for the atom initially in the excited state we have

$$\frac{1}{c}\frac{\partial \mathscr{E}}{\partial t} = \frac{k}{2\epsilon_0}\frac{\wp^2}{\hbar}\mathscr{E}\tau, \tag{5.6.6}$$

where we have neglected the cavity loss. It follows from Eq. (5.6.6) that the change in the electric field during the time τ is

$$\Delta\mathscr{E} \cong \frac{ck}{2\epsilon_0}\frac{\wp^2}{\hbar}\mathscr{E}\tau^2, \tag{5.6.7}$$

i.e., the incident field experiences gain.

Likewise for the atom initially in the ground state, we have

$$\frac{1}{c}\frac{\partial \mathscr{E}}{\partial t} = -\frac{k}{2\epsilon_0}\frac{\wp^2}{\hbar}\mathscr{E}\tau, \tag{5.6.8}$$

and therefore

$$\Delta\mathscr{E} \cong -\frac{ck}{2\epsilon_0}\frac{\wp^2}{\hbar}\mathscr{E}\tau^2, \tag{5.6.9}$$

i.e., the incident field experiences loss. Thus the atom acts essentially as a tiny oscillating electronic current induced by the incident light field. Attenuation of an incident field is then the result of the radiation from this current interfering destructively with the incident light (see Fig. 5.6). This simple physical picture of stimulated emission and absorption can be expanded to explain more complicated phenomena, e.g., lasing without inversion, as we shall see in Section 7.3.1.

5.7 Time delay spectroscopy

In the previous section, we saw how simple and intuitively pleasing the concepts of stimulated emission and absorption are when treated within the framework of semiclassical radiation theory. As an example

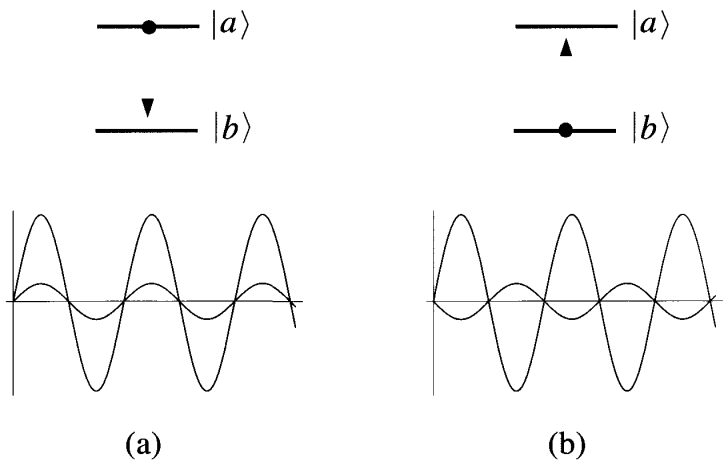


Fig. 5.6
(a) Emission: induced dipole radiation interferes constructively with incident radiation.
(b) Absorption: induced dipole radiation interferes destructively with incident radiation.

of unusual and counter-intuitive physics within the framework of semiclassical theory, we conclude this chapter with a discussion of time delay spectroscopy.

In conventional spectroscopy, the limit of resolution of the energy between two levels $|a\rangle$ and $|b\rangle$ is governed by the sum of the decay rates γ_a and γ_b out of these levels.

In this section we present a spectroscopic technique which provides resolution beyond the natural linewidth. These considerations are based on the fact that in the *transient regime*, the probability for induced transitions in a two-level system interacting with a monochromatic electromagnetic field is not governed by a Lorentzian of width $(\gamma_a+\gamma_b)/2 \equiv \gamma_{ab}$, but rather by $(\gamma_a-\gamma_b)/2 \equiv \delta_{ab}$. The Lorentzian width γ_{ab}, which usually appears in atomic physics, is regained only in the proper limits.

We proceed by considering the experimental situation in which an ensemble of two-level atoms is excited at time $t = t_0$ into the $|b\rangle$ state by some 'instantaneous' excitation mechanism, e.g., a picosecond optical pulse. The excited atoms are then driven by a monochromatic but tunable radiation field.

Consider the level scheme illustrated in Fig. 5.7. There we see an atom with two unstable levels $|a\rangle$ and $|b\rangle$ and a weak field driving the atom from the lower level $|b\rangle$ to the upper level $|a\rangle$. If one includes the lower levels ($|c\rangle$ and $|d\rangle$) to which $|a\rangle$ and $|b\rangle$ decay, this may be considered as a four-level atom. That is, we prepare the atom in level $|b\rangle$ at t_0, drive the atom to level $|a\rangle$, and count the number of atoms accumulating in level $|c\rangle$, starting a finite time t after the atom is

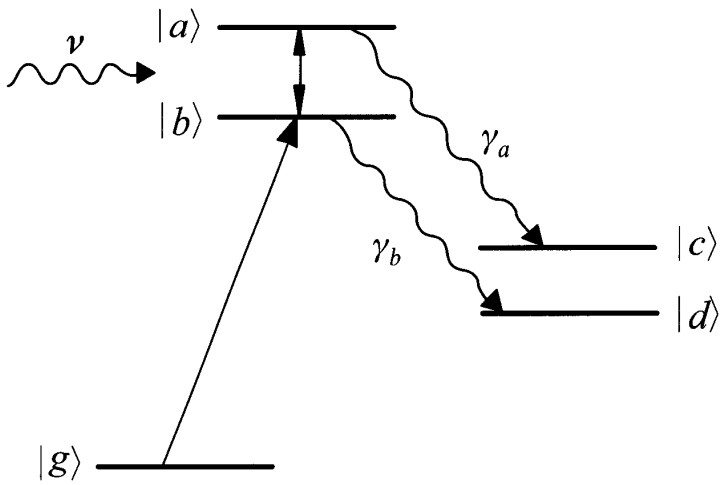


Fig. 5.7
Level diagram indicating excitation of atom from ground state to $|b\rangle$, subsequent interaction with resonant radiation promoting atom from $|b\rangle$ to $|a\rangle$ with atendant decays to states $|d\rangle$ and $|c\rangle$ at rates γ_b and γ_a, respectively.

prepared. The counting rate is measured as a function of the detuning between the laser and atomic frequencies.

We proceed by solving the density matrix equations of motion (5.3.22)–(5.3.24) for $\rho_{aa}(t)$ to lowest nonvanishing order. This yields

$$\rho_{aa}(t) = \frac{\Omega_R^2}{\Delta^2+\delta_{ab}^2}\left[e^{-\gamma_a(t-t_0)}+e^{-\gamma_b(t-t_0)}-2e^{-\gamma_{ab}(t-t_0)}\cos\Delta(t-t_0)\right], \tag{5.7.1}$$

where $\delta_{ab} = (\gamma_a - \gamma_b)/2$, Ω_R is the Rabi frequency of the driven transition and Δ is the detuning between the laser and ω_{ab}. The key point is that the Lorentzian factor in (5.7.1) goes as $\gamma_a-\gamma_b$ not $\gamma_a+\gamma_b$.

Now suppose we count the number of photons emitted when the excited atom makes the $|a\rangle \rightarrow |c\rangle$ transition. This will be equal to the total number of atoms accumulated in level $|c\rangle$ which is determined by the simple rate equation

$$\dot{\rho}_{cc}(t,t_0) = \gamma\rho_{aa}(t,t_0), \tag{5.7.2}$$

where the notation reminds us that the atoms are initially excited at time t_0. Then the total number of spontaneously emitted photons from time t_0 to a time long after the initial excitation to level $|b\rangle$ is given by

$$N(\Delta,t_0) = \eta\gamma_a\int_{t_0}^{\infty}\rho_{aa}(\Delta,t,t_0)dt, \tag{5.7.3}$$

where η is a constant determined by the efficiency of photon detection.

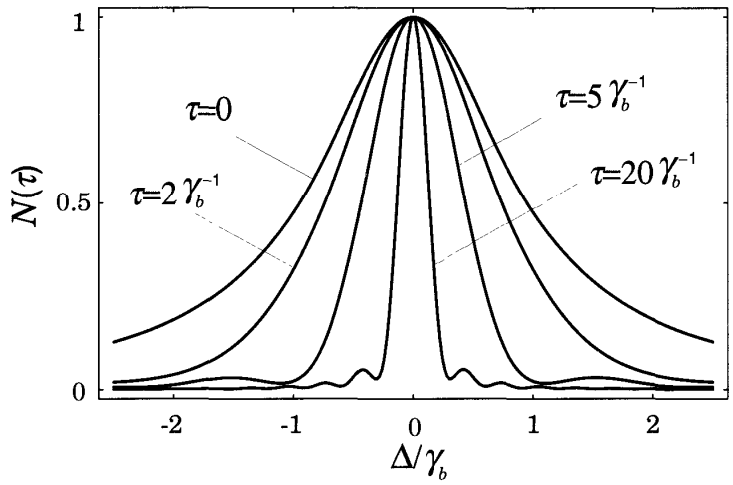


Fig. 5.8 Time delay spectroscopy signal $N(\tau)$ for different τ. The different curves have been normalized for simplicity. In fact the peak heights of the curves corresponding to larger τ are strongly reduced as indicated by Eq. (5.7.6). Nevertheless the line narrowing can be useful as discussed by Figger and Walther (1974).

Inserting (5.7.1) into (5.7.3) we find

$$N(\Delta, t_0) = \frac{\eta\gamma_a\Omega_R^2}{\Delta^2+\gamma_{ab}^2}\frac{2\gamma_{ab}}{\gamma_a\gamma_b}. \tag{5.7.4}$$

That is, when we carry out the above procedure, collecting the $|a\rangle \rightarrow |c\rangle$ photons from t_0 onwards we regain the usual Lorentzian of width γ_{ab}. This is reassuring since in most experiments it is indeed γ_{ab} that governs the resolution of our experiments.

However, let us now wait for a time $t_0 + \tau$ before accepting any counts. That is let us measure

$$N(\Delta, t_0+\tau) = \eta\gamma_a \int_{t_0+\tau}^{\infty} \rho_{aa}(\Delta, t, t_0)dt. \tag{5.7.5}$$

Inserting (5.7.1) into (5.7.5) we now find*

$$N(\Delta, t_o+\tau) = \frac{\eta\gamma_a\Omega_R^2}{\Delta^2+\delta_{ab}^2}\left[\frac{\exp(-\gamma_a\tau)}{\gamma_a}+\frac{\exp(-\gamma_b\tau)}{\gamma_b}\right.$$
$$\left.+\frac{2\exp(-\gamma_{ab}\tau)}{\Delta^2+\gamma_{ab}^2}(\Delta\sin\Delta\tau-\gamma_{ab}\cos\Delta\tau)\right]. \tag{5.7.6}$$

The point is clear. When we delay observation we find a line narrowing as is seen by comparing Eqs. (5.7.4) and (5.7.6). Equation (5.7.6) is plotted for various values of time delay in Fig. 5.8.

We conclude by noting that, as pointed out explicitly by Figger and Walther, the line narrowing in time delay spectroscopy provides a

* See Meystre, Scully, and Walther [1980].

high spectral *resolution* in the sense that we can separate closely spaced lines. However, this higher resolution does not always lead to a higher experimental *accuracy* in the final result for the atomic transition frequencies ω_{ab}. The reason for this is the exponential damping of the signal with the time delay τ by means of the prefactors $\exp(-\gamma_a\tau)$ and $\exp(-\gamma_b\tau)$ in Eq. (5.7.6) which decrease the signal. We will return to the question of enhancing spectroscopic resolution in later chapters, e.g., in Section 21.7.

5.A Equivalence of the r · E and the p · A interaction Hamiltonians

In Section 5.1 we noted that in the radiation gauge (R-gauge) and in the dipole approximation $(\mathbf{A}(\mathbf{r},t), U(\mathbf{r},t)) \equiv (\mathbf{A}(t), 0)$, the gauge transformation

$$\chi(\mathbf{r},t) = -\frac{e}{\hbar}\mathbf{A}(t)\cdot\mathbf{r} \tag{5.A.1}$$

yields the gauge $(0, -\mathbf{E}(t)\cdot\mathbf{r})$. We observe that the gauge $(0, -\mathbf{E}(t)\cdot\mathbf{r})$ leads to the electric–dipole interaction $\mathscr{H}$ (Eq. (5.1.19)), and thus we call it the electric field gauge (E-gauge). The two Hamiltonians $\mathscr{H}$ (Eq. (5.1.19)) and $\mathscr{H}'$ (Eq. (5.1.21)) are therefore related via the gauge transformation (5.A.1). A gauge transformation requires a transformation of the potentials according to Eqs. (5.1.6) and (5.1.7) and of the wave functions according to Eq. (5.1.4). Nonidentical, *wrong* results are obtained for physically measurable quantities in different gauges if only one of these two transformations is carried out. We will discuss how we have to handle the wave functions in the two different gauges in order to obtain gauge-invariant physical predictions. Before this, however, let us briefly discuss some examples of physical quantities.

5.A.1 Form-invariant physical quantities

A form-invariant physical quantity is defined as a quantity whose corresponding operator $G_\chi = G(A_\chi, U_\chi)$ is form invariant under a unitary transformation $T(\mathbf{r},t) = \exp[i\chi(\mathbf{r},t)]$, i.e.,

$$G_{\chi'} = TG_\chi T^\dagger, \tag{5.A.2}$$

where the wave function in the gauge χ is transformed to the gauge χ' by the unitary transformation

$$\psi_{\chi'}(\mathbf{r},t) = T(\mathbf{r},t)\psi_\chi(\mathbf{r},t). \tag{5.A.3}$$

The difference between physical and nonphysical quantities lies in the gauge invariance of the eigenvalues. The eigenvalues of a physical quantity are identical in all gauges, whereas the eigenvalues of nonphysical quantities depend on the chosen gauge. In order to show this, we denote the eigenvalues and eigenstates of the operator G_χ by g_n and $|\xi_{\chi,n}\rangle$, respectively:

$$G_\chi|\xi_{\chi,n}\rangle = g_n|\xi_{\chi,n}\rangle. \tag{5.A.4}$$

Only for physical quantities are the eigenvalues g_n gauge invariant, i.e.,

$$\begin{aligned} G_{\chi'}|\xi_{\chi',n}\rangle &= TG_\chi T^\dagger T|\xi_{\chi,n}\rangle \\ &= Tg_n|\xi_{\chi,n}\rangle \\ &= g_n|\xi_{\chi',n}\rangle. \end{aligned} \tag{5.A.5}$$

Hence, nonphysical quantities can only be used as calculational tools.

We next consider some examples of physical and nonphysical quantities. The starting point for these considerations is the fact that the operators $\mathbf{r}$ and $\mathbf{p}$ ($\mathbf{p} = -i\hbar\nabla$), associated with the position and the canonical momentum of the particle, are the same in all gauges, by which we mean that $\mathbf{p}$ is represented by $-i\hbar\nabla$ in all gauges. This ensures that, in any gauge, the commutation relation $[\mathbf{r}_j, \mathbf{p}_k] = i\hbar\delta_{jk}$ is satisfied. With this rule the operator for the mechanical momentum,

$$\pi_\chi = \mathbf{p} - e\mathbf{A}_\chi(\mathbf{r}, t), \tag{5.A.6}$$

is a physical, measurable quantity since

$$\begin{aligned} T\pi_\chi T^\dagger &= T[\mathbf{p} - e\mathbf{A}_\chi(\mathbf{r}, t)]T^\dagger \\ &= \mathbf{p} - e\mathbf{A}_\chi - \hbar\nabla\chi \\ &= \mathbf{p} - e\mathbf{A}_{\chi'} \\ &= \pi_{\chi'}. \end{aligned} \tag{5.A.7}$$

Similarly, the instantaneous energy operator of the system, consisting of the kinetic energy and the static potential (normally the atomic binding potential)

$$\mathscr{E}_\chi = \frac{1}{2m}[\mathbf{p} - e\mathbf{A}_\chi(\mathbf{r}, t)]^2 + V(r), \tag{5.A.8}$$

represents a physical quantity as well as any other operator which is only a function of other physical quantities like π_χ.

On the other hand, the canonical momentum $\mathbf{p}$ is not a physical quantity since

$$T\mathbf{p}T^\dagger = \mathbf{p} - \hbar\nabla\chi \neq \mathbf{p}. \tag{5.A.9}$$

In a similar way, the operator $\mathscr{H}_0 = p^2/2m$ (which does not depend on potentials) is not a physical quantity because

$$T\mathscr{H}_0T^\dagger = \mathscr{H}_0 - \frac{\hbar}{2m}[\mathbf{p}\cdot\nabla\chi + (\nabla\chi)\cdot\mathbf{p}] + \frac{\hbar^2}{2m}(\nabla\chi)^2 \neq \mathscr{H}_0. \tag{5.A.10}$$

In general, any operator which is a function of nonphysical quantities alone, like the canonical momentum $\mathbf{p}$ or the vector or the scalar potentials $\mathbf{A}_\chi$ or U_χ, represents a nonphysical quantity. The total Hamiltonian

$$\mathscr{H}_\chi = \frac{1}{2m}[\mathbf{p} - e\mathbf{A}_\chi(\mathbf{r}, t)]^2 + eU_\chi(\mathbf{r}, t) + V(r) \tag{5.A.11}$$

is also a nonphysical quantity, since it depends on the scalar potential U_χ.

We therefore conclude that the time evolution of a physical system is determined by Hamiltonians such as $\mathscr{H}_\chi$ or $\mathscr{H}_0$, which in general are not observable quantities. The physical quantities are, for example, the mechanical momentum and the instantaneous energy of the system.

5.A.2 *Transition probabilities in a two-level atom*

In this subsection we restrict the discussion to the large-wavelength dipole approximation (LWA) in which $\mathbf{A}$ may be considered to be independent of $\mathbf{r}$, i.e., $\mathbf{A}(\mathbf{r}, t) \equiv \mathbf{A}(t)$. Since the energy operator $\mathscr{E}_\chi$ (as given by Eq. (5.A.8)) is time dependent, its eigenstates $|\alpha_\chi(t)\rangle$, where $\alpha = a, b$, and its eigenvalues $E_\alpha = \hbar\omega_\alpha$ are also time dependent in general, namely

$$\mathscr{E}_\chi|\alpha_\chi(t)\rangle = E_\alpha|\alpha_\chi(t)\rangle. \tag{5.A.12}$$

However, in the LWA the eigenvalues of $\mathscr{E}_\chi$ are time independent. This can be seen with the help of the gauge transformation (5.A.1). In the LWA

$$\exp\left[-\frac{ie\mathbf{A}(t)\cdot\mathbf{r}}{\hbar}\right][\mathbf{p} - e\mathbf{A}(t)]^2\exp\left[\frac{ie\mathbf{A}(t)\cdot\mathbf{r}}{\hbar}\right] = p^2, \tag{5.A.13}$$

so that

$$\exp\left[-\frac{ie\mathbf{A}(t)\cdot\mathbf{r}}{\hbar}\right]\mathscr{E}_\chi\exp\left[\frac{ie\mathbf{A}(t)\cdot\mathbf{r}}{\hbar}\right] = \mathscr{H}_0. \tag{5.A.14}$$

The eigenstate $|\alpha_\chi\rangle$ is then related to the eigenstate $|\alpha(t)\rangle$ of $\mathscr{H}_0$ by

$$|\alpha_\chi\rangle = \exp\left[\frac{ie\mathbf{A}_\chi(t)\cdot\mathbf{r}}{\hbar}\right]|\alpha(t)\rangle, \tag{5.A.15}$$

and the eigenvalues E_α of $\mathscr{E}_\chi$ coincide with the time-independent eigenvalues E_α of $\mathscr{H}_0$ since the eigenvalues of physical quantities are gauge independent.

In the E-gauge the unperturbed energy operator $\mathscr{E}_E$ is equal to the *unperturbed* Hamiltonian $\mathscr{H}_0$. Hence the eigenstates of $\mathscr{H}_0$ are also the eigenstates of $\mathscr{E}_E$. Therefore, only in the E-gauge is the wave function expanded in terms of energy eigenstates, and the coefficients $c_\alpha(t)$, where $\alpha = a, b$, in Eqs. (5.2.10) and (5.2.11) are interpreted as probability amplitudes for finding the system in an eigenstate of the observable energy. In any other gauge, $\mathscr{H}_0$ is a nonphysical quantity and its eigenstates are not the energy eigenstates of the system. The expansion coefficients $c_\alpha(t)$ in Eqs. (5.2.10) and (5.2.11) are then the probability amplitudes for finding the system in an eigenstate of $\mathscr{H}_0$. However, if $\mathscr{H}_0$ is a nonphysical quantity, this *probability* is gauge dependent and has to be distinguished from the measurable, gauge-invariant probability of finding the system in an energy eigenstate.

It is, therefore, useful to expand the wave function of the system in terms of eigenstates of the energy operator $\mathscr{E}_\chi$

$$|\psi_\chi(t)\rangle = d_a(t)e^{-i\omega_a t}|a_\chi\rangle + d_b(t)e^{-i\omega_b t}|b_\chi\rangle. \tag{5.A.16}$$

The expansion coefficients d_a and d_b then coincide with the probability amplitudes for transitions of the system to the eigenstates $|a_\chi\rangle$ and $|b_\chi\rangle$, respectively of the energy operator $\mathscr{E}_\chi$ with energies $\hbar\omega_a$ and $\hbar\omega_b$:

$$d_a(t) = \langle a_\chi|\psi_\chi(t)\rangle e^{i\omega_a t}, \tag{5.A.17}$$

$$d_b(t) = \langle b_\chi|\psi_\chi(t)\rangle e^{i\omega_b t}. \tag{5.A.18}$$

We will now show explicitly that these amplitudes are gauge invariant.

In the E-gauge, the probability amplitude $d_a(t)$ is given by

$$d_a^E(t) = \langle a|U_0(t)U_I^{(1)}(t)|b\rangle e^{i\omega_a t} \tag{5.A.19}$$

and, in the R-gauge, by

$$d_a^R(t) = \langle a|\exp\left[-\frac{ie}{\hbar}\mathbf{A}(t)\cdot\mathbf{r}\right] U_0(t)U_I^{(2)}(t)\exp\left[\frac{ie}{\hbar}\mathbf{A}(0)\cdot\mathbf{r}\right]|b\rangle e^{i\omega_a t}, \tag{5.A.20}$$

where $U_0(t) = \exp(-i\mathscr{H}_0 t/\hbar)$ and

$$U_I^{(i)}(t) = \mathscr{T}\exp\left[-\frac{i}{\hbar}\int_0^t d\tau U_0^\dagger(\tau)\mathscr{H}_i(\tau)U_0(\tau)\right]. \tag{5.A.21}$$

Here, we assume that the atom is initially in the ground state $|b\rangle$. Similar expressions exist for the amplitudes $d_b^E(t)$ and $d_b^R(t)$. In the first order of perturbation theory, the time-evolution operator $U_I^{(1)}$ becomes

$$U_I^{(1)} = 1 - \frac{i}{\hbar}\int_0^t d\tau U_0^\dagger(\tau)\mathscr{H}_1 U_0(\tau), \tag{5.A.22}$$

and the probability amplitude of the excited state in the E-gauge takes the form

$$\begin{aligned} d_a^E(t) &= -\frac{i}{\hbar}\langle a|\int_0^t d\tau U_0^\dagger(\tau)\mathscr{H}_1 U_0(\tau)|b\rangle \\ &= \frac{ie}{2\hbar}\mathscr{E}\cdot\langle a|\mathbf{r}|b\rangle\int_0^t d\tau e^{i(\omega-\nu)\tau} \\ &= \frac{e}{2\hbar}\mathscr{E}\cdot\mathbf{r}_{ab}\frac{e^{i\Delta t}-1}{\Delta}. \end{aligned} \tag{5.A.23}$$

This result is now compared to the corresponding result in the R-gauge. In first-order perturbation theory,

$$\begin{aligned} d_a^R(t) = \langle a|\left[1-\frac{ie}{\hbar}\mathbf{A}(t)\cdot\mathbf{r}\right] U_0(t)\left[1-\frac{i}{\hbar}\int_0^t d\tau U_0^\dagger(\tau)\mathscr{H}_2 U_0(\tau)\right] \\ \times\left[1+\frac{ie}{\hbar}\mathbf{A}(0)\cdot\mathbf{r}\right]|b\rangle\exp(i\omega_a t). \end{aligned} \tag{5.A.24}$$

Using (5.1.26) and (5.2.32) and $A(t) = \frac{1}{2}\mathscr{A}e^{-i\nu t}$, to lowest order in $\mathscr{A}$, yields

$$d_a^R(t) = -\frac{ie}{\hbar}\frac{\mathscr{A}}{2}\left[-\mathbf{r}_{ab}e^{-i(\omega-\nu)t} + \mathbf{p}_{ab}\frac{e^{-i(\omega-\nu)t}-1}{i(\omega-\nu)} + \mathbf{r}_{ab}\right].$$

From (5.1.30) we have $\mathbf{p}_{ab} = +im\omega\mathbf{r}_{ab}$ and defining $\mathscr{E} = i\nu\mathscr{A}$ yields

$$d_a^R(t) = \frac{e}{2\hbar}\mathscr{E}\cdot\mathbf{r}_{ab}\frac{e^{i\Delta t}-1}{\Delta}. \tag{5.A.25}$$

Thus, the amplitudes $d_a^E(t)$ and $d_a^R(t)$ are seen to be identical. This resolves* the apparent contradiction pointed out at the end of Section 5.1.

* The present treatment is oversimplified in that the effects of atomic decay are not included. For the more general case, see Lamb, Schlicher and Scully [1987].

5.B Vector model of the density matrix

A physical picture of the density matrix is provided by reducing Eqs. (5.3.22), (5.3.23), and (5.3.30) into a form equivalent to the Bloch equations appearing in nuclear magnetic resonance. The present problem of a two-level atom interacting with an electromagnetic field is similar to that of a spin-1/2 magnetic dipole undergoing precession in a magnetic field. This formal similarity has led to the prediction, observation, and physical understanding of a number of phenomena associated with coherent pulse propagation in a system of two-level atoms.

We introduce the real quantities

$$R_1 = \rho_{ab}e^{ivt} + \text{c.c.}, \tag{5.B.1}$$

$$R_2 = i\rho_{ab}e^{ivt} + \text{c.c.}, \tag{5.B.2}$$

$$R_3 = \rho_{aa} - \rho_{bb}. \tag{5.B.3}$$

These quantities are components of the vector **R**, given by

$$\mathbf{R} = R_1\hat{\mathbf{e}}_1 + R_2\hat{\mathbf{e}}_2 + R_3\hat{\mathbf{e}}_3. \tag{5.B.4}$$

where $\hat{\mathbf{e}}_1$, $\hat{\mathbf{e}}_2$, and $\hat{\mathbf{e}}_3$ form a set of mutually perpendicular unit vectors. Here, R_1 and R_2 represent the atom's dipole moment, and R_3 is the population difference between the levels $|a\rangle$ and $|b\rangle$.

It follows from Eqs. (5.3.22), (5.3.23), and (5.3.30) that, in the rotating-wave approximation, (with $\phi = 0$)

$$\dot{R}_1 = -\Delta R_2 - \frac{1}{T_2}R_1, \tag{5.B.5}$$

$$\dot{R}_2 = \Delta R_1 - \frac{1}{T_2}R_2 + \Omega_R R_3, \tag{5.B.6}$$

$$\dot{R}_3 = -\frac{1}{T_1}R_3 - \Omega_R R_2, \tag{5.B.7}$$

where we have assumed $\gamma_a = \gamma_b = 1/T_1$ and $\gamma = 1/T_2$. The quantities T_1 and T_2 are called the longitudinal and the transverse relaxation times, respectively, in analogy with the corresponding quantities in the Bloch equations. Equations (5.B.5)–(5.B.7) are referred to as the optical Bloch equations.

When $T_1 = T_2$, these equations can be written in the following compact form

$$\dot{\mathbf{R}} = -\frac{1}{T_1}\mathbf{R} + \mathbf{R} \times \boldsymbol{\Omega}, \tag{5.B.8}$$

where the effective field is given by

$$\boldsymbol{\Omega} = \Omega_R\hat{\mathbf{e}}_1 - \Delta\hat{\mathbf{e}}_3. \tag{5.B.9}$$

The time dependence of **R**, as given by Eq. (5.B.8), is well known from classical mechanics. The vector **R** precesses clockwise about the effective field **Ω** with diminishing amplitude. The precessions for resonance and slightly off resonance are depicted in Fig. 5.9. Physically **R** pointing along $\hat{\mathbf{e}}_3$ ($R_3 = 1$, $R_1 = R_2 = 0$) represents a system in its upper level, $\rho_{aa} = 1$, $\rho_{bb} = 0$. Similarly, **R** pointing along $-\hat{\mathbf{e}}_3$ represents a system in its lower level.

5.C Quasimode laser physics based on the modes of the universe*

Most laser theories, e.g., that of Section 5.5, describe the electromagnetic field in terms of a discrete set of quasimodes of the laser cavity, each of which has a finite quality factor Q. In the present section, this theory is generalized for a laser with a cavity modeled by a semitransparent wall as one of the mirrors so that there are now many modes of the 'universe' corresponding to each quasimode. Here we show that the normal modes of the universe associated with a single 'mode' may, under proper conditions, lock together and the δ-function laser lineshape may be regained.

We consider the normal modes for a combined system of a laser cavity coupled to the outside world. We represent the 'universe' by a much larger cavity having perfectly reflecting walls. A simple one-dimensional model which carries the essential features of such a combined system is illustrated in Fig. 5.10. The mirrors at $z = L$ and $-L_0$ are completely reflective, while the one at $z = 0$ is semitransparent. Region 1 corresponds to a laser cavity and region 2 to the rest of the universe.

We represent a semitransparent mirror by a very thin plate with a very large dielectric constant. As an idealization of such a mirror we choose the dielectric constant around $z = 0$ to be

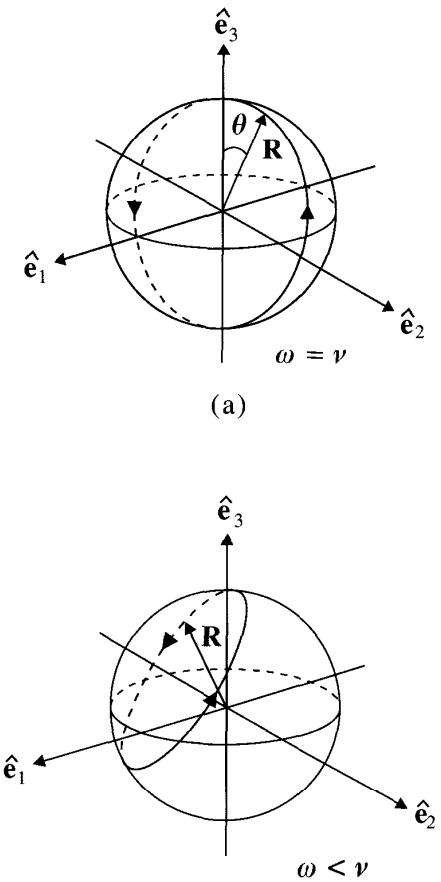
$$\epsilon(z) = \epsilon_0[1 + \eta\delta(z)], \tag{5.C.1}$$

where η is a parameter with the dimension of length which determines the transparency of this plate.

The normal mode functions of this system can be obtained by solving Maxwell's equations with the proper boundary conditions (see Problem 5.6). For those normal modes having frequency $v_k(= ck)$ close

* For further reading, see Lang, Scully and Lamb [1973].

Fig. 5.9
Precession of Bloch vector **R** about the effective field **Ω** for
(a) $\Delta = 0$ and
(b) $\Delta \neq 0$.



to a 'resonant' frequency $\nu_0 (= ck_0)$, the eigenfunctions of the entire cavity are

$$U_k(z) = \begin{cases} M_k \sin k(z-L) & (z>0), \\ \xi_k \sin k(z+L) & (z<0), \end{cases} \tag{5.C.2}$$

where ξ_k is a phase factor which alternates between 1 and -1 as k increases from one value to the next. The coefficients M_k in (5.C.2) are

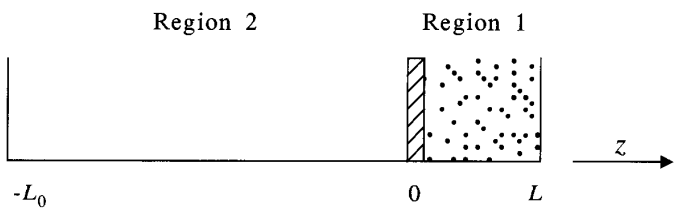


Fig. 5.10
Leaky cavity bounded by a perfect mirror at $z = L$ and a semitransparent mirror at $z = 0$. The auxiliary cavity which, along with the leaky cavity, constitutes the universe, is bounded by a perfect mirror at $z = -L_0$ $(L_0 \to \infty)$ and the mirror at $z = 0$.

$$M_k = \frac{\mathscr{C}\Lambda}{2}\left[(\nu_k - \nu_0)^2 + \frac{\mathscr{C}^2}{4}\right]^{-1/2}, \tag{5.C.3}$$

where $\mathscr{C}$ is the bandwidth associated with the mirror transparency and is given by

$$\mathscr{C} = 2c/\eta^2 k_0^2 L = 2c/\Lambda^2 L, \tag{5.C.4}$$

with

$$\Lambda = \eta\nu_0/c = \eta k_0, \tag{5.C.5}$$

and the frequency ν_0 of the nth quasimode is given by

$$\nu_0 = ck_0 = (n\pi + 1/\Lambda)c/L. \tag{5.C.6}$$

An arbitrary undriven field in the entire cavity can be expressed as the positive frequency part of the field

$$E^{(+)}(z,t) = \sum_k \mathscr{E}_k(0)U_k(z)e^{-i\nu t} = \sum_k \mathscr{E}_k(t)U_k(z), \tag{5.C.7}$$

which is to be understood as a sum over modes of the large cavity, i.e., 'the universe'.

We now demonstrate that the semitransparency of the mirror leads to a damping of free oscillations in the laser cavity. Let us assume that, at $t = 0$, the laser cavity (region 1) contains a field of the form

$$E^{(+)}(z,0) = |\mathscr{E}_0|e^{-i\varphi}\sin k_0(z-L), \tag{5.C.8}$$

whereas no field exists outside the cavity, i.e., in region 2. The coefficients $\mathscr{E}_k(0)$ for this case are obtained by multiplying (5.C.8) by $U_k(z)$ defined in (5.C.2) and integrating over z. We find

$$\mathscr{E}_k(t) = (|\mathscr{E}_0|M_k L/L_0)e^{-i(\nu_k t+\varphi)}. \tag{5.C.9}$$

Therefore at later times, $t > 0$,

$$E(z,t) = (|\mathscr{E}_0|L/L_0)\sum_k M_k U_k(z)e^{-i(\nu_k t+\varphi)}. \tag{5.C.10}$$

The summation can be approximated by an integral if the frequency separation between the normal modes is small compared to $\mathscr{C}$. Carrying out the integration over k in (5.C.10), the explicit form of $E(z,t)$ in the maser cavity turns out to be

$$E^{(+)}(z,0) = |\mathscr{E}_0| \sin k_0(z-L) e^{-i(\nu_k t+\varphi)-\mathscr{C}t/2}. \tag{5.C.11}$$

Equation (5.C.11) indicates that the field localized in the maser cavity decays exponentially owing to leakage through the mirror at a rate $\mathscr{C}/2$.

Problems

5.1 Show that the Schrödinger equation (5.1.5) is invariant under local gauge transformations (5.1.4), (5.1.6), and (5.1.7).

5.2 The finite lifetime of the atomic levels can be described by adding phenomenological decay terms to the probability amplitude equations (5.2.12) and (5.2.13):

$$\dot{c}_a = -\frac{\gamma}{2}c_a + \frac{i\Omega_R}{2}e^{-i\phi}c_b,$$
$$\dot{c}_b = -\frac{\gamma}{2}c_b + \frac{i\Omega_R}{2}e^{i\phi}c_a,$$

where γ is the decay constant and $\omega = \nu$. For an atom initially in the state $|a\rangle$, show that the inversion at time t is

$$W(t) = e^{-\gamma t}\cos(\Omega_R t).$$

5.3 Find the solution of Eq. (5.B.8) (with $T_1 \to \infty$):

$$\dot{\mathbf{R}} = \mathbf{R} \times \boldsymbol{\Omega}$$

for $\mathbf{R}(0) = 0$. Give a physical interpretation of this solution.

5.4 Show that, in general,

$$\mathrm{Tr}(\rho^2) \leq 1,$$

where the equality is valid only for a pure state.

5.5 Consider a three-level atom interacting with a classical field of frequency ν. The transitions $|a\rangle \to |b\rangle$ and $|b\rangle \to |c\rangle$ are allowed whereas the transition $|a\rangle \to |c\rangle$ is forbidden. It is also assumed that $\omega_a - \omega_b = \omega_b - \omega_c = \nu$. Assuming the atom to be initially in level $|c\rangle$, find the probabilities for the atom to be in levels $|a\rangle$ and $|c\rangle$ after making the rotating-wave approximation.

5.6 The electromagnetic field in the entire cavity (region 1 and region 2 in Fig. 5.10) is governed by the Maxwell wave equation

$$\frac{\partial^2 E}{\partial z^2} - \mu_0\epsilon_0[1+\eta\delta(z)]\frac{\partial^2 E}{\partial t^2} = 0,$$

where E can be written as

$$E = U_k(z)e^{-i\nu_k t}.$$

(a) Find $U_k(z)$ in the form (5.C.2) and prove that

$$\frac{M_k^2}{\xi_k^2} = \frac{\tan^2 kL + 1}{\tan^2 kL + (\Lambda \tan kL - 1)^2},$$

where Λ is given by Eq. (5.C.5). Derive Eq. (5.C.3).

(b) Show that

$$(\nu_k - \nu_{k'})^2 \int_{-L_0}^{L} dz U_k(z)U_{k'}(z)\epsilon(z) = 0,$$

where $\epsilon(z) = \epsilon_0[1+\eta\delta(z)]$. (Hint: see R. Lang, M. O. Scully, and W. E. Lamb, Jr., *Phys. Rev. A* **7**, 1788 (1973).)

5.7 The $m = +1$ level of Fig. 5.3 is weakly coupled to the $\psi_b(r)$ level by the left-circular polarized light of Eq. (5.2.55) via counter rotating terms. Note that in such a case ($m = 0$ to $m = +1$) we normally rule out such coupling on the grounds that right-circularly polarized light is needed for the $m = 0$ to $m = +1$ transition.

(a) Show that if we define

$$\psi_{a'}(\mathbf{r}) = \psi_{n=2,l=1,m_l=+1}(\mathbf{r}) = \eta(x+iy)e^{-r/2a_0},$$

where η is the uninteresting constant $[\sqrt{64\pi a_0^3}a_0]^{-1}$, then

$$\mathscr{V}_{a'b}(t) = -e\mathscr{E}\int d\mathbf{r}\psi_a^*(\mathbf{r})\mathbf{r}\cdot(\hat{x}\cos\nu t - \hat{y}\sin\nu t)\psi_b(\mathbf{r})e^{i\omega_{a'b}t} = -\wp\mathscr{E}e^{i(\omega_{a'b}+\nu)t}.$$

(b) Show, by specific example, that the counter terms associated with the $|b\rangle \rightarrow |a'\rangle$ transitions, which go like $[\omega_{a'b}+\nu]^{-1}$ can be much smaller than the usual counter terms $[\omega_{ab}+\nu]^{-1}$. Hint: consider a Rydberg atom in which $\omega_{ab} = \omega_{a'b} \cong 10^9$Hz. If we now apply a field of around 10^4 Gauss, we could arrange for the Zeeman shifted $\omega_{ab} \sim 10^3$Hz while $\omega_{a'b} \sim 10^{10}$Hz.

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CHAPTER 6

Atom–field interaction – quantum theory

In the preceding chapters concerning the interaction of a radiation field with matter, we assumed the field to be classical. In many situations this assumption is valid. There are, however, many instances where a classical field fails to explain experimentally observed results and a quantized description of the field is required. This is, for example, true of spontaneous emission in an atomic system which was described phenomenologically in Chapter 5. For a rigorous treatment of the atomic level decay in free space, we need to consider the interaction of the atom with the vacuum modes of the universe. Even in the simplest system involving the interaction of a single-mode radiation field with a single two-level atom, the predictions for the dynamics of the atom are quite different in the semiclassical theory and the fully quantum theory. In the absence of the decay process, the semiclassical theory predicts Rabi oscillations for the atomic inversion whereas the quantum theory predicts certain *collapse* and *revival* phenomena due to the quantum aspects of the field. These interesting quantum field theoretical predictions have been experimentally verified.

In this chapter we discuss the interaction of the quantized radiation field with the two-level atomic system described by a Hamiltonian in the dipole and the rotating-wave approximations. For a single-mode field it reduces to a particularly simple form. This is a very interesting Hamiltonian in quantum optics for several reasons. First, it can be solved exactly for arbitrary coupling constants and exhibits some true quantum dynamical effects such as collapse followed by periodic revivals of the atomic inversion. Second, it provides the simplest illustration of spontaneous emission and thus explains the effects of various kinds of quantum statistics of the field in more complicated systems such as a micromaser and a laser, which we shall study in later chapters. Third, and perhaps most importantly,* it has become possible to realize it experimentally through the spectacular advances in the development of high-Q microwave cavities.

The spontaneous decay of an atomic level is treated by considering the interaction of the two-level atom with the modes of the universe in the vacuum state. We examine the state of the field that is generated in the process of emission of a quantum of energy equal to the energy difference between the atomic levels. Such a state may be regarded as a single-photon state.

6.1 Atom–field interaction Hamiltonian

The interaction of a radiation field $\mathbf{E}$ with a single-electron atom can be described by the following Hamiltonian in the dipole approximation:

$$\mathscr{H} = \mathscr{H}_A + \mathscr{H}_F - e\mathbf{r}\cdot\mathbf{E}. \tag{6.1.1}$$

Here $\mathscr{H}_A$ and $\mathscr{H}_F$ are the energies of the atom and the radiation field, respectively, in the absence of the interaction, and $\mathbf{r}$ is the position vector of the electron. In the dipole approximation, the field is assumed to be uniform over the whole atom.

The energy of the free field $\mathscr{H}_F$ is given in terms of the creation and destruction operators by

$$\mathscr{H}_F = \sum_{\mathbf{k}} \hbar\nu_k \left(a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}} + \frac{1}{2}\right). \tag{6.1.2}$$

We can express $\mathscr{H}_A$ and $e\mathbf{r}$ in terms of the atom transition operators

$$\sigma_{ij} = |i\rangle\langle j|. \tag{6.1.3}$$

As before $\{|i\rangle\}$ represents a complete set of atomic energy eigenstates, i.e., $\sum_i |i\rangle\langle i| = 1$. It then follows from the eigenvalue equation $\mathscr{H}_A|i\rangle = E_i|i\rangle$ that

$$\mathscr{H}_A = \sum_i E_i|i\rangle\langle i| = \sum_i E_i\sigma_{ii}. \tag{6.1.4}$$

Also

$$e\mathbf{r} = \sum_{i,j} e|i\rangle\langle i|\mathbf{r}|j\rangle\langle j| = \sum_{i,j} \wp_{ij}\sigma_{ij}, \tag{6.1.5}$$

* Especially the micromaser of H. Walther and coworkers as discussed in Chapter 13. See also the Physics Today article by Haroche and Kleppner [1989] which presents the physics of cavity QED very nicely.

where $\wp_{ij} = e\langle i|\mathbf{r}|j\rangle$ is the electric-dipole transition matrix element. The electric field operator is evaluated in the dipole approximation at the position of the point atom. It follows from Eq. (1.1.27) that, for the atom at the origin, we have

$$\mathbf{E} = \sum_{\mathbf{k}} \hat{\epsilon}_{\mathbf{k}} \mathscr{E}_{\mathbf{k}} (a_{\mathbf{k}} + a_{\mathbf{k}}^\dagger), \tag{6.1.6}$$

where $\mathscr{E}_{\mathbf{k}} = (\hbar \nu_k / 2\epsilon_0 V)^{1/2}$. Here, for simplicity, we have taken a linear polarization basis and the polarization unit vectors to be real.

It now follows, on substituting for $\mathscr{H}_F, \mathscr{H}_A, e\mathbf{r}$, and $\mathbf{E}$ from Eqs. (6.1.2), (6.1.4), (6.1.5), and (6.1.6) into Eq. (6.1.1), that

$$\mathscr{H} = \sum_{\mathbf{k}} \hbar \nu_k a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + \sum_i E_i \sigma_{ii} + \hbar \sum_{i,j} \sum_{\mathbf{k}} g_{\mathbf{k}}^{ij} \sigma_{ij} (a_{\mathbf{k}} + a_{\mathbf{k}}^\dagger), \tag{6.1.7}$$

where

$$g_{\mathbf{k}}^{ij} = -\frac{\wp_{ij} \cdot \hat{\epsilon}_{\mathbf{k}} \mathscr{E}_{\mathbf{k}}}{\hbar}. \tag{6.1.8}$$

In Eq. (6.1.7), we have omitted the zero-point energy from the first term. For the sake of simplicity, we will assume $\wp_{ij}$ to be real throughout this chapter.

We now proceed to the case of a two-level atom. For $\wp_{ab} = \wp_{ba}$, we write

$$g_{\mathbf{k}} = g_{\mathbf{k}}^{ab} = g_{\mathbf{k}}^{ba}. \tag{6.1.9}$$

The following form of the Hamiltonian is obtained

$$\mathscr{H} = \sum_{\mathbf{k}} \hbar \nu_k a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + (E_a \sigma_{aa} + E_b \sigma_{bb}) + \hbar \sum_{\mathbf{k}} g_{\mathbf{k}} (\sigma_{ab} + \sigma_{ba})(a_{\mathbf{k}} + a_{\mathbf{k}}^\dagger). \tag{6.1.10}$$

The second term in Eq. (6.1.10) can be rewritten as

$$E_a \sigma_{aa} + E_b \sigma_{bb} = \frac{1}{2} \hbar\omega (\sigma_{aa} - \sigma_{bb}) + \frac{1}{2}(E_a + E_b), \tag{6.1.11}$$

where we use $(E_a - E_b) = \hbar\omega$ and $\sigma_{aa} + \sigma_{bb} = 1$. The constant energy term $(E_a + E_b)/2$ can be ignored. If we use the notation

$$\sigma_z = \sigma_{aa} - \sigma_{bb} = |a\rangle\langle a| - |b\rangle\langle b|, \tag{6.1.12}$$

$$\sigma_+ = \sigma_{ab} = |a\rangle\langle b|, \tag{6.1.13}$$

$$\sigma_- = \sigma_{ba} = |b\rangle\langle a|, \tag{6.1.14}$$

the Hamiltonian (6.1.10) takes the form

$$\mathscr{H} = \sum_{\mathbf{k}} \hbar \nu_k a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + \frac{1}{2} \hbar\omega \sigma_z + \hbar \sum_{\mathbf{k}} g_{\mathbf{k}} (\sigma_+ + \sigma_-)(a_{\mathbf{k}} + a_{\mathbf{k}}^\dagger). \tag{6.1.15}$$

It follows from the identity

$$[\sigma_{ij}, \sigma_{kl}] = \sigma_{il}\delta_{jk} - \sigma_{kj}\delta_{il}, \tag{6.1.16}$$

that σ_+, σ_-, and σ_z satisfy the spin-1/2 algebra of the Pauli matrices, i.e.,

$$[\sigma_-, \sigma_+] = -\sigma_z, \tag{6.1.17}$$

$$[\sigma_-, \sigma_z] = 2\sigma_-. \tag{6.1.18}$$

In the matrix notation, σ_-, σ_+, and σ_z are given by

$$\sigma_- = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix}, \quad \sigma_+ = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}, \quad \sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}. \tag{6.1.19}$$

The σ_- operator takes an atom in the upper state into the lower state whereas σ_+ takes an atom in the lower state into the upper state.

The interaction energy in Eq. (6.1.15) consists of four terms. The term $a_{\mathbf{k}}^\dagger \sigma_-$ describes the process in which the atom is taken from the upper state into the lower state and a photon of mode $\mathbf{k}$ is created. The term $a_{\mathbf{k}} \sigma_+$ describes the opposite process. The energy is conserved in both the processes. The term $a_{\mathbf{k}} \sigma_-$ describes the process in which the atom makes a transition from the upper to the lower level and a photon is annihilated, resulting in the loss of approximately $2\hbar\omega$ in energy. Similarly $a_{\mathbf{k}}^\dagger \sigma_+$ results in the gain of $2\hbar\omega$. Dropping the energy nonconserving terms corresponds to the rotating-wave approximation. The resulting simplified Hamiltonian is

$$\mathscr{H} = \sum_{\mathbf{k}} \hbar \nu_k a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + \frac{1}{2} \hbar\omega \sigma_z + \hbar \sum_{\mathbf{k}} g_{\mathbf{k}} (\sigma_+ a_{\mathbf{k}} + a_{\mathbf{k}}^\dagger \sigma_-). \tag{6.1.20}$$

This form of the Hamiltonian describing the interaction of a single two-level atom with a multi-mode field is the starting point of many calculations in the field of quantum optics.

6.2 Interaction of a single two-level atom with a single-mode field

It follows from Eq. (6.1.20) that the interaction of a single-mode quantized field of frequency ν with a single two-level atom is described

by the Hamiltonian

$$\mathscr{H} = \mathscr{H}_0 + \mathscr{H}_1, \tag{6.2.1}$$

where

$$\mathscr{H}_0 = \hbar v a^\dagger a + \frac{1}{2}\hbar\omega\sigma_z, \tag{6.2.2}$$

$$\mathscr{H}_1 = \hbar g(\sigma_+ a + a^\dagger \sigma_-). \tag{6.2.3}$$

Here we have removed the subscript from the coupling constant g. The Hamiltonian, given by Eqs. (6.2.1)–(6.2.3), describes the atom–field interaction in the dipole and rotating-wave approximations. As we show below, this important Hamiltonian of quantum optics provides us with an exactly solvable example of the field–matter interaction.

It is convenient to work in the interaction picture. The Hamiltonian, in the interaction picture, is given by

$$\mathscr{V} = e^{i\mathscr{H}_0 t/\hbar}\mathscr{H}_1 e^{-i\mathscr{H}_0 t/\hbar}. \tag{6.2.4}$$

Using

$$e^{\alpha A}Be^{-\alpha A} = B + \alpha[A,B] + \frac{\alpha^2}{2!}[A,[A,B]] + \ldots, \tag{6.2.5}$$

it can be readily seen that

$$e^{iva^\dagger at}ae^{-iva^\dagger at} = ae^{-ivt}, \tag{6.2.6}$$

$$e^{i\omega\sigma_z t/2}\sigma_+ e^{-i\omega\sigma_z t/2} = \sigma_+ e^{i\omega t}. \tag{6.2.7}$$

Combining Eqs. (6.2.1)–(6.2.3), (6.2.4), (6.2.6), and (6.2.7), we have

$$\mathscr{V} = \hbar g(\sigma_+ a e^{i\Delta t} + a^\dagger \sigma_- e^{-i\Delta t}), \tag{6.2.8}$$

where $\Delta = \omega - v$.

In this section, we present three different but equivalent methods to solve for the evolution of the atom–field system described by the Hamiltonian (6.2.1)–(6.2.3) based on the solutions of the probability amplitudes, the Heisenberg field and atomic operators, and the unitary time-evolution operator.

6.2.1 Probability amplitude method

We first proceed to solve the equation of motion for $|\psi\rangle$, i.e.,

$$i\hbar\frac{\partial|\psi\rangle}{\partial t} = \mathscr{V}|\psi\rangle. \tag{6.2.9}$$

At any time t, the state vector $|\psi(t)\rangle$ is a linear combination of the states $|a,n\rangle$ and $|b,n\rangle$. Here $|a,n\rangle$ is the state in which the atom is in