SPONTANEOUS EMISSION

Atomic systems can interact with electromagnetic radiation in different ways. Atoms can absorb photons from the radiation field making transitions from a lower state to a higher state of energy. Atoms can also emit photons under the influence of an applied radiation field. This process is called *stimulated emission*. There is a third elementary process in which atomic systems can make spontaneous transitions from an excited state to a state of lower energy with the emission of photons even in the absence of any externally applied radiation field. This process is called *spontaneous emission.* Spontaneous emission is a remarkable manifestation of the interaction between an atom and vacuum radiation fields. Figure 1 sketches the three processes described here.

The development of coherent sources of electromagnetic fields has generated considerable interest in the interaction of matter and radiation. Soon after tunable lasers became widely available, atom-radiation interaction turned into a very active topic of study allowing for high precision spectroscopy measurements, including spontaneous emission rates. However, the recent development of high quality techniques to fabricate microcavities (1–8) and new methods to confine and observe a small number of atoms (9,10) have provided new observation tools for the study of the basic phenomena associated with the interaction between atoms and electromagnetic radiation.

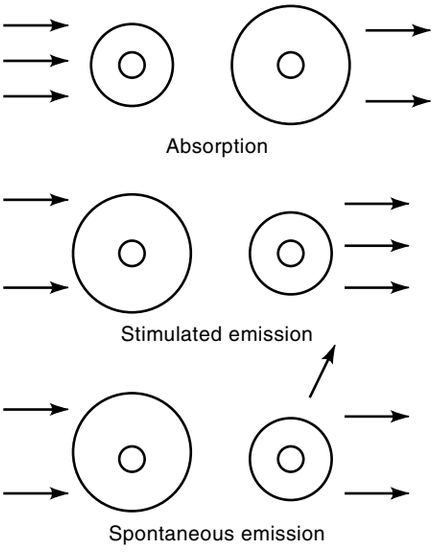

Figure 1. Basic processes of atomic excitation outlined in the text. Photon absorption, stimulated photon emission and spontaneous emission. Photons are represented by arrows.

The main goal of this article is to introduce in the simplest possible way some fundamental processes of atomic excitation and spontaneous emission of radiation. Following a presentation of basic definitions, we trace a succession of elementary concepts beginning with classical rate equations and concluding with the quantum mechanical description of the two-state atom excited by quantized fields and atomic excitation in cavities. The final section introduces a description of the mechanical effects of spontaneous emission on atom dynamics.

The elementary processes described here are concerned only with single-photon atomic transitions. More complex processes may occur in which the number of photons involved may increase or decrease in several units. Such processes are called multiphoton processes and their description is beyond the scope of this introductory article. A detail description could be found in Refs. 11–13 and references therein.

Over the recent years the research into spontaneous emission of radiation has progressed rapidly. Nowadays, the field is so wide that it is totally impossible to make a complete review of the achievements. A general review of the situation can be found in Refs. 11–13. A presentation of the theoretical and experimental situation and a discussion of future prospects are given in Refs. 14–16.

CLASSICAL ASPECTS OF SPONTANEOUS EMISSION

In the classical electrodynamics picture any accelerated charged-particle emits radiation. Indeed, the classical radiation theory treats changing charge and currents as the source of all electromagnetic radiation. Spontaneous emission fits this criterion since it can occur in the absence of any applied radiation as opposed to stimulated emission. A simple estimate of the spontaneous emission rate can be obtained assuming a semiclassical description of a moving charge. The average power emitted by an electron of charge $-e$ excited by an oscillating radiation field can be derived from the Larmor formula (11)

$$P = \frac{4e^2\langle a_e^2\rangle}{3c^3} = \frac{4e^2\omega_0^4\langle r_e^2\rangle}{3c^3} \tag{1}$$

where $a_e(t) = -\omega_0^2\, r_e(t)$ is the acceleration of the electron produced by a harmonic field of frequency ω_0, and $r_e(t)$ is the amplitude of the electron oscillation. Assuming that the emitted radiation is quantized in units of $\hbar\omega_0$, the rate at which radiation is emitted results

$$\tilde{\Gamma} = \frac{P}{\hbar\omega_0} = \frac{\omega_0^3 e^2\langle r_e^2\rangle}{3c^3\hbar} \tag{2}$$

This radiation emission rate is similar to the spontaneous emission rate for atoms (11) when frequency ω_0 coincides with the natural frequency of an atomic transition.

Although the existence of spontaneous emission can be regarded as a consequence of classical electrodynamics, it should be emphasized, however, that a complete picture of spontaneous emission requires a description of atoms and radiation in terms of quantum theory. Such a description is provided in later sections.

In the following a statistical description of the interaction of atoms and radiation is introduced. The method, developed by Albert Einstein (17) before the advent of quantum mechanics, relates the transition rate for spontaneous emission to those for absorption and stimulated emission.

Radiative Rate Equations

The fundamental radiative processes remain those first postulated by Einstein (17), who identified absorption, spontaneous emission, and stimulated emission as the elementary events by which atoms interact with radiation.

Let us review the elementary description of these processes. Consider an ideal two-level atomic system with populations N_1 and N_2 and energies $E_2 > E_1$. Let us denote $\omega_0 = (E_2 - E_1)/\hbar$ (Bohr frequency) and assume the two states to be nondegenerated.

Our discussion of populations must distinguish between energy states and energy levels. An energy state is the most elementary of distinguishable states of motion; no further subdivision of attributes is possible within the constraints of quantum mechanics. An energy level is a set of states with common energy (a set of degenerate states). A nondegenerate level consists of a single state.

The following description of radiative processes presumes that atoms initially unexcited become exposed to a broadband isotropic radiation field. Radiation is in thermal equilibrium at an absolute temperature T. The energy density of radiation at any frequency is given by the Planck distribution for blackbody radiation (11,18).

As time elapses populations move to the excited state, until a steady-state equilibrium is reached in which the number of atoms being excited by the radiation field just balances the number of being de-excited.

Einstein postulated that the rate at which atoms absorb energy from the radiation field is proportional to N_1, the number of atoms in the lowest energy level, and to $u(\omega_0)$, the energy density of radiation at the resonant frequency ω_0. The rate at which atoms in the high energy level undergo stimulated emission is proportional to N_2, the population of level 2, and to the radiation energy density $u(\omega_0)$. Finally, spontaneous emission from level 2 to level 1 occur at rate proportional only to the excited population N_2. The coefficients of proportionality (termed B_{12}, B_{21}, and A_{21} respectively) are not independent of each other. The connection between them may be established by considering thermodynamic equilibrium. Under this circumstance, the rate at which population arrives in a level must equal the rate at which population leaves that level (11,18).

$$B_{12}N_1u(\omega_0) = B_{21}N_2u(\omega_0) + A_{21}N_2 \tag{3}$$

Taking the high-temperature limit of the Planck radiation distribution $u(\omega) \sim \omega^2/\pi^2c^3k_BT$, the following relation (11,18) can be obtained for the two emission coefficient A_{21} and B_{21} and the absorption coefficient B_{12}

$$A_{21} = \hbar\omega_0 h(\omega_0)B_{21} \qquad B_{12} = \frac{h(E_2/\hbar)}{h(E_1/\hbar)}B_{21} \tag{4}$$

where the continuum density of states $h(\omega) = \omega^2/\pi^2c^3$ counts the number of electromagnetic modes having frequency ω.

The temporal changes in population as they approach to equilibrium from an arbitrary initial condition follow the radiative rate equation

$$\frac{dP_1(t)}{dt} = -\frac{dP_2(t)}{dt} = A_{21}\left[(n+1)P_2(t) - n\frac{h(E_2/\hbar)}{h(E_1/\hbar)}P_1(t)\right] \tag{5}$$

Here we introduced $P_1(t)$ and $P_2(t)$, the excitation probabilities, and n, the mean photon number of a radiation field of energy density $u(\omega_0)$

$$\begin{aligned} P_1 &= \frac{N_1}{N_1+N_2} \\ P_2 &= \frac{N_2}{N_1+N_2} \\ n &= \frac{u(\omega_0)}{\hbar\omega_0 h(\omega_0)} = u(\omega_0)\frac{B_{21}}{A_{21}} \end{aligned} \tag{6}$$

Spontaneous emission appears in Eq. (5) as a contribution independent of the photon number n. The vacuum energy density $u_v = A_{21}/2B_{21}$ associated with this term is present even in the absence of any applied radiation field.

Note that in this two-level model each loss of population from one level exactly balances gain to the other level. Therefore, the dynamics of both levels can be completely described by a single variable. A convenient variable is the population inversion $P(t) = P_2(t) - P_1(t)$ which, for the case of constant number of photons, reduces Eq. (5) to

$$\frac{dP}{d\tau} = -P - \left[\frac{n(1 - h(E_2/\hbar)/h(E_1/\hbar)) + 1}{n(1 + h(E_2/\hbar)/h(E_1/\hbar)) + 1}\right] \tag{7}$$

where $\tau = A_{21}\,[n(1 + h(E_2/\hbar)/h(E_1/\hbar)) + 1]\,t$. Note that spontaneous emission fixes the response time of the system and produces the exponential decay of the population inversion.

Coherent Excitation

In writing rate equations we assume that the excitation radiation is broadband and isotropic and that the absorption and emission of photons are uncorrelated events. When near-resonance monochromatic excitation radiation is used the atom has no time to fully randomize behavior between absorption and emission, and the two events become correlated.

Now let us consider a two-level atomic system interacting with a monochromatic radiation field of frequency ω. Under the continuous action of a monochromatic radiation field, population moves out of the initial concentration in the ground state into the excited state. As more population moves out of the ground state, fewer atoms are available to absorb radiation, and absorption becomes less frequent. In contrast, stimulated emission becomes increasingly important and population begins to flow back to the ground state. The cycle of excitation and de-excitation between the two states can continue as long as the radiation remains steady. The frequency of the excitation cycle Ω_f (known as the Rabi flopping frequency) is given in terms of the coupling strength between the radiation field and the electric dipole moment of the atom (Rabi frequency) (11,13).

$$\Omega_f = \sqrt{(\omega - \omega_0)^2 + \Omega^2} \qquad \Omega = \mathbf{d}\cdot\epsilon\mathscr{E}/\hbar \tag{8}$$

Here **d** is the atomic dipole moment, ϵ and $\mathscr{E}$ are the polarization and strength of the radiation field respectively. The Rabi frequency plays a fundamental role in coherent excitation. Note the contrast between the oscillatory behavior presented here with the approach to equilibrium produced by rate equations.

Note also that there are three frequencies associated with coherent excitation: ω, ω_0, and Ω. The radiation frequency ω is set by appropriate choice of a laser or radiation source. The excitation frequency ω_0 (Bohr frequency) depends upon the atomic system. Finally, the Rabi frequency, Ω, depends upon the radiation field strength and upon the atomic dipole moment.

A monochromatic wave of intensity $I[\mathrm{W/cm^2}]$ creates an electric field whose magnitude is

$$\mathscr{E} = 5.33 \times 10^{-9}\sqrt{I[\mathrm{W/cm^2}]}\,\mathscr{E}_{\mathrm{AU}} \tag{9}$$

Here $\mathscr{E}_{\mathrm{AU}} = e/a_0^2$ is the atomic unit of field strength and a_0 is the Bohr radius. Notice that low intensity laser excitation acts as a very minor perturbation on the internal atomic dynamics. By expressing the field strength in terms of laser intensity the Rabi frequency reads

$$\Omega = 35.12\sqrt{I}\,\frac{|\mathbf{d}\cdot\epsilon|}{ea_0}\,\mathrm{MHz} \tag{10}$$

An atomic-unit dipole moment exposed to a laser intensity of 1 $\mathrm{MW/cm^2}$ has a Rabi frequency of about 35 GHz. This value is much smaller than the characteristic frequency of optical transitions ~100 THz.

Incoherence and Spontaneous Emission

Monochromatic radiation, expressible as an ideal wave train, provides a convenient idealization of electromagnetic radiation. However, no real source of radiation fits this ideal exactly. Deviations from this ideal may occur as result of fluctuations in phase, frequency, or in amplitude. The spectrum of an atomic radiation source, or alternatively the autocorrelation function of the field, provides a simple description of the distribution of frequencies within the radiation.

Consider a wave train characterized, at a fixed point in space, by the positive-frequency part of the electromagnetic field

$$E(t) = \mathscr{E}e^{-j\omega_0 t + j\phi(t)} \tag{11}$$

where $j = \sqrt{-1}$.

Let the frequency ω_0 be fixed but let the phase $\phi(t)$ fluctuate stochastically with time. More precisely, suppose the phase $\phi(t)$ remains constant until a chance event interrupts the phase and reassigns it a random value. During the interval between interruptions the phase shift is $\delta\phi = 0$. After the random interruption the new phase value is completely uncorrelated with the previous value. Thus we require a zero phase shift between interruptions weighted by the probability $p(\tau)$ that the interval τ has passed without interruption. If interruptions are independent and the mean time between phase interruptions is γ^{-1}, then the probability $p(t + dt)$ that the interval $t + dt$ passed without interruptions provided that there were no interruptions before interval t is

$$p(t + dt) = (1 - \gamma dt)p(t) \tag{12}$$

Therefore the probability of an uninterrupted interval τ is

$$p(\tau) = e^{-\gamma\tau} \tag{13}$$

The relevant quantity for describing this random process is the autocorrelation function of the electric field. The particular weighting described produces the normalized autocorrelation function

$$g(\tau) = \frac{\langle E(t+\tau)E(t)\rangle}{\langle E(t)E(t)\rangle} = e^{-j\omega_0\tau - \gamma\tau} \tag{14}$$

The Fourier transform of this autocorrelation function produces an area normalized spectral density

$$G(\omega - \omega_0) = \frac{1}{\pi}\,\mathrm{Re}\int_0^\infty e^{j\omega\tau}g(\tau)\,d\tau = \frac{\gamma}{\pi[(\omega-\omega_0)^2 + \gamma^2]} \tag{15}$$

This line profile, sketched in Fig. 2, is known as the Lorentz profile. It describes a spectral line that is broadened because of memory-erasing phase interruptions. The exponential nature of the correlation function $g(\tau)$ is a reasonable model for the behavior of light from an atom undergoing spontaneous emission. Thus, the Lorentz profile describes the spectrum expected from spontaneous emission. When applied to this situation, it is customary to parameterize the decay of the radiation intensity rather than the field amplitude replacing γ by $\Gamma/2$, where Γ^{-1} is the radiative lifetime of the atom.

Note that the intensity of the spectral line is concentrated around the central value ω_0. The width γ of this distribution gives a measure of the spread in frequencies (bandwidth) that are present in the radiation. Interruptions of monochromatic wave trains decrease coherence and thereby increase the bandwidth.

Typical radiative lifetimes $\Gamma^{-1} \sim 1$ ns $-$ 100 ns are much longer than the period of $2\pi/\omega_0 \sim 1$ fs associated with an excitation frequency in the optical range. Thus, we can regard spontaneous emission as a weak perturbation of atomic orbital motion.

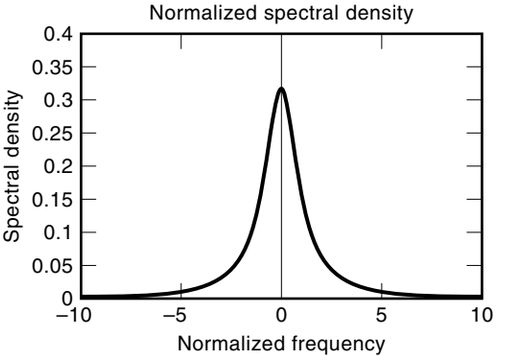

Figure 2. Spectral density expected from spontaneous emission as function of the normalized frequency $(\omega - \omega_0)/\gamma$.

THE ORIGIN OF SPONTANEOUS EMISSION

Spontaneous emission does not fit completely within classical electrodynamics. Quantum mechanical properties of the electromagnetic field play an essential part in the quantitative explanation of spontaneous emission. More precisely, quantum theory predicts that spontaneous emission occurs as a consequence of coupling between the atom and a continuum of unpopulated field modes (11–13).

In the following we introduce a Heinserberg picture of the conventional quantum-mechanical description of spontaneous emission. In this picture it is the operators which evolve in time and the state vector remains fixed and equal to its initial value. The Heisenberg picture of the atom-radiation system provides a straightforward approach that reveals the quantum mechanical origin of spontaneous emission (11–13).

The description of atomic systems poses a very difficult theoretical problem in practice because, except for single electron atoms, the motion of each electron affects that of all others. Neither classical dynamics nor quantum mechanics provides exact solutions for complex atomic systems interacting with radiation. For practical purposes, one introduces simpler models of atomic dynamics. This section examines some of the most elementary processes of excitation of an ideal two-level atom. The atom is assumed to be infinitely heavy and at rest, which allows us to study the evolution of just the internal degrees of freedom. A discussion of the mechanical effects on the translation degrees of freedom of the atom is presented in the last section. The simplicity of the two-level atom makes the model popular as a description of atomic excitation induced by coherent radiation. A more detailed description of the atomic excitation caused by quantized radiation fields can be found in Refs. 19–21 and references therein.

The idealized excitation of a two-level system provides a deep insight into the origin of spontaneous emission. However, it is important to keep in mind the limitations of such a model of atomic excitation.

By definition a two-level system can exist in only two possible states and its state vector must be expressible as an element in a two-dimensional abstract vector space. The dynamics of a perturbed two-level atomic system is determined by a Hamiltonian with a structure

$$H(t) = \begin{pmatrix} E_1 & V_{12}(t) \\ V_{21}(t) & E_2 \end{pmatrix} \tag{16}$$

where $V_{12}(t)$ and $V_{21}(t)$ are the perturbations to the atomic levels 1 and 2.

To ensure that the Hamiltonian retains real-valued eigenvalues (energies) the diagonal elements of H should be real, and the off diagonal elements must have the property $V_{12}(t) = V_{21}^{*}(t)$. The Hamiltonian operator of Eq. (16) can be conveniently expressed as the combination of independent elementary operators $\Pi(n, m; t)$ whose initial values are

$$\begin{aligned} \Pi(1,1;0) &= \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \quad \Pi(1,2;0) = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} \\ \Pi(2,1;0) &= \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} \quad \Pi(2,2;0) = \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} \end{aligned} \tag{17}$$

In the Heisenberg picture operators $\Pi(n, m; t)$ evolve in time from the basic operators $\Pi(n, m, 0)$ in accord with the equation (11,13)

$$\hbar \frac{d}{dt}\Pi(n,m;t) = i[H(t), \Pi(n,m;t)] \tag{18}$$

The equal-time product of a pair of Π operators remains at all times

$$\Pi(n,m;t)\Pi(p,q;t) = \delta(m,p)\Pi(n,q;t) \tag{19}$$

Using these operators we can write Eq. (16), the Hamiltonian of the perturbed atomic system, as

$$\begin{aligned} H(t) =& E_1\Pi(1,1,t) + E_2\Pi(2,2,t) + V_{12}(t)\Pi(1,2,t) \\ &+ V_{21}(t)\Pi(2,1,t) \end{aligned} \tag{20}$$

The expectation values of the two diagonal operators $\Pi(1, 1; t)$ and $\Pi(2, 2; t)$ yield the probability that the two-state atom will be found in energy state 1 or 2

$$\begin{aligned} P_1(t) =& < \Pi(1,1;t) > = \mathrm{Tr}(\rho\Pi(1,1,t)) \\ P_2(t) =& < \Pi(2,2;t) > = \mathrm{Tr}(\rho\Pi(2,2,t)) \end{aligned} \tag{21}$$

Here ρ is the statistical operator, which describes the statistical mixture of atomic states (11,22). The off-diagonal operators $\Pi(1, 2; t) = \Pi(2, 1; t)\dagger$ act as transition operators. Their expectation values (termed coherence) are complex conjugated of each other.

Next, let us examine the dynamics of the two-level atom interacting with a quantized electromagnetic field. The Hamiltonian of the atom-radiation system is

$$H(t) = H_{\mathrm{A}} + H_{\mathrm{R}} + H_{\mathrm{I}} \tag{22}$$

Here

$$\begin{aligned} H_{\mathrm{A}} =& \frac{1}{2}\hbar\omega_0(\Pi(2,2;t) - \Pi(1,1;t)) \\ &+ \frac{1}{2}(E_2 + E_1)(\Pi(2,2;t) + \Pi(1,1;t)) \end{aligned} \tag{23}$$

is the Hamiltonian operator of a free atom, and

$$H_{\mathrm{R}} = \int_{\lambda} \left(\hbar\omega_\lambda a_\lambda^{\dagger} a_\lambda + \frac{1}{2} \right) \tag{24}$$

represents the radiation-field Hamiltonian described in terms of creation and annihilation operator $a_\lambda^{\dagger}$ and a_λ of normal-modes of frequency $\omega_\lambda = ck_\lambda$, where k_λ is the magnitude of the wave-vector in mode λ. Integration over λ encompasses all allowed normal modes (continuous and discrete spectrum). Radiation field operators a_λ evolve with Heinsenberg equations similar to Eq. (18) where $a_\lambda(t)$ replaces $\Pi(n, m; t)$. The evolution of the radiation field is entirely determined by that of the operators $a_\lambda(t)$ and their adjoints. The coupling between the atom and the radiation field is represented by H_{I}.

Our primary interest is with excitation by frequencies near the fundamental frequency ω_0 because such tunings produce great excitation. Let us define the detuning of the excitation frequency as $\Delta = \omega - \omega_0$. If $|\Delta| \ll \omega$ (so the excitation is always

close to resonance) then terms in Eq. (18) with frequency $\omega + \omega_0 \sim 2\omega$ (counter-rotating terms) can be averaged out because they oscillate rapidly. This is known as the rotating wave approximation or RWA (11,23), and is a near-resonance weak-field approximation in which we explicitly recognize the dominance of population oscillations and ignore high-frequency oscillations. The RWA exhibiting Rabi oscillations is the basis for much of the analysis of coherent excitation of atomic systems.

The occurrence of Rabi oscillations requires special conditions that are not always encountered in practice. Basically, such population oscillations occur when the excitation is coherent and monochromatic, sufficiently intense that the Rabi period is shorter than possible relaxation times, yet not so intense that the Rabi frequency exceeds the Bohr frequency or the driving frequency

$$|\Omega| \ll \omega \quad \text{or} \quad \omega_0 \tag{25}$$

In our discussion of resonance excitation we shall be interested in cases for which the detuning is comparable (same order of magnitude) to the Rabi frequency

$$|\Delta| \sim |\Omega| \tag{26}$$

and, therefore, also in the case for which the detuning is much less than ω or ω_0

$$|\Delta| \ll \omega \quad \text{or} \quad \omega_0 \tag{27}$$

For radiation within the optical region of the spectrum, wavelengths are much larger than the atomic dimensions. Therefore, the coupling between the excitation radiation and the atom can be approximated by a spatially uniform, but time varying, electric field interacting with an atomic dipole moment.

Within the rotating-wave approximation and dipole interaction approximation, the coupling between the atom and the radiation field is represented by the interaction Hamiltonian (11,13)

$$H_{\mathrm{I}} = -\mathbf{d}(t) \cdot \mathbf{E}(t) = -\frac{1}{2} \int_{\lambda} (\hbar \Omega_{\lambda}^{*} a_{\lambda}^{\dagger} \Pi(1,2;t) + \hbar \Omega_{\lambda} \Pi(2,1;t) a_{\lambda}) \tag{28}$$

Here the strength of the coupling between the two-state atom and the radiation in mode λ is given by the single-photon Rabi frequency

$$\Omega_{\lambda} = \mathbf{d}_{21} \cdot \epsilon_{\lambda} \mathscr{E}_{\lambda} / \hbar \tag{29}$$

where $\mathscr{E}_{\lambda}$ is the electric field strength in mode λ, and ϵ_{λ} its polarization. In this description the dipole moment operator is expressed in terms of the elementary atomic operators

$$\mathbf{d}(t) = \mathbf{d}_{12} \Pi(1,2;t) + \mathbf{d}_{21} \Pi(2,1;t) \tag{30}$$

where $\mathbf{d}_{mn} = \langle n|\mathbf{d}|m\rangle$.

From Eq. (22), together with commutation properties of the elementary atom and radiation-field operators, it is possible to deduce some properties of a two-state atom system interacting with an infinite set of radiation modes.

The fields that appear in Eqs. (22)–(24) include not only those modes of the fields that are initially highly populated by the applied radiation field, but those modes of the field that become populated after a time because of coupling to the atom. These latter modes represent the field that is radiated as a result of the time varying atomic dipole moment, and incorporate the spontaneous emission field.

Since we are mainly interested in the atomic dynamics, we proceed to eliminate reference to the field dynamics. After integration of the equations of motion for the operators a_{λ}, the radiation field can be decomposed as (11,13)

$$\begin{aligned} a_{\lambda}(t) &= a_{\lambda f}(t) + a_{\lambda \mathrm{s}}(t) \\ &= a_{\lambda}(0) e^{-j\omega_{\lambda} t} + \frac{j}{2} \Omega_{\lambda}^{*} \int_{0}^{t} e^{-j\omega_{\lambda}(t-t')} \Pi(1,2;t')\, dt' \end{aligned} \tag{31}$$

The first part of this operator represents the field in absence of the atom (source-free field) composed of vacuum field together with any incident radiation field. The second contribution to the field operator represents the field generated by the atom (source field). This field can only be found after the dynamics of the atomic operators is known.

Typically thousands of Bohr oscillations periods pass during the course of a spontaneous emission lifetime. It is therefore justified to assume observation times t much longer than a Bohr period, and that the atomic operators primarily oscillate at the Bohr frequency ω_0. Introducing these approximations into Eq. (31) yields (11,13)

$$\begin{aligned} \Pi(1,2;t+\tau) &\simeq \Pi(1,2;t) e^{-j\omega_0 \tau} \\ \Theta(t) a_{\lambda s} &\simeq -\frac{1}{2} \Omega_{\lambda}^{*} \Pi(1,2;t) \zeta(\omega_0 - \omega_{\lambda}) \end{aligned} \tag{32}$$

Here $\Theta(x)$ is the Heaviside function and $\zeta(x) = \mathscr{P}(1/x) - j\pi\delta(x)$ is composed of the principal part of function $1/x$ and the contribution of the delta function. Note that these approximations [based on the Wigner–Weisskopf approximation (11,24)] assume that spontaneous emission is a small perturbation upon otherwise stationary behavior.

Introducing the results of Eq. (32) into the equations of motion Eq. (18) for the atomic operators, and retaining only those operators that are consistent with the RWA, we obtain the following equations for the atomic operators (11)

$$\begin{aligned} \frac{d}{dt}\Pi(1,2;t) &= -j\omega_0 \Pi(1,2;t) - \frac{j}{2}\Phi(t) \int_{\lambda} \Omega_{\lambda} a_{\lambda f} \\ &\quad - (\gamma + j\omega_{\mathrm{s}}) \Phi(t) \Pi(1,2;t) \\ \frac{d}{dt}\Phi(t) &= j \int_{\lambda} \left(\Pi(2,1;t) \Omega_{\lambda} a_{\lambda f} - \Omega_{\lambda}^{*} a_{\lambda f}^{\dagger} \Pi(1,2;t) \right) \\ &\quad - 4\gamma \Pi(2,1;t) \Pi(1,2;t) \end{aligned} \tag{33}$$

where we introduced the population inversion operator

$$\Phi(t) = \Pi(2,2;t) - \Pi(1,1;t) \tag{34}$$

Note that the above approximations (11,13) introduce a frequency shift ω_{s} and a rate (or width) γ.

The preceding equations all follow from the RWA. This approximation neglects counter rotating terms although their inclusion is straightforward. When these terms are included

a complete description of ω_s and γ can be obtained

$$\gamma = \frac{\pi}{4}\int_\lambda \left(|\Omega_\lambda|^2\delta(\omega_\lambda - \omega_0) + |\Omega_\lambda^C|^2\delta(\omega_\lambda + \omega_0)\right)$$
$$\omega_s = \frac{1}{4}\mathscr{P}\int_\lambda \left(\frac{|\Omega_\lambda|^2}{\omega_0 - \omega_\lambda} + \frac{|\Omega_\lambda^C|^2}{\omega_0 + \omega_\lambda}\right) \tag{35}$$

Here $\Omega_\lambda^C = \mathbf{d}_{12} \cdot \boldsymbol{\epsilon}_\lambda \mathscr{E}_\lambda/\hbar$ is the Rabi frequency of the counter rotating terms. For the particular case of an atom in free space, a description of field modes in terms of plane waves yields (11)

$$A_{21} = \Gamma = 2\gamma = \frac{4\omega_0^3|\mathbf{d}_{21}|^2}{3\hbar c^3} \tag{36}$$

which is similar to the semi-classical result described in Eq. (2).

The frequency ω_s, related to the *Lamb shift,* is a shift of the original Bohr transition frequency as a result of the mode structure of the radiation field. The spontaneous emission rate $\Gamma = 2\gamma$ occurs as a consequence of coupling between the atom and the radiation field. Both ω_s and γ are independent of field occupation numbers and they must be viewed as vacuum effects.

The physical interpretation of the emission process described in the preceding equations follows immediately. When an atom in a radiation field makes a spontaneous transition between states, the system cannot be described only as an ideal two-level atom because the final state of the system is composed of the atom in a low energy state together with an emitted photon which may be of any frequency ω and direction, although the chance of finding a value of ω outside a narrow region about ω_0 is very small. The initial states are, in fact, coupled to a continuum of final states. These states are incoherent and cannot act cooperatively to build up the reverse transitions. The exponential decay of population associated with spontaneous emission occurs as a consequence of coupling between the atom and a continuum of unpopulated field modes.

SPONTANEOUS EMISSION IN CAVITIES

The rate of spontaneous emission is not a fixed property of the emitter, but depends on the density of electromagnetic modes and the field intensity of vacuum fluctuations. Under most circumstances the enclosure surrounding an atom is sufficiently large that there is no significant error in treating the mode structure of the radiation field as forming a continuum appropriate to free space. However, when the atom is confined to a cavity the radiation field no longer forms a continuum of modes but a set of discrete modes. Choices of the cavity geometry may either enhance or diminish the number of modes at the Bohr frequency. Thereby, the radiative properties of an atom can be altered by modifying the structure of the radiation field modes. Spontaneous emission rate can be increased when the cavity is resonant with the atomic transition and inhibited when it is out of resonance and subtends a large solid angle at the atom (13,25–27).

The modification of the free-space radiation modes could be produced by changing the boundary conditions of the electromagnetic field in the vicinity of the atom. The simplest alteration of the field boundary conditions is a single reflecting surface (28). For illustration purposes, let us assume an atomic system emitting radiation close to a mirror. The atom is considered to be a dipole oscillator responding to its own field reflected from the mirror. If the reflected field is in phase with the dipole then the decay rate will be enhanced. In contrast, if the reflected field is out of phase the emission rate will be reduced. The phase of the reflected field depends on the distance between the atom and the surface. Thus, the emission rate oscillates with increasing distance as the phase of the reflected field changes. Figure 3 sketches the dependence of the spontaneous emission rate as a function of the distance between the atom and the mirror assuming a classical description of the radiation field. A detailed description of this phenomenon can be found in Refs. 28 and 29.

Because cavities have a more complex structure than single mirrors, they can have a dramatic effect upon the dynamics of the radiation-atom system (15). If the atom is in free space, the emitted radiation travels away and is irreversibly lost. In contrast, if the atom is in a cavity, then the radiation may be reflected from the walls and may be reabsorbed by the atom. The discreteness of modes and the presence of cavity walls can thus prevent purely exponential decay and give rise to sustained oscillations. In the case of strong coupling to the cavity the atom may exchange its energy many times with the cavity before a photon is finally emitted into the vacuum field. The cavity capability of modifying the radiative properties of atoms led to proposals for new efficient lasers (15,30).

Cavities with dimensions on the order of one wavelength are commonly referred to as microcavities. The simplest of these cavities may be the planar dielectric Bragg-mirror cavity. Planar structures are simple to fabricate. However, since there is no lateral confinement of electromagnetic modes, the transverse modes of the cavity may be poorly defined. A typical planar dielectric microcavity emits most of its spontaneous emission into radiation continuum modes. Recently, many different microcavity geometries, such as post, disk, droplet, and hemispherical (1–8) have been proposed to achieve high coupling between the atom and the radiation field. Radiative

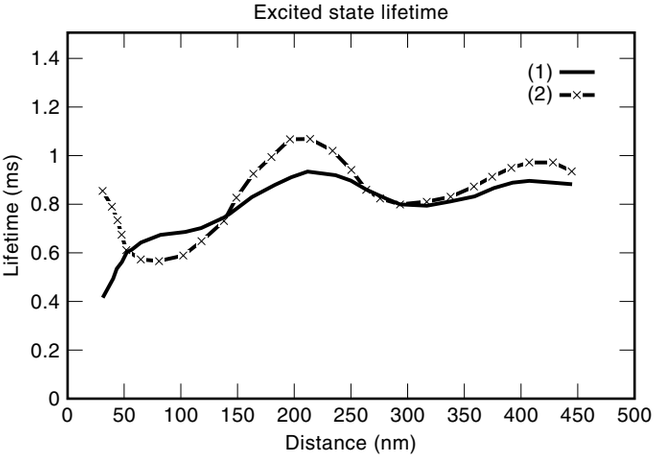

Figure 3. The excited state lifetime of atoms (Γ^{-1}) as function of the distance from (1) a thin mirror (14 nm) and (2) a thick mirror (200 nm). Both curves are based on the classical dipole model outlined in Ref. 28.

properties of semiconductors embedded in microcavities have also been reported (31,32).

In general, the efficiency of the coupling between a cavity and an atom is measured in terms of its spontaneous emission factor. This factor is defined as the ratio between the spontaneous emission radiated into a mode and the total spontaneous emission radiated by the atomic system. It is important to realize that the value of the coupling factor is the result of the interaction between the cavity and the atomic system. The same cavity will generate different spontaneous emission coupling ratios depending on the radiating system that is put in it. Calculations of spontaneous emission rates in microcavities of simple geometry can be found in references (1–8).

The development of new sources of electromagnetic radiation spanning the range of frequencies from microwave to optical frequencies has generated considerable interest in the radiative properties of atoms in microcavities. Unfortunately, alteration of the spontaneous emission rate at optical frequencies is much more difficult than at microwave frequencies because of cavity losses. A metal-clad optical cavity has large absorption loss. Dielectric structures have been proposed to alter the radiation field but they would involve difficult microfabrication technologies (33).

Atomic Excitation in Cavities

Rather than considering the most general situation of atomic excitation in cavities the present discussion will be limited to a sufficiently simple model so that formalism will not be a major obstacle. More precisely, we present an elementary description of atomic excitation in a cavity without losses, having only one mode (Jaynes–Cummings model) (11,22). This model cannot, of course, include all the details of atomic excitation in cavities, but it will allow us to point out some essential features of spontaneous emission. The frequency of the field mode in the cavity is ω and the Bohr frequency of the atomic system is ω_0. The Hamiltonian for this atom-radiation system is obtained from Eq. (22) as the single mode limit of the RWA. Setting the energy zero-point to be midway between the atomic levels E_1 and E_2 Eq. (22) reduces to

$$\begin{aligned} H(t) = {} & \frac{\omega_0}{2}(\Pi(2,2,t) - \Pi(1,1,t)) + \omega a^{\dagger}(t)a(t) \\ & - \frac{1}{2}\left(\Omega^* a(t)^{\dagger}\Pi(1,2,t) + \Omega\Pi(1,2,t)^{\dagger}a(t)\right) \end{aligned} \tag{37}$$

where the single-mode label λ has been omitted.

The Heisenberg equations resulting from this Hamiltonian are the equations (11,22)

$$\begin{aligned} \frac{d}{dt}\Pi(1,2;t) &= -j\omega_0\Pi(1,2;t) - \frac{j}{2}\Phi(t)\Omega a(t) \\ \frac{d}{dt}\Phi(t) &= j\left(\Pi(2,1;t)\Omega a(t) - \Omega_{\lambda}^* a^{\dagger}(t)\Pi(1,2;t)\right) \\ \frac{d}{dt}a(t) &= -j\omega a(t) + \frac{j}{2}\Omega\Pi(1,2;t) \end{aligned} \tag{38}$$

together with adjoint equations for $\Pi(2, 1, t)$ and $a^{\dagger}(t)$. These equations include both spontaneous and stimulated emission.

Let us consider the evolution of the population inversion $\Phi(t)$. Converting Eq. (38) for the population inversion to a second order differential equation yields (11,26)

$$\frac{d^2\Phi}{dt^2} = -\tilde{\Omega}_{\mathrm{f}}^2\Phi - \Delta\left(2\frac{H}{\hbar} - \omega(2N-1)\right) \tag{39}$$

Here

$$\begin{aligned} \tilde{\Omega}_{\mathrm{f}}^2 &= \Delta^2 + |\Omega|^2 N \\ N &= a^{\dagger}a + \frac{1}{2}\Phi + \frac{1}{2} \end{aligned} \tag{40}$$

The operator $\hat{\Omega}_{\mathrm{f}}$ is the operator equivalent to the flopping frequency of population excitations given in Eq. (8). The excitation number operator N combines the photon number operator $a^{\dagger}a(t)$ with the population inversion $\Phi(t)$. This operator commutes with the Hamiltonian and its expectation value remains constant. The constancy of N means that each change of photon number accompanies a balancing change of atomic excitation.

Note that Eq. (39) is the operator counterpart of the equation of motion of a forced harmonic oscillator. Let us study the expectation values of the solutions of Eq. (39) by assuming that, initially, the atom is in a well-defined excitation state ($\langle\Pi(1, 2; t)\rangle = \langle\Pi(2, 1; t)\rangle = 0$), and that there are n photons in the radiation field. There are two limit cases which show the effects introduced by the cavity: (1) an initially unexcited atom interacting with the cavity mode, and (2) an initially excited atom interacting with the cavity mode.

First, consider the dynamics of an atom that is in its lowest energy state and enters the cavity. Then the expectation value of the initial population inversion is $\langle\Phi(0)\rangle = -1$, the expectation value of the excitation number is $\langle N\rangle = n$ and $\langle\hat{\Omega}\rangle = \sqrt{\Delta^2 + n\,|\Omega|^2}$. In this case, the expectation value of the population inversion evolves in accordance with the expression

$$\langle\Phi(t)\rangle = -\frac{1}{\Delta^2 + n|\Omega|^2}\left[\Delta^2 + n|\Omega|^2\cos\left(\sqrt{\Delta^2 + n|\Omega|^2}t\right)\right] \tag{41}$$

Equation (41) indicates that the population inversion undergoes periodic oscillations at a flopping frequency which depends on n photons.

Consider next the dynamics of an atom that is in its excited state and enters the cavity. Then the expectation value of the initial population inversion is $\langle\Phi(0)\rangle = 1$, the expectation value of the excitation number is $\langle N\rangle = n + 1$ and $\langle\hat{\Omega}\rangle = \sqrt{\Delta^2 + (n+1)\,|\Omega|^2}$. In this case, the expectation value of the population inversion evolves in accordance with the expression

$$\begin{aligned} \langle\Phi(t)\rangle = {} & \frac{1}{\sqrt{\Delta^2 + (n+1)|\Omega|^2}} \\ & \left[\Delta^2 + (n+1)|\Omega|^2\cos\left(\sqrt{\Delta^2 + (n+1)|\Omega|^2}t\right)\right] \end{aligned} \tag{42}$$

In contrast to the case of an initially unexcited atom, Eq. (42) indicates that the population inversion undergoes periodic oscillations at a flopping frequency which depends on $n + 1$ photons.

Although the two cases reveal the same type of population oscillations, there is a significant difference between them. When there are no photons initially present, $n = 0$, there are

no population oscillations for an unexcited atom passing through a cold cavity. The situation is different when the atom is initially excited because the oscillation frequency involve $n + 1$ excitations. Even when there are no photons initially, population oscillations will arise. These oscillations originate with spontaneous emission. An excited atom moving into a cold cavity will spontaneously emit radiation if the cavity frequency matches the Bohr frequency.

MECHANICAL EFFECTS OF SPONTANEOUS EMISSION

The goal of this final section is to discuss some of the mechanical effects on atomic motion introduced by the absorption and emission of radiation. The atom is still represented by a two-level system but we now take into account the motion of its center of mass. The mode structure of the radiation field is assumed a continuum appropriate to free space. The emphasis here will be put on simple physical arguments based on a semiclassical approach. A more quantitative discussion of the quantum and classical aspects of the phenomena described here can be found in Refs. 13, 14, 19, 21, 34–36.

In general, the approach to describing the effects of radiation pressure concentrates on the study of radiation forces resulting during the atom-radiation interaction. This approach could lead to misleading interpretations if an inconsistent division of the atomic dynamics into internal and translational components is used. An example of inconsistent choices occurs when the internal degrees of freedom are modeled as an ideal two-level system described by the Jaynes–Cummings Hamiltonian, while the translational motion is only described in terms of the kinetic energy of the atom (21).

The fluctuations of the radiation force are responsible for a diffusion of the atomic momentum and the heating of the translational degrees of freedom of the atom (34,35). When atoms interchange energy with resonant radiation, the atomic motion must compensate for the momentum change in the radiation field. The corresponding energy is gained by the atom in the form of recoil kinetic energy. By suitable arrangement of the source of the radiation pressure force, cooling or heating of atoms can be achieved (19,34,35).

The physical basis of the cooling effect is the irradiation of atoms with a laser beam whose frequency is chosen slightly lower than the frequency of an atomic transition. Thus, when photons are moving towards the atom the frequency of the light in the frame of reference moving with the atom is Doppler shifted to resonance and the absorption of photons is largely increased (19,34,35).

Photon absorption, and subsequent emission, produces transitions between the atomic levels, modifying both, the atom energy, and momentum. When the atom absorbs a photon, it goes into an excited state undergoing a change of momentum along the direction of propagation of the radiation field. After an induced emission, the atom loses the momentum gained during the photon absorption. In contrast, when a spontaneous emission occurs the atom momentum is reduced on average since the radiated photon direction is randomly distributed and the momentum is irreversibly lost (19). When the atom moves away from the laser beam the opposite effect occurs and its momentum is increased (heating effect). As photons carry a momentum $\hbar\mathbf{k}$, the absorption and emission of radiation leads to a recoil of the atom. If a two-level atom with an initial momentum $\mathbf{p}$ and mass M absorbs a photon of energy $\hbar\omega$, energy and momentum conservation require that (23)

$$\frac{\mathbf{p}^2}{2M} + \hbar\omega = \frac{(\mathbf{p} + \hbar\mathbf{k})^2}{2M} + \hbar\omega_0 \tag{43}$$

This expression reduces to

$$\frac{\mathbf{k}\cdot\mathbf{p}}{M} = \Delta - \delta \tag{44}$$

where $\delta = \hbar\mathbf{k}^2/2M$ is known as the recoil frequency. Thus, energy conservation requires that the absorbed photon has a Doppler shift equal to the detuning minus the recoil frequency. For emission, the Doppler shift is

$$\frac{\mathbf{k}\cdot\mathbf{p}}{M} = \Delta + \delta \tag{45}$$

In previous sections atom recoil was neglected (Raman–Nath approximation). This approximation is usually valid (23) because typically the recoil frequency is several orders of magnitude smaller than the other relevant frequencies such as the excitation frequency ω, the transition frequency ω_0, the Rabi frequency Ω, and the spontaneous emission rate γ.

The rate at which momentum is transferred to the atom in absorption-spontaneous emission cycles is given by (19,35)

$$\eta(\mathbf{v}) = \frac{\Omega^2}{2}\frac{\gamma}{(\Delta - \mathbf{k}\cdot\mathbf{v})^2 + \Omega^2/2 + \gamma^2} \tag{46}$$

where $\mathbf{v} = \mathbf{p}/M$ is the atom velocity.

For atom velocities much smaller than the resonant velocity $\Delta = \mathbf{k}\cdot\mathbf{v}$, the radiation force acting on the atom is η_0 $\mathbf{k} \times (1 - \beta\mathbf{n}\cdot\mathbf{v})$, where $\eta_0 = \eta(\mathbf{v} = 0)$, $\beta = (\partial\eta/\partial\mathbf{v})/\eta_0$ and $\mathbf{n} = \mathbf{k}/k$ is the direction of propagation of the radiation field. The first term corresponds to the average pressure exerted by the radiation field, while the second term corresponds to the radiation pressure damping which contains the projection of the atom velocity along the propagation direction of the field. As a consequence of the stochastic nature of the spontaneous emission process, the atom will also exhibit a velocity spreading of order $\sqrt{2\hbar\gamma/M}$, resulting from the uncertainty of the emitted radiation energy.

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