

## CHAPTER TWO

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# Optical Frequency Amplifiers

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## Optical Frequency Amplifiers

### 2.1 Introduction

We saw in Chapter 1 how the stimulated emission and absorption processes can cause the intensity of a wave propagating through a medium to change. The overall intensity will increase if the number of stimulated emissions can be made larger than the number of absorptions. If this situation occurs then we have built an amplifier that operates through the mechanism of stimulated emission. This *laser amplifier*, in common with electronic amplifiers, only has useful gain over a particular frequency bandwidth. The operating frequency range will be determined by the lineshape of the transition, and, roughly speaking, has the same order of frequency width as the width of the lineshape. To consider in more detail how laser amplifiers operate, we need to examine the various mechanisms by which transitions between different energy states of a particle turn out to cover a range of frequencies. This *line broadening* affects in a fundamental way not only the frequency bandwidth of the amplifier, but also its gain.

To turn a laser amplifier into an oscillator we need to supply an appropriate amount of positive feedback. The level of oscillation will stabilize because the amplifier saturates. We shall see that laser amplifiers fall into two categories that saturate in different ways. The *homogeneously* broadened amplifier consists of a number of amplifying particles that are essentially equivalent while the *inhomogeneously* broadened amplifier contains amplifying particles with a distribution of amplification

characteristics. Finally we shall introduce the concept of the complex susceptibility for absorbing transitions by considering the classical electron oscillator model for interaction of an electron with an external electromagnetic field.

## 2.2 Homogeneous Line Broadening

So far we have been treating the energy levels of atoms, molecules or ions as if they were sharp distinct states of clearly defined energy. In practice this is not so, all energy states are *smearred out* over a finite (although usually very small) range of energies. At the fundamental level this smearing out of the energy is caused by the uncertainty involved in the energy measurement process. This gives rise to an intrinsic and unavoidable amount of line broadening called *natural broadening*.

### 2.2.1 Natural Broadening

This most fundamental source of line broadening arises, as just mentioned, because of uncertainty in the exact energy of the states involved. This uncertainty in measured energy,  $\Delta E$ , arises from the time,  $\Delta t$ , involved in making such a measurement. Heisenberg's uncertainty principle<sup>[2.1]–[2.3]</sup> tells us that the product of these *uncertainties*  $\Delta E \Delta t \sim \hbar^\dagger$ . Now, since an excited particle can only be observed for a time that is of the order of its lifetime, the measurement time  $\Delta t$  is roughly the same as the lifetime, so

$$\Delta E \sim \hbar/\tau = A\hbar. \quad (2.1)$$

The uncertainty in emitted frequency  $\Delta\nu$  is  $\Delta E/h$ , so

$$\Delta\nu \sim A/2\pi. \quad (2.2)$$

When the decay of the excited atom is viewed as a photon emission process we can think of the atom, initially placed in the excited state at time  $t' = 0$ , emitting a photon at time  $t$ . The distribution of these times  $t$  among many such atoms varies as  $e^{-t/\tau}$ . Our knowledge of when the photon is likely to be emitted with respect to the time origin restricts our ability to be sure of its frequency. For example, if a photon is observed at time  $t$ , and is known to have come from a state with lifetime  $\tau$ , we know that the *probable* time  $t'$  at which the atom became excited was  $t - \tau < t' \leq t$ . The longer the lifetime of the state the greater the

<sup>†</sup>  $\hbar = h/2\pi$ .

Fig. 2.1.

uncertainty about when the atom acquired its original excitation. In the limit as  $\tau \rightarrow \infty$ , our knowledge of the time of excitation becomes infinitely uncertain and we can ascribe a very well defined frequency to the emitted photon, in this limit the electromagnetic waveform emitted by the atom approaches infinite length and is undamped.

We can put the above approximate determination of  $\Delta\nu$  on a more exact basis by considering the exponential intensity decay of a group of excited atoms: The decay of each individual excited atom is modelled as an exponentially decaying (damped) cosinusoidal oscillation, as shown schematically in Fig. (2.1). It must be stressed, however, that this is only a convenient way of *picturing* how an excited particle decays and emits electromagnetic radiation. It would not be possible in practice to observe such an electromagnetic field by watching a single excited particle decay. We can only observe a classical field by watching many excited particles simultaneously. However, within the framework of our model we can represent the electric field of a decaying excited particle as

$$e(t) = E_0 e^{-t/\tau_c} \cos \omega_0 t. \quad (2.3)$$

What time constant  $\tau_c$  applies to this damped oscillation? The instantaneous intensity  $i(t)$  emitted by an individual excited atom is

$$i(t) \propto |e(t)|^2 = E_0^2 e^{-2t/\tau_c} \cos^2 \omega_0 t. \quad (2.4)$$

If we observe many such atoms the total observed intensity is

$$\begin{aligned} I(t) &= \sum_{particles} i(t) = \sum_i E_0^2 e^{-2t/\tau_c} \cos^2(\omega_0 t + \epsilon_i) \\ &= \sum_i \frac{E_0^2}{2} e^{-2t/\tau_c} [1 + \cos 2(\omega_0 t + \epsilon_i)], \end{aligned} \quad (2.5)$$

where  $\epsilon_i$  is the phase of the wave emitted by atom  $i$ . In the summation the cosine term gets smeared out because individual atoms are emitting with random phases. So,  $I(t) \propto e^{-2t/\tau_c}$ . However, we know that  $I(t) \propto e^{-t/\tau}$ , where  $\tau$  is the lifetime of the emitting state, so the time constant  $\tau_c$  is in fact  $= 2\tau$ . Thus:

$$e(t) = E_0 e^{-t/2\tau} \cos \omega_0 t. \quad (2.6)$$

To find the frequency distribution of this signal we take its Fourier transform

$$E(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e(t) e^{-i\omega t} dt, \quad (2.7)$$

where

$$\begin{aligned} e(t) &= \frac{E_0}{2} (e^{i(\omega_0 + i/2\tau)t} + e^{-i(\omega_0 - i/2\tau)t}) \text{ for } t > 0 \\ &= 0 \text{ for } t < 0. \end{aligned} \quad (2.8)$$

The start of the period of observation at  $t = 0$ , taken for example at an instant when all the particles are pushed into the excited state, allows the lower limit of integration to be changed to 0, so

$$\begin{aligned} E(\omega) &= \frac{1}{2\pi} \int_0^{\infty} e(t) e^{-i\omega t} dt \\ &= \frac{E_0}{4\pi} \left[ \frac{i}{(\omega_0 - \omega + i/2\tau)} - \frac{i}{(\omega_0 + \omega - i/2\tau)} \right]. \end{aligned} \quad (2.9)$$

The intensity of emitted radiation is

$$I(\omega) \propto |E(\omega)|^2 = E(\omega) E^*(\omega) \propto \frac{1}{(\omega - \omega_0)^2 + (1/2\tau)^2}. \quad (2.10)$$

Or, in terms of ordinary frequency

$$I(\nu) \propto \frac{1}{(\nu - \nu_0)^2 + (1/4\pi\tau)^2}. \quad (2.11)$$

The full width at half maximum height (FWHM) of this function is found from the half intensity points of  $I(\nu)$  that occur at frequencies  $\nu_{\pm\frac{1}{2}}$  as shown in Fig. (2.2). This occurs where

$$\left( \frac{1}{4\pi\tau} \right)^2 = (\nu_{\pm\frac{1}{2}} - \nu_0)^2. \quad (2.12)$$

The FWHM is  $\Delta\nu = \nu_{+\frac{1}{2}} - \nu_{-\frac{1}{2}}$ , which gives<sup>†</sup>

$$\Delta\nu = \frac{1}{2\pi\tau} = \frac{A}{2\pi}. \quad (2.13)$$

<sup>†</sup> A more exact treatment gives  $\Delta\nu = (A_1 + A_2)/2\pi$  where  $A_2$  and  $A_1$  are the Einstein coefficients of the upper and lower levels of the transition.

Fig. 2.2.

So from Eq. (2.11)

$$I(\nu) \propto \frac{1}{(\nu - \nu_0)^2 + (\Delta\nu/2)^2}. \quad (2.14)$$

The normalized form of this function is the lineshape function for natural broadening

$$g(\nu)_N = \frac{(2/\pi\Delta\nu)}{1 + [2(\nu - \nu_0)/\Delta\nu]^2}. \quad (2.15)$$

This type of function is called a Lorentzian. Since natural broadening is the same for each particle it is said to be a *homogeneous* broadening mechanism. Other such mechanisms of homogeneous broadening exist, for example:

- (i) In a crystal the constituent particles of the lattice are in constant vibrational motion. This collective vibration can be treated as equivalent to sound waves bouncing around inside the crystal. These sound waves, just like electromagnetic waves, can only carry energy in discrete amounts. The packets of acoustic energy are called *phonons*, and are analogous in many ways to photons, the principal differences between them being that the phonons only travel at the speed of sound and can only exist in a material medium. Collisions of phonons with the particles of the lattice perturb the phase of any excited, emitting particles present. This type of collision, which does not abruptly terminate the lifetime of the particle in its emitting state, is called a *soft* collision.
- (ii) By *pressure* broadening, particularly in the gaseous and liquid phase: interaction of an emitting particle with its neighbors causes perturbation of its emitting frequency and subsequent broadening of the transition. This interaction may arise in a number of ways:

Fig. 2.3.

- (a) Collisions with neutral particles, which may be *soft* or *hard*. A hard collision causes abrupt decay of the emitting species.
- (b) Collisions with charged particles. These collisions need not be very direct, but may involve a very small interaction that occurs when the charged particle passes relatively near, but perhaps as far as several tens of atomic diameters away from, the excited particle. In any case the relative motion of the charged and excited particles leads to a time-varying electric field at the excited particle that perturbs its energy states. This general effect in which an external electric field perturbs the energy levels of an atom (molecule or ion) is called the *Stark* effect; hence line broadening caused by charged particles (ions or electrons) is called Stark broadening.
- (c) By Van der Waals and resonance interactions (usually small effects). Resonance interactions occur when an excited particle can easily exchange energy with like neighbors, the effect is most important for transitions involving the ground state since in this case there are generally many particles near an excited particle for which the possibility of energy exchange exists. Such a process is shown schematically in Fig. (2.3). Broadening occurs because the *possibility* of energy exchange exists, not because an actual emission/reabsorption process occurs.

### 2.3 Inhomogeneous Broadening

When the environment or properties of particles in an emitting sample are nonidentical, *inhomogeneous* broadening can occur. In this type of broadening the shifts and perturbations of emission frequencies differ from particle to particle.

For example, in a real crystal the presence of imperfections and impurities in the crystal structure alters the physical environment of atoms from one lattice site to another. The random distribution of lattice point environments leads to a distribution of particles whose center frequencies are shifted in a random way throughout the crystal.

### 2.3.1 Doppler Broadening

In a gas the random distribution of particle velocities leads to a distribution in the emission center frequencies of different emitting particles seen by a stationary observer. For an atom whose component of velocity towards the observer is  $v_x$  the observed frequency of the transition, whose stationary center frequency is  $\nu_0$ , is

$$\nu = \nu_0 + \frac{v_x}{c}\nu_0, \quad (2.16)$$

where  $c$  is the velocity of light in the gas.

The Maxwell–Boltzmann distribution of atomic velocities for particles of mass  $M$  at absolute temperature  $T$  is<sup>[2.4],[2.5]</sup>

$$f(v_x, v_y, v_z) = \left(\frac{M}{2\pi kT}\right)^{3/2} \exp\left[-\frac{M}{2kT}(v_x^2 + v_y^2 + v_z^2)\right]. \quad (2.17)$$

The number of atoms per unit volume that have velocities simultaneously in the range  $v_x \rightarrow v_x + dv_x, v_y \rightarrow v_y + dv_y, v_z \rightarrow v_z + dv_z$  is  $Nf(v_x, v_y, v_z)dv_x dv_y dv_z$  where  $N$  is the total number of atoms per unit volume. The  $(M/2\pi kT)^{3/2}$  factor is a normalization constant that ensures that the integral of  $f(v_x, v_y, v_z)$  over all velocities is equal to unity, i.e.,

$$\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} f(v_x, v_y, v_z) dv_x dv_y dv_z = 1. \quad (2.18)$$

The normalized one-dimensional velocity distribution is

$$f(v_x) = \sqrt{\frac{M}{2\pi kT}} e^{-\frac{Mv_x^2}{2kT}}. \quad (2.19)$$

This Gaussian-shaped function is shown in Fig. (2.4). It represents the probability that the velocity of a particle towards an observer is in the range  $v_x \rightarrow v_x + dv_x$ . This is the same as the probability that the frequency be in the range

$$\nu_0 + \frac{v_x}{c}\nu_0 \rightarrow \nu_0 + \left(\frac{v_x + dv_x}{c}\right)\nu_0 = \nu_0 + \frac{v_x}{c}\nu_0 + \frac{dv_x}{c}\nu_0. \quad (2.20)$$

The probability that the frequency lies in the range  $\nu \rightarrow \nu + d\nu$  is the

Fig. 2.4.

same as the probability of finding the velocity in the range  $(\nu - \nu_0)c/\nu_0 \rightarrow (\nu - \nu_0)c/\nu_0 + c/\nu_0 d\nu$ , so the distribution of the emitted frequencies is

$$g(\nu) = \frac{c}{\nu_0} \sqrt{\frac{M}{2\pi kT}} \exp \left[ \left( -\frac{M}{2kT} \right) \left( \frac{c^2}{\nu_0^2} \right) (\nu - \nu_0)^2 \right]. \quad (2.21)$$

This is already normalized (since  $f(v_x)$  was normalized). It is called the normalized Doppler-broadened lineshape function. Its FWHM is

$$\Delta\nu_D = 2\nu_0 \sqrt{\frac{2kT \ln 2}{Mc^2}}. \quad (2.22)$$

It can be seen that this increases with  $\sqrt{T}$  and falls with atomic mass as  $1/\sqrt{M}$ . In terms of the Doppler-broadened linewidth  $\Delta\nu_D$  the normalized Doppler lineshape function is

$$g(\nu) = \frac{2}{\Delta\nu_D} \sqrt{\frac{\ln 2}{\pi}} e^{-[2(\nu - \nu_0)/\Delta\nu_D]^2 \ln 2}. \quad (2.23)$$

This is a Gaussian function. It is shown in Fig. (2.5) compared with a Lorentzian function of the same FWHM. The Gaussian function is much more sharply peaked while the Lorentzian has considerable intensity far away from its center frequency, in its *wings*.

*Example.* The 632.8 nm transition of neon is the most important transition to show laser oscillation in the helium–neon laser. The atomic mass of neon is 20. Therefore, using

$$\begin{aligned} M &= 20 \times 1.67 \times 10^{-27} \text{ kg}, \\ \nu_0 &= 3 \times 10^8 / 632.8 \times 10^{-9} \text{ Hz}, \\ T &= 400 \text{ K}, \end{aligned}$$

which we shall see is an appropriate temperature to use for the gas in

Fig. 2.5.

a He–Ne laser, the Doppler width is  $\Delta\nu_D \sim 1.5$  GHz.<sup>†</sup> Doppler broadening usually dominates over all other sources of broadening in gaseous systems, except occasionally in very heavy gases at high pressures and in highly ionized plasmas of light gases, in the latter case Stark broadening frequently dominates.

Doppler broadening gives rise to a Gaussian lineshape and is a form of inhomogeneous broadening. The inhomogeneous lineshape covers a range of frequencies because many *different* particles are being observed. The particles are *different* in the sense that they have different velocities, and consequently different center frequencies. Homogeneous broadening also always occurs at the same time as inhomogeneous broadening, to a greater or lesser degree. To illustrate this, imagine a hypothetical experiment in which only those particles in a gas within a certain narrow velocity range are observed. The center frequencies of these particles are confined to a narrow frequency band and in this sense there is no inhomogeneous broadening – all the *observed* particles are the same. However, a broadened lineshape would still be observed – the homogeneous lineshape resulting from natural and pressure broadening. This is illustrated in Fig. (2.6). When all particles are observed, irrespective of their velocity, an *overall* lineshape called a *Voigt* profile will be observed. This overall lineshape will be considered in more detail later, but results from the superposition of Lorentzian lineshapes spread across the Gaussian distribution of Doppler-shifted center frequencies, as shown in Fig. (2.7). If the constituent Lorentzians have FWHM  $\Delta\nu_L \ll \Delta\nu_D$  then the overall lineshape remains Gaussian and the system is properly said

<sup>†</sup> The gigahertz (GHz) is a unit of frequency  $\equiv 10^9$  Hz, another designation of high frequency is the terahertz (THz)  $\equiv 10^{12}$  Hz.

Fig. 2.6.

Fig. 2.7.

to be *inhomogeneously* broadened. On the other hand if all observed particles are identical, or almost identical, so that  $\Delta\nu_D \ll \Delta\nu_L$  then the system as a whole is *homogeneously* broadened.

In solid materials inhomogeneous broadening, when it is important, results from lattice imperfections and impurities that cause the local environment, of individual excited particles to differ in a random way. We shall assume that the broadening that thereby results also gives rise to a Gaussian lineshape of appropriate FWHM, which we shall also designate as  $\Delta\nu_D$ .<sup>†</sup>

## 2.4 Optical Frequency Amplification with a Homogeneously Broadened Transition

<sup>†</sup> The designation  $\Delta\nu_D$  originates from Doppler broadening, but is used generally to designate inhomogeneous linewidth.

Fig. 2.8.

In an optical frequency amplifier we are generally concerned with the interaction of a monochromatic radiation field with a transition between two energy states whose center frequency is at, or near, the frequency of the monochromatic field. The magnitude of this interaction with each particle is controlled by the homogeneous lineshape function of the transition.

In the general case, the monochromatic radiation field and the center frequency of the transition are not the same. This situation is shown schematically in Fig. (2.8). The stimulating radiation field is taken to be at frequency  $\nu$  whilst the center frequency of the transition is at  $\nu'$ . The closer  $\nu$  is to  $\nu'$ , the greater the number of transitions that can be stimulated. The stimulated transitions occur at frequency  $\nu$ , since this is the frequency of the stimulating radiation. The number of stimulated transitions is proportional to the homogeneous lineshape function  $g(\nu', \nu)$  and can be written as

$$N_S = N_2 B_{21} \rho(\nu) g(\nu', \nu). \quad (2.24)$$

We have written  $g(\nu', \nu)$  to indicate that this lineshape function has its center frequency at  $\nu'$  but is being evaluated at frequency  $\nu$ . For example, for a Lorentzian broadened line

$$g(\nu', \nu) = \frac{(2/\pi\Delta\nu)}{1 + [2(\nu - \nu')/\Delta\nu]^2}. \quad (2.25)$$

$g(\nu', \nu)$  has its maximum value when  $\nu' = \nu$ . It is important to stress that this lineshape function  $g(\nu', \nu)$  that is used here is the *homogeneous* lineshape function of the individual particles in the system, even though the contribution of homogeneous broadening to the overall broadening in the system may be small, for example when the overall broadening is pre-

Fig. 2.9.

dominantly inhomogeneous. The important point about the interaction of a particle with radiation is that an excited atom, molecule or ion can only interact with a monochromatic radiation field that overlaps its homogeneous (usually Lorentzian-shaped) lineshape profile. For example, consider the case of two excited atoms with different center frequencies, these may be the different center frequencies of atoms with different velocities relative to a fixed observer. The homogeneous lineshapes of these two atoms are shown in Fig. (2.9) together with a monochromatic radiation field at frequency  $\nu$ . Particle  $A$  with center frequency  $\nu_A$  and *homogeneous width*  $\Delta\nu$  can interact strongly with the field while the interaction of particle  $B$  is negligible.

We can analyze the interaction between a plane monochromatic case and a collection of homogeneously broadened particles with reference to Fig. (2.10). As the wave passes through the medium it grows in intensity if the number of stimulated emissions exceeds the number of absorptions. The change in intensity of the wave in travelling a small distance  $dz$  through the medium is

$$\begin{aligned} dI_\nu &= (\text{number of stimulated emissions} - \text{number of absorptions})/\text{vol} \\ &\quad \times h\nu \times dz \\ &= \left( N_2 B_{21} g(\nu', \nu) \frac{I_\nu}{c} - N_1 B_{12} g(\nu', \nu) \frac{I_\nu}{c} \right) h\nu dz \end{aligned} \quad (2.26)$$

Use of the Einstein relations, Eqs. (1.64) and (1.65), gives

$$dI_\nu = \frac{I_\nu}{c} \left( N_2 - \frac{g_2}{g_1} N_1 \right) \frac{c^3 A_{21}}{8\pi h\nu^3} h\nu g(\nu', \nu) dz. \quad (2.27)$$

Fig. 2.10.

Therefore,

$$\frac{dI_\nu}{dz} = \left( N_2 - \frac{g_2}{g_1} N_1 \right) \frac{c^2 A_{21}}{8\pi\nu^2} g(\nu', \nu) I_\nu, \quad (2.28)$$

which has the solution

$$I_\nu = I_\nu(0) e^{\gamma(\nu)z}, \quad (2.29)$$

where  $I_\nu(0)$  is the initial intensity at  $z = 0$ , and

$$\gamma(\nu) = \left( N_2 - \frac{g_2}{g_1} N_1 \right) \frac{c^2 A_{21}}{8\pi\nu^2} g(\nu', \nu). \quad (2.30)$$

$\gamma(\nu)$  is called the gain coefficient of the medium and has the same frequency dependence as  $g(\nu', \nu)$ . If

$$N_2 > \frac{g_2}{g_1} N_1$$

then  $\gamma(\nu) > 0$  and we have an optical frequency amplifier. If

$$N_2 < \frac{g_2}{g_1} N_1$$

then  $\gamma(\nu) < 0$  and net absorption of the incident radiation occurs.

For a system in thermal equilibrium

$$\frac{N_2}{N_1} = \frac{g_2}{g_1} e^{-h\nu/kT} \quad (2.31)$$

and for  $T > 0$ ,  $e^{-h\nu/kT} < 1$ , which implies, that in thermal equilibrium at positive temperatures, we cannot have positive gain. However, if we allow the formal existence of a negative temperature, at least for our system of two levels, then we can have

$$N_2 > \frac{g_2}{g_1} N_1.$$

Such a situation, which is essential for the construction of an optical frequency amplifier, is called a state of *population inversion* or *negative*

*temperature.* This is not a true state of thermal equilibrium and can only be maintained by feeding energy into the system.

In the above discussion we have neglected the occurrence of spontaneous emission: this is reasonable for a truly plane wave as the total number of spontaneous emissions into the zero solid angle subtended by the wave is zero. If the wave being amplified were diverging into a small solid angle  $\delta\omega$ , then  $N_2 A_{21} \delta\omega / 4\pi$  spontaneous emissions per second per unit volume would contribute to the increase in intensity of the wave. However, such emissions, being independent of the incident wave, are not in a constant phase relationship with this wave as are the stimulated emissions.

#### 2.4.1 The Stimulated Emission Rate in a Homogeneously Broadened System

The stimulated emission rate  $W_{21}(\nu)$  is the number of stimulated emissions per particle per second per unit volume caused by a monochromatic input wave at frequency  $\nu$ :

$$W_{21}(\nu) = B_{21} g(\nu', \nu) \rho_\nu. \quad (2.32)$$

$\rho_\nu$  ( $\text{J m}^{-3}$ ) is the energy density of the stimulating radiation. Eq. (2.32) can be rewritten in terms of more practical parameters as

$$W_{21}(\nu) = \frac{A_{21} c^2 I_\nu}{8\pi h \nu^3} g(\nu', \nu). \quad (2.33)$$

$W_{21}(\nu)$  has units  $\text{m}^{-3} \text{s}^{-1}$  per particle. Note that the frequency variation of  $W_{21}(\nu)$  follows the lineshape function. The total number of stimulated emissions is

$$N_s = N_2 W_{21}(\nu). \quad (2.34)$$

### 2.5 Optical Frequency Amplification with Inhomogeneous Broadening Included

Although in this discussion so far we have been restricting our attention to homogeneous systems we can show that Eqs. (2.26–2.30) hold generally, even in a system with inhomogeneous broadening, if we take  $g(\nu', \nu)$  as the total lineshape function.

In an inhomogeneously broadened system we can divide the atoms up into classes, each class consisting of atoms with a certain range of center emission frequencies and the same homogeneous lineshape. For example, the class with center frequency  $\nu''$  in the frequency range  $d\nu''$  has

$N g_D(\nu', \nu'') d\nu''$  atoms in it, where  $g_D(\nu', \nu'')$  is the normalized inhomogeneous distribution of center frequencies – the inhomogeneous lineshape function centered at  $\nu'$ . This class of atoms contributes to the change in intensity of a monochromatic wave at frequency  $\nu$  as

$$\begin{aligned} \Delta(dI_\nu) (\text{from the group of particles in the band } d\nu'') = \\ (N_2 B_{21} g_D(\nu', \nu'') d\nu'' g_L(\nu'', \nu) \frac{I_\nu}{c} \\ - N_1 B_{12} g_D(\nu', \nu'') d\nu'' g_L(\nu'', \nu) \frac{I_\nu}{c}) h\nu dz, \end{aligned} \quad (2.35)$$

where  $g_L(\nu'', \nu)$  is the homogeneous lineshape function of an atom at center frequency  $\nu''$ . Eq. (2.35) is equivalent to Eq. (2.26). The increase in intensity from all the classes of atoms is found by integrating over these classes, that is, over the range of center frequencies  $\nu''$ , so Eq. (2.35) becomes

$$dI_\nu = \frac{I_\nu}{c} (N_2 B_{21} - N_1 B_{12}) \left[ \int_{-\infty}^{\infty} g_D(\nu', \nu'') g_L(\nu'', \nu) d\nu'' \right] \nu dz, \quad (2.36)$$

which in a similar fashion to Eqs. (2.26–2.30) gives

$$\gamma(\nu) = \left( N_2 - \frac{g_2}{g_1} N_1 \right) \frac{c^2 A_{21}}{8\pi\nu^2} g(\nu', \nu), \quad (2.37)$$

where  $g(\nu', \nu)$  is now the overall lineshape function defined by the equation

$$g(\nu', \nu) = \int_{-\infty}^{\infty} g_D(\nu', \nu'') g_L(\nu'', \nu) d\nu''. \quad (2.38)$$

In other words the overall lineshape function is the convolution<sup>[2,6]</sup> of the homogeneous and inhomogeneous lineshape functions. The convolution integral in Eq. (2.38) can be put in more familiar form if we measure frequency relative to the center frequency of the overall lineshape, that is, we put  $\nu' = 0$ , and Eq. (2.38) becomes

$$\begin{aligned} g(0, \nu) &= \int_{-\infty}^{\infty} g_D(0, \nu'') g_L(\nu'', \nu) d\nu'' \\ &= \int_{-\infty}^{\infty} g_D(0, \nu'') g_L(0, \nu - \nu'') d\nu'', \end{aligned} \quad (2.39)$$

which can be written in simple form as

$$g(\nu) = \int_{-\infty}^{\infty} g_D(\nu'') g_L(\nu - \nu'') d\nu''. \quad (2.40)$$

This is recognizable as the standard convolution integral of two functions  $g_D(\nu)$  and  $g_L(\nu)$ .

If  $g_D(\nu', \nu'')$  is indeed a normalized Gaussian lineshape as in Eq. (2.23)

and  $g_L(\nu'', \nu)$  is a Lorentzian, then Eq. (2.38) can be written in the form

$$g(\nu', \nu) = \frac{2}{\Delta\nu_D} \sqrt{\frac{\ell n 2}{\pi}} \frac{y}{\pi} \int_{-\infty}^{\infty} \frac{e^{t^2}}{y^2 + (x - t)^2} dt, \quad (2.41)$$

where  $y = \Delta\nu_D \sqrt{\ell n 2} / \Delta\nu_D$  and  $x = 2(\nu - \nu') \sqrt{\ell n 2} / \Delta\nu_D$ .

This is one way of writing a normalized Voigt profile<sup>[2.7],[2.8]</sup>. The integral in Eq. (2.40) cannot be evaluated analytically but must be evaluated numerically. For this purpose the Voigt profile is often written in terms of the error function for complex argument  $W(z)$ , which is available in tabulated form<sup>[2.9]</sup>

$$g(\nu', \nu) = \frac{2}{\Delta\nu_D} \sqrt{\frac{\ell n 2}{\pi}} \mathcal{R}[W(z)], \quad (2.42)$$

where  $z = x + iy$  and  $\mathcal{R}[w(z)]$  denotes the real part of the function.

## 2.6 Optical Frequency Oscillation – Saturation

If we can force a medium into a state of population inversion for a pair of its energy levels, then the transition between these levels forms an optical frequency amplifier. To turn this amplifier into an oscillator we need to apply appropriate feedback by inserting the amplifying medium between a pair of suitable mirrors. If the overall gain of the medium exceeds the losses of the mirror cavity and ancilliary optics then oscillation will result. The level at which this oscillation stabilizes is set by the way in which the amplifier saturates. How does this happen?

### 2.6.1 Homogeneous Systems

Consider an amplifying transition at center frequency  $\nu_0$  between two energy levels of an atom. Suppose we maintain this pair of levels in population inversion by feeding in energy. In equilibrium in the absence of an external radiation field, the rates  $R_2$  and  $R_1$  at which atoms are fed into these levels must be balanced by spontaneous emission and non-radiative loss processes (such as collisions). The population densities and effective lifetimes of the two levels are  $N_2, \tau_2$  and  $N_1, \tau_1$  respectively as shown in Fig. (2.11). These effective lifetimes include the effect of non-radiative deactivation. If  $X_{2j}$  is the rate per particle per unit volume by which collisions depopulate level 2 and cause the particle to end up in a lower state  $j$  we can write

$$\frac{1}{\tau_2} = \sum_j (A_{2j} + X_{2j}). \quad (2.43)$$

Fig. 2.11.

In equilibrium

$$\frac{dN_2^o}{dt} = R_2 - \frac{N_2^o}{\tau_2} = 0, \quad (2.44)$$

where the term  $N_2^o/\tau_2$  is the total loss rate per unit volume from spontaneous emission and other deactivation processes, so

$$N_2^o = R_2\tau_2, \quad (2.45)$$

where the superscript  $o$  indicates that the population is being calculated in the absence of a radiation field. Similarly, for the lower level of the transition, in equilibrium

$$\frac{dN_1^o}{dt} = R_1 + N_2^o A_{21} - \frac{N_1^o}{\tau_1} = 0. \quad (2.46)$$

The term  $N_1^o/\tau_1$  is the total loss rate per unit volume from the level by spontaneous emission and other deactivation processes, while the term  $N_2^o A_{21}$  is the rate at which atoms are feeding into level 1 by spontaneous emission from level 2. So from Eqs. (2.45) and (2.46)

$$N_1^o = (R_1 + N_2^o A_{21})\tau_1 = (R_1 + R_2\tau_2 A_{21})\tau_1. \quad (2.47)$$

The population inversion is

$$\left(N_2^o - \frac{g_2}{g_1} N_1^o\right) = \Delta N^o = R_2\tau_2 - \frac{g_2}{g_1}\tau_1(R_1 + R_2\tau_2 A_{21}), \quad (2.48)$$

or when the level degeneracies are equal

$$\Delta N^o = R_2\tau_2 - \tau_1(R_1 + R_2\tau_2 A_{21}). \quad (2.49)$$

If we now feed in a monochromatic (or other) signal then stimulated emission and absorption processes will occur. We take the energy density of this signal at some point within the medium to be  $\rho(\nu) = I(\nu)/c$ . The rate at which this signal causes stimulated emissions is

$$W_{21}(\nu) = \int_{-\infty}^{\infty} B_{21}g(\nu_0, \nu)\rho(\nu)ds \quad \text{per particle per second,} \quad (2.50)$$

where  $g(\nu_0, \nu)$  is the *homogeneous* lineshape function. If the input radiation were *white*, which in this context means that  $\rho(\nu)$  is a constant over the range of frequencies spanned by the lineshape function, then

$$W_{21}(\nu) = B_{21}\rho(\nu) \int_{-\infty}^{\infty} g(\nu_0, \nu) d\nu = B_{21}\rho(\nu). \quad (2.51)$$

The total rate at which a monochromatic plane wave causes stimulated transitions is

$$\begin{aligned} W_{21}(\nu) &= \int_{-\infty}^{\infty} B_{21}g(\nu_0, \nu)\rho(\nu) d\nu = \frac{I_\nu}{c} B_{21}g(\nu_0, \nu) \int_{-\infty}^{\infty} \delta(\nu - \nu'') d\nu'' \\ &= B_{21}g(\nu_0, \nu) \frac{I_\nu}{c}. \end{aligned} \quad (2.52)$$

Returning once more to a consideration of the pair of energy levels shown in Fig. (2.11), in equilibrium, in the presence of a radiation field,

$$\frac{dN_2}{dt} = R_2 - \frac{N_2}{\tau_2} - N_2 B_{21}g(\nu_0, \nu)\rho(\nu) + N_1 B_{12}g(\nu_0, \nu)\rho(\nu) = 0 \quad (2.53)$$

and

$$\frac{dN_1}{dt} = R_1 + N_2 A_{21} - \frac{N_1}{\tau_1} + N_2 B_{21}g(\nu_0, \nu)\rho(\nu) - N_1 B_{12}g(\nu_0, \nu)\rho(\nu) = 0. \quad (2.54)$$

If we write  $B_{21}g(\nu_0, \nu)\rho(\nu) = W_{12}(\nu)$ , the stimulated emission rate at frequency  $\nu$  per particle, and neglect degeneracy factors so that we can assume that  $W_{12}(\nu) = W_{21}(\nu) = W$  (equivalent to  $B_{12} = B_{21}$ ), then we have

$$R_2 - \frac{N_2}{\tau_2} - N_2 W + N_1 W = 0, \quad (2.55)$$

and

$$R_1 + N_2 A_{21} - \frac{N_1}{\tau_1} + N_2 W - N_1 W = 0. \quad (2.56)$$

From Eq. (2.55)

$$N_2 = \frac{N_1 W + R_2}{1/\tau_2 + W} \quad (2.57)$$

and from Eq. (2.56)

$$N_2 = \frac{N_1/\tau_1 + N_1 W - R_1}{A_{21} + W}, \quad (2.58)$$

so

$$N_1 = \frac{R_1/\tau_2 + R_2 A_{21} + W(R_1 + R_2)}{1/\tau_1 \tau_2 + W(1/\tau_1 + 1/\tau_2 - A_{21})}. \quad (2.59)$$

The population inversion in the system is now

$$N_2 - N_1 = \frac{N_1 W + R_2}{1/\tau_2 + W} - N_1 = \frac{R_2 - N_1/\tau_2}{1/\tau_2 + W}. \quad (2.60)$$

From Eq. (2.59) this gives

$$\begin{aligned} N_2 - N_1 &= \frac{1/\tau_2(R_2/\tau_1 - R_2A_{21} - R_1/\tau_2) + W(R_2/\tau_1 - R_2A_{21} - R_1/\tau_2)}{(1/\tau_2 + W)(1/\tau_1\tau_2 + W/\tau_2 + W/\tau_1 - WA_{21})} \\ &= \frac{R_2/\tau_1 - R_1/\tau_2 - R_2A_{21}}{1/\tau_1\tau_2 + W(1/\tau_2 + 1/\tau_1 - A_{21})}. \end{aligned} \quad (2.61)$$

Multiplying the numerator and denominator of Eq. (2.61) by  $\tau_1\tau_2$  we get

$$N_2 - N_1 = \frac{R_2\tau_2 - R_1\tau_1 - R_2\tau_1\tau_2A_{21}}{1 + W\tau_2(1 + \tau_1/\tau_2 - A_{21}\tau_1)}. \quad (2.62)$$

The numerator of this expression is just the population inversion in the absence of any light signal,  $\Delta N^o$ . Thus,

$$N_2 - N_1 = \frac{\Delta N^o}{1 + W\tau_2(1 + \tau_1/\tau_2 - A_{21}\tau_1)} \quad (2.63)$$

or with the substitution

$$\phi = A_{21}\tau_2 \left[ 1 + (1 - A_{21}\tau_2) \frac{\tau_1}{\tau_2} \right] \quad (2.64)$$

$$N_2 - N_1 = \frac{\Delta N^o}{1 + \phi W/A_{21}}. \quad (2.65)$$

Now from Eq. (2.33)

$$W = \frac{c^2 A_{21}}{8\pi h\nu^3} I_\nu g(\nu_0, \nu). \quad (2.66)$$

If we define

$$I_s(\nu) = \frac{8\pi h\nu^3}{c^2 \phi g(\nu_0, \nu)} \quad (2.67)$$

then

$$N_2 - N_1 = \frac{\Delta N^o}{1 + I_\nu/I_s(\nu)} \quad (2.68)$$

and  $I_s(\nu)$ , called the saturation intensity, is the intensity of an incident light signal (power area<sup>-1</sup>) that reduces the population inversion to half its value when no signal is present. Note that the value of the saturation intensity depends on the frequency of the input signal relative to the line center.

Returning to our expression for the gain constant of a laser amplifier

$$\gamma(\nu) = (N_2 - N_1) \frac{c^2 A_{21}}{8\pi\nu^2} g(\nu_0, \nu) \quad (2.69)$$

the gain as a function of intensity is, in a *homogeneously* broadened system,

$$\gamma(\nu) = \frac{\Delta N^o}{[1 + I_\nu/I_s(\nu)]} \frac{c^2 A_{21}}{8\pi\nu^2} g(\nu_0, \nu), \quad (2.70)$$

which is reduced, that is, *saturates* as the strength of the amplified signal increases. A good optical amplifier should have a large value of saturation intensity, from Eq. (2.67) this implies that  $\phi$  should be a minimum. In such systems often  $A_{21} \simeq 1/\tau_2$  so  $\phi \simeq 1$ .

### 2.6.2 Inhomogeneous Systems

The problem of gain saturation in inhomogeneous media is more complex. For example, in a gas a plane monochromatic wave at frequency  $\nu$  interacts with a medium whose individual particles have Lorentzian homogeneous lineshapes with FWHM  $\Delta\nu_N$ , but whose center frequencies are distributed over an inhomogeneous (Doppler) broadened profile of width (FWHM)  $\Delta\nu_D$ . The Lorentzian contribution to the overall lineshape is

$$g_L(\nu', \nu) = \frac{(2/\pi\Delta\nu_N)}{1 + [2(\nu - \nu')/\Delta\nu_N]^2}, \quad (2.71)$$

where  $\nu'$  is the center frequency of a particle set by its velocity relative to the observer. The Doppler broadened profile of all the particles is

$$g_D(\nu_0, \nu') = \frac{2}{\Delta\nu_D} \sqrt{\frac{\ell n 2}{\pi}} e^{-[(2(\nu' - \nu_0)/\Delta\nu_D)^2 \ell n 2]}, \quad (2.72)$$

where  $\nu_0$  is the center frequency of a particle at rest. The overall lineshape (from all the particles) is a sum of Lorentzian profiles spread across the particle velocity distribution, as shown in Fig. (2.7).

If  $\Delta\nu_N \gg \Delta\nu_D$  the overall profile remains approximately Lorentzian and the observed behavior of the system will correspond to *homogeneous* broadening. Such a situation is likely to arise for long wavelength transitions in a gas, particularly if this has a high atomic or molecular weight, at pressures where pressure broadening (which is a homogeneous process) is important, and frequently in solid materials. If  $\Delta\nu_D \gg \Delta\nu_N$  (as is often the case in gases) the overall lineshape remains Gaussian and the system is *inhomogeneously* broadened.

Once again we reduce the problem to a consideration of the interaction of a plane electromagnetic wave with a two-state system as shown in Fig. (2.11). As the wave passes through the system its intensity changes according to whether the medium is amplifying or absorbing. We take the intensity of the monochromatic wave to be  $I(\nu, z)$  at plane  $z$  within the medium. The individual particles of the medium have a distribution of emission center frequencies (or absorption center frequencies) because of their random velocities (or, for example, their different crystal environments). We take the population density functions (atoms

$\text{vol}^{-1} \text{Hz}^{-1}$ ) in the upper and lower levels whose center frequency is at  $\nu'$  to be  $N_2(\nu', z)$  and  $N_1(\nu', z)$ , respectively, at plane  $z$ . Atoms are fed into levels 2 and 1 at rates  $R_2(\nu')$  and  $R_1(\nu')$ . These rates are assumed uniform throughout the medium.  $N_2(\nu', z)$ ,  $N_1(\nu', z)$ ,  $R_2(\nu')$ , and  $R_1(\nu')$  are assumed to follow the Gaussian frequency dependence set by the particle velocity distribution, and are normalized so that, for example, the total pumping rate of level 2 is

$$R_2 = \int_{-\infty}^{\infty} R_2(\nu') d\nu' = R_{20} \int_{-\infty}^{\infty} e^{[2(\nu' - \nu_0)/\Delta\nu_D]^2 \ell n^2} d\nu'. \quad (2.73)$$

In practice, the primary pumping process may not have this Gaussian dependence, but even when this is the case, the effect of collisions among particles that have been excited will be to *smear out* any non-Gaussian pumping process into a near-Gaussian form. This conclusion is justified by observations of Doppler-broadened lines under various excitation conditions where deviations from a true Gaussian lineshape are found to be minimal. From Eq. (2.73)

$$R_2 = \sqrt{\frac{\pi}{\ell^2}} \frac{\Delta\nu_D}{2} R_{20}, \quad (2.74)$$

where  $R_{20}$  is a pumping rate constant and the total population density of level at plane  $z$  is

$$N_2 = \int_{-\infty}^{\infty} N_2(\nu', z) d\nu'. \quad (2.75)$$

The rate equations for the atoms whose center frequencies are at  $\nu'$  are

$$\begin{aligned} \frac{dN_2}{dt}(\nu', z) &= R_2(\nu') - N_2(\nu', z) \left[ \frac{1}{\tau_2} + B'_{21}(\nu', \nu) \frac{I(\nu, z)}{c} \right] \\ &+ N_1(\nu', z) B'_{12}(\nu', \nu) \frac{I(\nu, z)}{c} \end{aligned} \quad (2.76)$$

and

$$\begin{aligned} \frac{dN_1}{dt}(\nu', z) &= R_1(\nu') + N_2(\nu', z) \left[ A_{21} + B'_{21}(\nu', \nu) \frac{I(\nu, z)}{c} \right] \\ &- N_1(\nu', z) \left[ \frac{1}{\tau_1} + B'_{12}(\nu', \nu) \frac{I(\nu, z)}{c} \right]. \end{aligned} \quad (2.77)$$

Here we have used the modified Einstein coefficients  $B'_{21}(\nu', \nu)$ ,  $B'_{12}(\nu', \nu)$  that describe stimulated emission processes when the stimulating radiation is at frequency  $\nu$  and the particle's center emission frequency is at  $\nu'$ . Written out in full

$$B'_{21}(\nu', \nu) = B_{21} g(\nu', \nu), \quad (2.78)$$

where  $g(\nu', \nu)$  is the *homogeneous* lineshape function. The rate of change of intensity of the incident wave due to atoms with center frequencies in

a small range  $d\nu'$  at  $\nu'$  is

$$\left[ \frac{dI}{dz}(\nu, z) \right]_{d\nu'} = h\nu \frac{I(\nu, z)}{c} [B'_{21}(\nu', \nu)N_2(\nu', z) - B'_{12}(\nu', \nu)N_1(\nu', z)]d\nu'. \quad (2.79)$$

The total rate of change of intensity due to all the atoms, that is from all possible center frequencies  $\nu'$ , is

$$\frac{dI(\nu, z)}{dz} = \frac{h\nu I(\nu, z)}{c} \int_{-\infty}^{\infty} [B'_{21}(\nu', \nu)N_2(\nu', z) - B'_{12}(\nu', \nu)N_1(\nu', z)]d\nu'. \quad (2.80)$$

In the steady state

$$\frac{dN_2}{dt}(\nu', z) = \frac{dN_1}{dt}(\nu', z) = 0, \quad (2.81)$$

so from Eqs. (2.76) and (2.77)

$$\begin{aligned} & B'_{21}(\nu', \nu)N_2(\nu') - B'_{12}(\nu', \nu)N_1(\nu') \\ &= \frac{B'_{21}(\nu', \nu) \left[ \frac{R_2(\nu')}{1/\tau_2} - \left( \frac{g_2}{g_1} \right) \frac{(R_2(\nu')A_{21} + R_1(\nu')/\tau_2)}{1/\tau_1\tau_2} \right]}{1 + \left[ \frac{(g_1/g_2)(1/\tau_2 - A_{21})}{1/\tau_1\tau_2} + \tau_2 \right] B'_{21}(\nu', \nu) \frac{I(\nu, z)}{c}}. \end{aligned} \quad (2.82)$$

We note that

$$R_2(\nu') = R_{20}e^{-[2(\nu' - \nu_0)/\Delta\nu_D]^2 \ell n 2}. \quad (2.83)$$

Substituting in Eq. (2.80) from (2.82) and (2.83) and bearing in mind that  $B'_{21}(\nu', \nu)$  has a Lorentzian form,

$$B'_{21}(\nu', \nu) = \frac{B_{21} \frac{2}{\pi \Delta\nu_N}}{1 + \left[ \frac{2(\nu - \nu')}{\Delta\nu_N} \right]^2}, \quad (2.84)$$

where  $\Delta\nu_N$  is the *homogeneous* FWHM of the transition, gives

$$\begin{aligned} & \frac{1}{I(\nu, z)} \frac{dI(\nu, z)}{dz} = \gamma(\nu) \\ &= \frac{\gamma_0 \int_{-\infty}^{\infty} d\nu' \left( \frac{2}{\pi \Delta\nu_N} \right) \{1 + [2(\nu - \nu')/\Delta\nu_N]^2\}^{-1} e^{-[2(\nu' - \nu_0)/\Delta\nu_D]^2 \ell n 2}}{1 + \eta I(\nu, z) \left( \frac{2}{\pi \Delta\nu_N} \right) [1 + [2(\nu - \nu')/\Delta\nu_N]^2]^{-1}}. \end{aligned} \quad (2.85)$$

We have made the substitutions

$$\gamma_0 = \frac{h\nu}{c} B_{21} \left[ R_{20}\tau_2 - \frac{g_2}{g_1} \left( \frac{R_{20}A_{21} + R_{10}A_2}{1/\tau_1\tau_2} \right) \right], \quad (2.86)$$

$$\eta = \left[ \frac{g_2}{g_1} \frac{(1/\tau_2 - A_{21})}{1/\tau_1\tau_2} + \tau_2 \right] \frac{B_{21}}{c}. \quad (2.87)$$

Eq. (2.85) can be written

$$\gamma(\nu) = \frac{2\gamma_0 \Delta\nu_N}{\pi} \int_{-\infty}^{\infty} \frac{e^{-[2(\nu' - \nu_0)/\Delta\nu_D]^2 \ell n 2} d\nu'}{4(\nu - \nu')^2 + \Delta\nu_N^2 [1 + 2\eta I(\nu, z)/\pi \Delta\nu_N]}. \quad (2.88)$$

Although it is fairly clear from Eq. (2.86) that the gain of the amplifier falls as  $I(\nu, z)$  increases, it is not easy to see from the integral exactly how this occurs. If the intensity is small the gain approaches its *unsaturated* value

$$\gamma_0(\nu) = \frac{2\gamma_0\Delta\nu_N}{\pi} \int_{-\infty}^{\infty} \frac{e^{-[2(\nu' - \nu_0)\Delta\nu_D]^2 \ell n 2} d\nu'}{4(\nu - \nu')^2 + \Delta\nu_N^2}. \quad (2.89)$$

If Eq. (2.88) is examined closely, for frequencies  $\nu'$  close to the input frequency  $\nu$  the integrand can be written approximately as

$$\frac{e^{-[2(\nu' - \nu_0)/\Delta\nu_D]^2 \ell n 2}}{\Delta\nu_N^2 [1 + 2\eta I(\nu, z)/\pi\Delta\nu_N]}.$$

On the other hand for frequencies  $\nu'$  far from the input frequency, the integrand can be written approximately as

$$\frac{e\{-[2(\nu' - \nu_0)/\Delta\nu_D]^2 \ell n 2\}}{4(\nu - \nu')^2 + \Delta\nu_N^2},$$

which can be seen to be identical to the integrand in the unsaturated gain expression, Eq. (2.89). Thus, we conclude that in making their contribution to the overall gain, particles whose frequencies are far from the input frequency are relatively unaffected by the input radiation, whereas particles whose frequencies are close to that of the input show strong saturation effects. The gain in the system comes largely from those particles whose frequencies are within (roughly) a homogeneous linewidth of the input radiation frequency. The consequences of this are best illustrated by considering a hypothetical experiment, shown schematically in Fig. (2.12), in which the small-signal gain of a predominantly inhomogeneously broadened amplifier is measured with and without a strong saturating signal simultaneously present. Without the presence of a strong signal at a fixed frequency  $\nu_S$  the observed (small-signal) gain follows the Gaussian curve of the overall line profile of the amplifier, as shown in Fig. (2.13a). However if we perform this experiment again when a strong fixed frequency field is also present, which causes saturation of the gain, we find the gain is reduced locally by the saturating effect of the strong field as shown in Fig. (2.13b). This phenomenon is called *hole burning*<sup>[2.10]</sup>. The width of the *hole* that is thus produced is determined by the quantity

$$\Delta\nu_N^2 \left[ 1 + \frac{2\eta I(\nu, z)}{\pi\Delta\nu_N} \right].$$

Fig. 2.12.

Fig. 2.13a.

Fig. 2.13b.

If

$$\Delta\nu_N \sqrt{1 + \frac{2\eta I(\nu, z)}{\pi \Delta\nu_N}} \ll \Delta\nu_D,$$

for example, in a gaseous system where Doppler broadening is the largest

contribution to the total observed line broadening, Eq. (2.88) can be integrated by bringing the much less sharply peaked exponential factor outside the integral. In this case, the sharply peaked Lorentzian lineshape makes the integrand largest for frequencies  $\nu'$  near to  $\nu$ ; over a small range of frequencies  $\nu'$  near  $\nu$  the exponential factor remains approximately constant so Eq. (2.88) can be written

$$\gamma(\nu) = \frac{2\gamma_0\Delta\nu_N}{\pi} e^{-[2(\nu-\nu_0)\Delta\nu_D]^2\ell n2} \times \int_{-\infty}^{\infty} \frac{d\nu'}{4(\nu-\nu')^2 + \Delta\nu_N^2[1 + 2\eta I(\nu, z)/\pi\Delta\nu_N]}. \quad (2.90)$$

Now the integral can be evaluated to give

$$\gamma(\nu) = \gamma_0 \left[ 1 + \frac{2\eta I(\nu, z)}{\pi\Delta\nu_N} \right]^{-\frac{1}{2}} e^{-[2(\nu-\nu_0)/\Delta\nu_D]^2\ell n2}, \quad (2.91)$$

which gives

$$\gamma(\nu) = \gamma_0 \left[ 1 + \frac{I(\nu, z)}{I'_s(\nu)} \right]^{-\frac{1}{2}} e^{-[2(\nu-\nu_0)/\Delta\nu_D]^2\ell n2} \quad (2.92)$$

where  $I'_s(\nu) = \pi\Delta\nu_N/2\eta$  is called the saturation intensity for inhomogeneous broadening. Note that  $\gamma_0$  is the small-signal gain at line center of the inhomogeneously broadened line. It is left as an exercise to the reader to show that  $\gamma_0$  can be written in the form

$$\gamma_0 = \frac{1}{4\pi} \sqrt{\frac{\ell n2}{\pi}} \frac{\lambda^2 A_{21}}{\Delta\nu_D} \left( N_2 - \frac{g_2}{g_1} N_1 \right). \quad (2.93)$$

When Doppler broadening dominates in a system, incident radiation at frequency  $\nu$  cannot interact with those atoms whose Doppler-shifted frequency is different from  $\nu$  by much more than  $\Delta\nu_N$ .

If the amplifier is homogeneously broadened, that is, if

$$\Delta\nu_N \sqrt{1 + \frac{\eta I(\nu, z)}{\pi\Delta\nu_N}} \gg \Delta\nu_D,$$

Eq. (2.88) can be integrated by bringing the less-sharply peaked Lorentzian factor outside the integral to give

$$\begin{aligned} \gamma(\nu) &= \frac{\gamma_0\Delta\nu_D}{\Delta\nu_N\sqrt{\pi\ell n2}} \left\{ \left[ \frac{2(\nu-\nu_0)}{\Delta\nu_N} \right]^2 + 1 + \frac{I(\nu, z)}{I'_s(\nu)} \right\}^{-1} \\ &= \frac{\Delta\nu_D}{2} \sqrt{\pi/\ell n2} \frac{\gamma_0 g(\nu_0, \nu)}{[1 + I(\nu, z)/I'_s(\nu)]}, \end{aligned} \quad (2.94)$$

where  $g(\nu_0, \nu)$  is the homogeneous lineshape function

$$g(\nu_0, \nu) = \frac{(2/\pi\Delta\nu_N)}{1 + [2(\nu-\nu_0)\Delta\nu_N]^2} \quad (2.95)$$

and  $I_s(\nu)$  is the saturation intensity for homogeneous broadening given by

$$I_s(\nu) = \frac{2I'_s(\nu)}{\pi\Delta\nu_N g(\nu_0, \nu)} = \frac{1}{\eta g(\nu_0, \nu)}. \quad (2.96)$$

It can be seen from Eq. (2.87) that for  $g_2 = g_1$ ,  $\eta$  reduces to the expression

$$\eta = \left[ \left( \frac{1/\tau_2 - A_{21}}{1/\tau_1\tau_2} \right) + \tau_2 \right] \frac{B_{21}}{c} \quad (2.97)$$

and  $I_s(\nu)$  reduces to the expression obtained previously as the saturation intensity for homogeneous broadening. Namely,

$$I_s(\nu) = \frac{8\pi h\nu^3}{c^2\phi g(\nu_0, \nu)}, \quad (2.98)$$

where

$$\phi = A_{21}\tau_2 \left[ 1 + (1 - A_{21}\tau_2) \frac{\tau_1}{\tau_2} \right]. \quad (2.99)$$

## 2.7 Power Output from a Laser Amplifier

For a laser amplifier of length  $\ell$  and gain coefficient  $\gamma(\nu)$  the output intensity for a monochromatic input intensity of  $I_0$  ( $\text{W m}^{-2}$ ) at frequency  $\nu$  is

$$I = I_0 e^{\gamma(\nu)\ell} \quad (2.100)$$

if saturation effects are neglected. If saturation effects cannot be neglected then the differential equation that describes how intensity increases must be re-examined. This is

$$\gamma(\nu) = \frac{1}{I} \frac{dI}{dz}. \quad (2.101)$$

For a homogeneously broadened amplifier with saturation an explicit solution to this equation can be found. In this case, if  $\gamma_0(\nu)$  is the small-signal gain,

$$\gamma(\nu) = \frac{\gamma_0(\nu)}{1 + I/I_s(\nu)} = \frac{1}{I} \frac{dI}{dz}, \quad (2.102)$$

which can be rewritten in the form

$$\frac{dI}{I} + \frac{dI}{I_s(\nu)} = \gamma_0(\nu) dz. \quad (2.103)$$

The solution to this equation is

$$I = I_0 e^{\gamma_0(\nu)\ell - (I - I_0)/I_s(\nu)}. \quad (2.104)$$

We can best illustrate how this equation can be used to find the output of a saturated amplifier with a numerical example.

Table (2.1). Iterative Solution of Eq. (2.104).

| LHS    | RHS    |
|--------|--------|
| 10     | 2.57   |
| 8      | 5      |
| 7      | 6.98   |
| 6.9    | 7.21   |
| 6.99   | 7.00   |
| 6.993  | 6.994  |
| 6.9934 | 6.9934 |

*Example.* A homogeneously broadened laser has a saturation intensity of  $3 \text{ W m}^{-2}$  and a small-signal gain at line center of  $0.5 \text{ m}^{-1}$ . Calculate the output intensity if a  $5 \text{ W m}^{-2}$  monochromatic signal at line center enters an amplifier 2 m long.

If saturation were negligible the output intensity would be

$$I = I_0 e^{\gamma_0 \ell} = 5 e^{0.5 \times 2} = 13.59 \text{ W m}^{-2}.$$

Eq. (2.104) must be solved iteratively, we know the solution will be somewhere between the input intensity and the output when saturation is neglected, i.e.,  $5 < I < 13.59$ . We make an initial guess for  $I$  (LHS) and evaluate the RHS of Eq. (2.104). The successive calculation of the LHS and RHS for difficult guesses for  $I$  is shown in Table (2.1). The answer, obtained when the LHS and RHS agree, is  $6.9934 \text{ W m}^{-2}$ .

### 2.8 The Electron Oscillator Model of a Radiative Transition

When a particle decays from an excited state into a lower state, we can model the resultant electric field as a damped oscillation. This behavior leads us to point out the analogy between the decay of an excited particle and the damped oscillation of an electric circuit. For example, for the *RLC* circuit shown in Fig. (2.14) the resonant frequency is  $\nu_0 = 1/2\pi\sqrt{LC}$ . If the sinusoidal driving voltage is disconnected from the circuit, then the oscillation of the circuit decays exponentially – provided the circuit is underdamped, as shown in detail in Appendix 4. The power spectrum of the decaying electric current is Lorentzian, just as it is for a spontaneous transition. The FWHM of the circuit resonance, is  $\nu_0/Q$ , where  $Q$  is called the *quality factor* of the circuit, analogous to the homogeneously broadened linewidth  $\Delta\nu_N$ . A transition

Fig. 2.15.

between states almost always has  $\Delta\nu_N \ll \nu_0$  so clearly has a very high  $Q$ .

In the classical theory of how a particle responds to electromagnetic radiation each of the  $n$  electrons attached to the particle is treated as a damped harmonic oscillator. For example, when an electric field acts on an atom, the nucleus, which is positively charged, moves in the direction of the field while the electron cloud, which is negatively charged, moves in the opposite direction to the field. The resultant separation of the centers of positive and negative charge causes the atom to become an elemental dipole. If the separation of the nucleus and electron cloud is  $d$ , then the resultant dipole has magnitude  $ed$  and points from the negative towards the positive charge.<sup>†</sup> As the frequency of the electric field that acts on the atom increases, the amount of nuclear motion decreases much more rapidly than that of the electrons. At optical frequencies we generally neglect the motion of the nucleus, its great inertia compared to the electron cloud prevents it following the rapidly oscillating applied electric field. If the vector displacement of the  $i$ th electron on the atom from its equilibrium position is  $\mathbf{x}_i$  then at any instant the atom has acquired a dipole moment

$$\mathbf{n} = - \sum_{i=1}^n e\mathbf{x}_i, \quad (2.105)$$

where the summation runs over all the  $n$  electrons on the atom. The magnitude of the displacement of each electron depends on the value of

<sup>†</sup> The magnitude of the electronic charge is  $e \simeq 1.9 \times 10^{-19} \text{C}$ , the charge on an electron is  $-e$ .

the electric field  $\mathbf{E}_i$  at the electron

$$k_i \mathbf{x}_i = -e\mathbf{E}_i, \quad (2.106)$$

where  $k_i$  is a force constant. A time-varying field  $\mathbf{E}$  leads to a time-varying dipole moment. This dipole moment can become large if there is a resonance between the applied field and a particular electron on the atom. This happens if the frequency of the field is near the natural oscillation frequency of a particular electron. Classically, the resonance frequency of electron  $i$  is, by analogy with a mass attached to a spring,  $\omega_i = \sqrt{k_i/m}$ . If the applied electric field is near this frequency, one electron in the summation in Eq. (2.106) makes a dominant contribution to the dipole moment and we can treat the atom as a single electron oscillator. The physical significance of the resonant frequencies of the electrons is that they correspond to the frequencies of transitions that the electrons of the atom can make from one energy state to another. If we confine our attention to one of these resonances then we can treat an  $n$ -electron atom as a one-electron classical oscillator. A time-varying electric field  $E(t)$ , which we assume *a priori* to be at a frequency near to an atomic resonance, perturbs only a single electron to a significant degree, thereby inducing a dipole moment which varies at the same frequency as the applied field. The electron and nucleus are perturbed in opposite directions by the field as shown in Fig. (2.15). We shall see that classically the atom absorbs energy from this field and that the maximum absorption of energy leads to the maximum induced dipole moment (polarization).

The motion of the electrons on each of the particles in the medium, which for simplicity can be assumed to be identical, obeys the differential equation

$$\frac{d^2x}{dt^2} + 2\Gamma \frac{dx}{dt} + \frac{k}{m}x = -\frac{e}{m}E(t). \quad (2.107)$$

The terms on the left hand side of the equation represent, reading from left to right: the acceleration of the electron, a damping term proportional to the electron velocity, and a restoring force. These terms are balanced by the effect of the applied electric field  $E(t)$ . The restoring force is analogous to the restoring force acting on a mass suspended from a spring and given a small displacement from its equilibrium position. The damping can be regarded as a viscous drag that the moving electron experiences because of its interaction with the other electrons on the particle.

We take  $E(t) = \mathcal{R}(Ee^{i\omega t})$  and  $x(t) = \mathcal{R}[X(\omega)e^{i\omega t}]$ . The possibility

that  $E(t)$  and  $x(t)$  are not in phase is taken into account by allowing the function  $X(\omega)$  to include a phase factor. If we define a resonant frequency by  $\omega_0 = \sqrt{k/m}$  then the differential equation (2.107) above becomes

$$(\omega_0^2 - \omega^2)X + 2i\omega\Gamma X = -\frac{e}{m}E, \quad (2.108)$$

giving

$$X(\omega) = \frac{-(e/m)E}{\omega_0^2 - \omega^2 + 2i\omega\Gamma}. \quad (2.109)$$

This is the amplitude of the displacement of the electron from its equilibrium position as a function of the frequency of the applied field.

Near resonance  $\omega \simeq \omega_0$  so

$$X(\omega \simeq \omega_0) = \frac{-(e/m)E}{2\omega_0(\omega_0 - \omega) + 2i\omega_0\Gamma}. \quad (2.110)$$

Now, the dipole moment of a single electron is  $\mu(t) = -e[x(t)]$ . This dipole moment arises from the separation of the electron charge cloud and the nucleus.

If there are  $N$  electron oscillators per unit volume there results a net polarization (dipole moment per unit volume) of:

$$p(t) = -Nex(t) = \mathcal{R}[P(\omega)e^{i\omega t}], \quad (2.111)$$

where  $P(\omega)$  is the complex amplitude of the polarization

$$\begin{aligned} P(\omega) = -NeX(\omega) &= \frac{(Ne^2/m)E_0}{2\omega_0(\omega_0 - \omega) + 2i\omega_0\Gamma} \\ &= \frac{-i(Ne^2/(2m\omega_0\Gamma))}{1 + i(\omega - \omega_0)/\Gamma} E_0. \end{aligned} \quad (2.112)$$

The electronic susceptibility  $\chi(\omega)$  is defined by the equation

$$P(\omega) = \epsilon_0\chi(\omega)E_0, \quad (2.113)$$

where  $\epsilon_0$  is the permittivity of free space.  $\chi(\omega)$  is complex and can be written in terms of its real and imaginary parts as

$$\chi(\omega) = \chi'(\omega) - i\chi''(\omega), \quad (2.114)$$

so

$$p(t) = \mathcal{R}[\epsilon_0\chi(\omega)E_0e^{i\omega t}] = \epsilon_0E_0\chi'(\omega)\cos\omega t + \epsilon_0E_0\chi''(\omega)\sin\omega t. \quad (2.115)$$

It is a commonly used convention that Eq. (2.114) has the negative sign.

Therefore  $\chi'(\omega)$ , the real part of the susceptibility, is related to the in-phase polarization, while  $\chi''(\omega)$ , the complex part, is related to the out of phase (quadrature) component. From Eqs. (2.112) and (2.113)

$$\chi(\omega) = -i \left( \frac{Ne^2}{2m\omega_0\Gamma\epsilon_0} \right) \frac{1}{1 + i(\omega - \omega_0)/\Gamma} \quad (2.116)$$

Fig. 2.16.

so

$$\chi'(\omega) = \left( \frac{Ne^2}{2m\omega_0\Gamma\epsilon_0} \right) \frac{(\omega_0 - \omega)/\Gamma}{1 + (\omega - \omega_0)^2/\Gamma^2}, \quad (2.117)$$

$$\chi''(\omega) = \left( \frac{Ne^2}{2m\omega_0\Gamma\epsilon_0} \right) \frac{1}{1 + (\omega - \omega_0)^2/\Gamma^2}. \quad (2.118)$$

Changing to conventional frequency,  $\nu = \omega/2\pi$ , and putting  $\Delta\nu = \Gamma/\pi$ , which is the FWHM of the Lorentzian shape that describes  $\chi''(\omega)$ , we obtain

$$\chi''(\nu) = \left( \frac{Ne^2}{16\pi^2 m\nu_0\epsilon_0} \right) \frac{\Delta\nu}{(\Delta\nu/2)^2 + (\nu - \nu_0)^2}, \quad (2.119)$$

$$\chi'(\nu) = \frac{2(\nu_0 - \nu)}{\Delta\nu} \chi''(\nu) = \left( \frac{Ne^2}{8\pi^2 m\nu_0\epsilon_0} \right) \frac{\nu_0 - \nu}{(\Delta\nu/2)^2 + (\nu - \nu_0)^2}, \quad (2.120)$$

Fig. (2.16) is a graph of  $\chi''$  and  $\chi'$  normalized to the peak value  $\chi''_0$  of  $\chi''$ . Note that  $\chi''$  has the characteristic Lorentzian shape common to the frequency response of *RLC* circuits and homogeneously broadened spectral lines.<sup>†</sup>

## 2.9 What Are the Physical Significances of $\chi'$ and $\chi''$ ?

To understand the physical significance of a complex susceptibility it will be helpful if we briefly review some basic electromagnetic theory.

<sup>†</sup> It can be shown that although Eqs. (2.119) and (2.120) have the correct frequency dependence for the susceptibility of a real atom, they have incorrect magnitude. This arises because of inadequacies in the classical electron oscillator model and these inadequacies should be borne in mind before the classical model is used to make detailed predictions about the behavior of an atom.

The relationship between the applied electric field  $\mathbf{E}$  and the electron displacement vector  $\mathbf{D}$  is

$$\begin{aligned}\mathbf{D} &= \epsilon_0 \mathbf{E} + \mathbf{P} \\ &= \epsilon_0(1 + \chi) \mathbf{E},\end{aligned}\quad (2.121)$$

which by introducing the dielectric constant  $\epsilon_r = 1 + \chi$  can be written as

$$\mathbf{D} = \epsilon_0 \epsilon_r \mathbf{E}. \quad (2.122)$$

The refractive index  $n$  of the medium is related to  $\epsilon_r$  by  $n = \sqrt{\epsilon_r}$ .

When an external electric field interacts with a group of particles there are two contributions to the induced polarization, a macroscopic contribution  $\mathbf{P}_m$  from the collective properties of the particles, for example their arrangement in a crystal lattice, and a contribution from the polarization  $\mathbf{P}_t$  associated with transitions in the medium, so, in general

$$\mathbf{P} = \mathbf{P}_m + \mathbf{P}_t. \quad (2.123)$$

Usually there are many transitions possible for the particles of the medium but only one will be in near resonance with the frequency of an applied field.  $\mathbf{P}_t$  is dominated by the contribution of this single transition near resonance. Far from any such resonance  $\mathbf{P}_t$  is negligible and  $\mathbf{P} = \mathbf{P}_m$ , which allows us to define the macroscopic dielectric constant  $\epsilon_r$  (far from resonance) from

$$\mathbf{P} = \epsilon_0 \mathbf{E} + \mathbf{P}_m = \epsilon_0 \mathbf{E} + \chi_m \epsilon_0 \mathbf{E} = \epsilon_r \epsilon_0 \mathbf{E}. \quad (2.124)$$

where  $\chi_m$  is the macroscopic, nonresonant, susceptibility. However, if the frequency of the electric field is near the frequency of a possible transition within the medium then there is a significant contribution to  $\mathbf{P}$  from this transition. Other possible transitions far from resonance do not contribute and we can write

$$\mathbf{D} = \epsilon_0 \mathbf{E} + \mathbf{P}_m + \mathbf{P}_t = \epsilon_r \epsilon_0 \mathbf{E} + \mathbf{P}_t. \quad (2.125)$$

$\mathbf{P}_t$  is related to the complex susceptibility that results from the transition according to  $\mathbf{P}_t = \epsilon_0 \chi(\omega) \mathbf{E}$ . Therefore we can rewrite Eq. (2.120) as

$$\mathbf{D} = \epsilon_0 [\epsilon_r + \chi(\omega)] \mathbf{E} = \epsilon_0 \epsilon_r^* \mathbf{E}, \quad (2.126)$$

so the complex susceptibility modifies the effective dielectric constant from  $\epsilon_r$  to  $\epsilon_r^*$ .

When an electromagnetic wave propagates through a medium with a complex susceptibility, both the amplitude and phase velocity of the wave are affected. This can be illustrated easily for a plane wave propagating in the  $z$  direction with a field variation  $\sim e^{i(\omega t - kz)}$ .  $k$  is the

propagation constant, given by the expression

$$k = \omega \sqrt{\mu \epsilon} = \omega \sqrt{\mu_r \mu_0 \epsilon_r \epsilon_0}, \quad (2.127)$$

where for optical materials the relative permeability  $\mu_r$  is usually unity. For a complex dielectric constant Eq. (2.127) can be rewritten as

$$k' = \omega \sqrt{\mu_0 \epsilon_r \epsilon_0} \sqrt{1 + \frac{\chi(\omega)}{\epsilon_r}} = k \sqrt{1 + \frac{\chi(\omega)}{\epsilon_r}}, \quad (2.128)$$

where  $k'$  is now the new propagation constant, which differs from the nonresonant propagation constant  $k$  because of the complex susceptibility resulting from a transition. If  $|\chi(\omega)| \ll \epsilon_r$  Eq. (2.128) can be simplified by the use of the binomial theorem to give

$$k' = k \left[ 1 + \frac{\chi(\omega)}{2\epsilon_r} \right] = k \left[ 1 + \frac{\chi'(\omega)}{2\epsilon_r} - \frac{i\chi''(\omega)}{2\epsilon_r} \right]. \quad (2.129)$$

The wave now propagates through the medium as  $e^{-ik'z}$ . Written out in full the electric field varies as

$$\begin{aligned} E &= E_0 \exp \left( i \left\{ \omega t - k \left[ 1 + \frac{\chi'(\omega)}{2\epsilon_r} - \frac{i\chi''(\omega)}{2\epsilon_r} \right] z \right\} \right) \\ &= E_0 \exp \left( i \left\{ \omega t - k \left[ 1 + \frac{\chi'(\omega)}{2\epsilon_r} \right] z \right\} \right) \exp \left[ -\frac{k\chi''(\omega)}{2\epsilon_r} z \right]. \end{aligned} \quad (2.130)$$

Clearly, this is a wave whose phase velocity is

$$c' = \frac{\omega}{k[1 + \chi'(\omega)/2\epsilon_r]} = \frac{\omega}{k + \Delta k} \quad (2.131)$$

and whose field amplitude changes exponentially with distance. If we write

$$\gamma = -\frac{k\chi''(\omega)}{\epsilon_r} \quad (2.132)$$

then the wave changes its electric field amplitude with distance according to  $e^{(\gamma/2)z}$ .

Now, the intensity of the wave is  $I \propto E(z, t)E^*(z, t)$ , so the intensity of the wave changes as it passes through the medium as  $I \propto e^{\gamma z}$ . We can identify  $\gamma$  as the familiar gain coefficient of the medium, which was calculated previously by considering the spontaneous and stimulated radiative jumps between two energy levels.

Now from Eq. (2.132),

$$\gamma(\nu) = -\frac{k\chi''(\nu)}{n^2} = -\left( \frac{2\pi\nu_0 n}{c_0} \right) \frac{\chi''(\nu)}{n^2}, \quad (2.133)$$

which from Eq. (2.118), obtained by consideration of the system as a collection of classical oscillators, is

$$\gamma(\nu) = -\left( \frac{2\pi\nu}{nc_0} \right) \left( \frac{Ne^2}{16\pi^2 m\nu_0 \epsilon_0} \right) \frac{\Delta\nu}{(\Delta\nu/2)^2 + (\nu - \nu_0)^2}, \quad (2.134)$$

which is *always negative*. Clearly, this is an incorrect result since we have previously shown that

$$\gamma(\nu) = \left( N_2 - \frac{g_2}{g_1} N_1 \right) \frac{c^2 A_{21}}{8\pi\nu^2} g(\nu_0, \nu), \quad (2.135)$$

which can be positive or negative depending on the sign of  $N_2 - (g_2/g_1)N_1$ . Thus, the classical electron oscillator model appears to predict only absorption of incident radiation. It is possible, however, within the framework of the classical electron oscillator model to show that in certain conditions stimulated emission can occur. This will be explained in the last section of this chapter. Although the classical electron oscillator model is instructive, it is not entirely adequate in describing the interaction between particles and radiation. It is better to accept that  $\gamma(\nu) = -k\chi''(\nu)/n^2$  and use Eq. (2.135) as the expression for  $\gamma(\nu)$ . In this case we find that the imaginary part of the complex susceptibility of the medium is

$$\chi''(\nu) = -\frac{n^2\gamma(\nu)}{k} = -\left( \frac{n^2c}{2\pi\nu} \right) \gamma(\nu), \quad (2.136)$$

which from Eq. (2.132) gives

$$\chi''(\nu) = -\frac{[N_2 - (g_2/g_1)N_1] n^2 c^3 A_{21}}{8\pi^3 \nu^3 \Delta\nu} \frac{1}{1 + [2(\nu - \nu_0)/\Delta\nu]^2}. \quad (2.137)$$

This quantum mechanical susceptibility is negative or positive depending on whether  $N_2 - (g_2/g_1)N_1$  is positive or not. A *negative* value of  $\chi''(\nu)$  corresponds to a system in population inversion.

Now it is shown in Appendix 3 that the power expended in inducing polarization in a medium is  $\mathbf{E} \cdot d\mathbf{P}/dt$ . If we write

$$|\mathbf{P}| = P(t) = \mathcal{R}(Pe^{i\omega t}), \quad (2.138)$$

which in an isotropic medium is parallel to  $\mathbf{E}$ , where

$$|\mathbf{E}| = \mathcal{R}(Ee^{i\omega t}), \quad (2.139)$$

then the rate of expenditure of energy is

$$\frac{\text{power}}{\text{volume}} = \mathcal{R}(Ee^{i\omega t}) \frac{\partial}{\partial t} \mathcal{R}(Pe^{i\omega t}). \quad (2.140)$$

Remembering that

$$\mathbf{P} = \epsilon_0 \chi \mathbf{E}, \quad (2.141)$$

and averaging we have

$$\frac{\overline{\text{power}}}{\text{volume}} = \overline{\mathcal{R}(Ee^{i\omega t}) \mathcal{R}(i\omega\epsilon_0\chi Ee^{i\omega t})} \quad (2.142)$$

$$= \frac{1}{2} \mathcal{R}(i\omega\epsilon_0\chi EE^*) \quad (2.143)$$

$$= \frac{\omega}{2} \epsilon_0 |E|^2 \mathcal{R}(i\chi). \quad (2.144)$$

Since  $\chi = \chi' - i\chi''$

$$\frac{\overline{\text{power}}}{\text{volume}} = \frac{\omega\epsilon_0\chi''}{2} |E|^2. \quad (2.145)$$

If our system is in a state of population inversion, that is with a negative  $\chi''$ , then from Eq. (2.145) the average power per volume used in inducing polarization in the medium is *negative* – and the system provides us with more energy than is put in by the electromagnetic wave.

## 2.10 The Classical Oscillator Explanation for Stimulated Emission

If there were no applied electric field acting on the electron, then from Eq. (2.107) the position of the electron would satisfy the equation

$$x(t) = x_0 e^{-\Gamma t} \cos(\omega_0 t + \phi_0), \quad (2.146)$$

where  $\omega_0$  is the resonant frequency and  $\phi_0$  is a phase factor set by the initial conditions. If at time  $t = 0$  the position and velocity of the electron are  $a_0, v_0$ , respectively, then

$$\begin{aligned} a_0 &= x_0 \cos \phi_0, \\ v_0 &= -\Gamma x_0 \cos \phi_0 - \omega_0 x_0 \sin \phi_0. \end{aligned} \quad (2.147)$$

When the electric field is applied we have already seen that in the steady state energy is apparently only absorbed. However, this impression is erroneous. It neglects that in reality no electromagnetic field interacts indefinitely with an electron. Therefore, we must consider what happens when an electron, which can already be regarded as oscillating if it is in an excited state, is suddenly subjected to the additional perturbation of an applied field. We are going to be interested in the behavior of the electron over the first few cycles of the applied field so we neglect damping and write

$$\frac{d^2 x}{dt^2} + \omega_0^2 x = -\frac{eE_0}{2m} (e^{i\omega t} + e^{-i\omega t}), \quad (2.148)$$

where the applied field is  $E_0 \cos \omega t$  and has been written in its complex exponential form. By introducing a new variable  $z = \dot{x} + i\omega_0 x$ , where the dot indicates differentiation with respect to time, Eq. (2.148) can be rewritten in the form

$$\frac{dz}{dt} - i\omega_0 z = -\frac{eE_0}{2m} (e^{i\omega t} + e^{-i\omega t}). \quad (2.149)$$

This equation can be solved by multiplying each term by  $e^{-i\omega_0 t}$  and then integrating to give

$$z e^{-i\omega_0 t} = -\frac{eE_0}{2m} \int (e^{i(\omega-\omega_0)t} + e^{-i(\omega+\omega_0)t}) dt. \quad (2.150)$$

This gives

$$\frac{dx}{dt} + i\omega_0 x = -\frac{eE_0}{2m} \left[ -\frac{ie^{i\omega t}}{(\omega - \omega_0)} + \frac{ie^{-i\omega t}}{(\omega + \omega_0)} + Ae^{i\omega t} \right], \quad (2.151)$$

where  $A$  is a constant of integration. By integrating a second time in a similar manner and introducing the initial values of position and velocity it is left as an exercise to the reader to show that the final solution is

$$x(t) = -\frac{eE_0}{m} \left[ \frac{\cos \omega_0 t - \cos \omega t}{(\omega^2 - \omega_0^2)} \right] + \sqrt{\left(\frac{v_0}{\omega_0}\right)^2 + x_0^2} \cos(\omega_0 t + \phi), \quad (2.152)$$

where  $\tan \phi = -v_0/\omega_0 x_0$ .

By the use of the trigonometrical identity

$$\cos X - \cos Y = -2 \sin \left( \frac{X+Y}{2} \right) \sin \left( \frac{X-Y}{2} \right) \quad (2.153)$$

and assuming that the applied frequency is close to resonance Eq. (2.152) can be written

$$x(t) = -\frac{eE_0}{2m\omega_0} t \sin \omega_0 t + \sqrt{\left(\frac{v_0}{\omega_0}\right)^2 + x_0^2} \cos(\omega_0 t + \phi). \quad (2.154)$$

Thus, near resonance the amplitude of oscillation will increase linearly with time, which is, of course, a consequence of our neglect of damping. However, it is interesting to use Eq. (2.154) to calculate the work done during the first  $n$  cycles of the applied field. This work is calculated as the work done by the electric field in polarizing the medium: the polarization  $\mathbf{P}$  is proportional to electron displacement. The work done is  $\mathbf{E} \cdot \partial \mathbf{P} / \partial t$  (see Appendix 3). During the first  $n$  cycles the total work done by the field is

$$W = \int_0^{2n\pi/\omega_0} \mathbf{E} \cdot \frac{\partial \mathbf{P}}{\partial t} dt = -NeE_0 \int_0^{2n\pi/\omega_0} (\cos \omega_0 t) \dot{x}(t) dt, \quad (2.155)$$

where  $N$  is the total number of particles per unit volume. Writing  $(v_0/\omega_0)^2 + x_0^2 = a^2$  and substituting from Eq. (2.154) gives

$$W = -NeE_0 \int_0^{2n\pi/\omega_0} \left[ -\frac{eE_0}{m\omega_0} \sin 2\omega_0 t - \frac{eE_0 t}{4m} (1 + \cos 2\omega_0 t) - \frac{a\omega_0}{2} \sin \phi + \frac{a\omega_0}{2} \sin(2\omega_0 t + \phi) \right] dt \quad (2.156)$$

Clearly, the first and last terms of the integrand average to zero over a whole number of cycles. The remaining terms can be integrated to give

$$W = \frac{Ne^2 E_0^2}{m} \left( \frac{n^2 \pi^2}{2\omega_0^2} + \frac{n\pi m a}{eE_0} \sin \phi \right). \quad (2.157)$$

This work done by the applied field is negative, implying that the oscillating electrons supply energy to the field if  $\sin \phi < 0$  and  $|\sin \phi| >$

$eE_0n\pi/2m\omega_0^2$ . This is the condition set by classical theory for stimulated emission to occur. Because the charge on the electron is negative, stimulated emission can only result if the electron velocity when the applied field is turned on is in the direction of the field. If the electron velocity is in the same direction as the field the electron is decelerated by the field and consequently radiates energy. If the electron were accelerated by the field then absorption of energy from the field would occur.

There is a maximum number of cycles of the applied field after which the oscillating electrons start, and continue indefinitely, to absorb energy. This is set by the condition

$$n < 2m\omega_0^2/eE_0\pi. \quad (2.158)$$

After a long enough time the motion of the electron is dominated by the first term in Eq. (2.151) and can be written

$$x(t) = -\frac{eE_0}{2m\omega_0}t \sin \omega_0 t \quad (2.159)$$

and the electron velocity is, for large enough  $t$

$$\dot{x}(t) \simeq -\frac{eE_0}{2m}t \cos \omega_0 t. \quad (2.160)$$

The electron now has a velocity that is oppositely directed from the applied field, the electron is being accelerated and absorbs energy from the field.

We can conclude by saying that when an electric field near resonance is applied to an already oscillating electron, stimulated emission can occur at early times provided the initial velocity of the electron is in the same direction as the field.

## 2.11 Problems

(2.1) A homogeneously broadened laser amplifier has a small-signal gain at line center of  $0.6 \text{ m}^{-1}$  and is at line center  $3 \text{ m}$  long. The saturation intensity of the medium at line center is  $3 \text{ W m}^{-2}$ . An input signal of intensity  $2.5 \text{ W m}^{-2}$  enters the amplifier. What is the output intensity if: (a) The input is at line center and gain saturation is neglected? (b) The input is one linewidth (FWHM) from line center and gain saturation is neglected? (c) As in (a) but gain saturation is included, (d) As in (b) but gain saturation is included.

Hint: (c) and (d) need to be solved numerically.

(2.2) A laser amplifier contains two groups of particles centered at frequencies  $\nu_0 - a/2$ ,  $\nu_0 + a/2$ . Each group is individually homogeneously

broadened with homogeneous FWHM  $\Delta\nu_L$ . (a) At what two frequencies  $\nu_0 \pm b$  is the gain of the amplifier a maximum? Consider all three cases: (i)  $a \gg \Delta\nu_L$ , (ii)  $a \sim \Delta\nu_L$ , (iii)  $a \ll \Delta\nu_L$ . (b) What is the relative gain at frequency  $\nu_0$  as a function of  $a/\Delta\nu_L$ ?

(2.3) What is the overall lineshape function if

$$g_D(\nu_0, \nu) = \begin{cases} 1/\Delta\nu_D & \text{for } \nu_0 - \Delta\nu_D/2 \leq \nu \leq \nu_0 + \Delta\nu_D/2; \\ 0, & \text{elsewhere} \end{cases}$$

$$g_L(\nu', \nu'') = \frac{(2/\pi\Delta\nu_L)}{1 + [2(\nu' - \nu'')/\Delta\nu_L]^2}$$

(2.4) What is the stimulated emission rate if:  $A_{21} = 10^8 \text{ s}^{-1}$ , the line is homogeneously broadened with  $\Delta\nu = 1 \text{ GHz}$ , the input radiation is “white” with  $\rho(\nu) = 1 \text{ J m}^{-3} \text{ Hz}^{-1}$ ? What would be the stimulated emission rate if the input radiation were monochromatic, one FWHM from line center, and had intensity  $1 \text{ W m}^{-2}$ ? Take the line center frequency to be  $10^{14} \text{ Hz}$ .

(2.5) A homogeneously broadened amplifier with FWHM=1 GHz and a small-signal gain at line center of  $1 \text{ m}^{-1}$  has a saturation intensity at line center of  $1 \text{ W m}^{-2}$ . An input signal of intensity  $2 \text{ W m}^{-2}$  enters the amplifier, which is 0.5 m long. Calculate: (a) The output intensity neglecting saturation if the input is at 500 MHz above line center. (b) The output intensity including gain saturation if the input signal is 500 MHz above line center. (c) The output intensity including gain saturation if the input signal is at line center (d) The output intensity 500 MHz above line center if an additional  $1 \text{ W m}^{-2}$  signal is also injected 500 MHz below line center.

(2.6) What is the saturation intensity one FWHM from line center in a naturally broadened amplifier if the amplifier has the following parameters:  $A_{21}=10^8 \text{ s}^{-1}$ ,  $\tau_1=1 \text{ ns}$ ,  $\tau_2=5 \text{ ns}$ ,  $\lambda_0=1 \text{ }\mu\text{m}$ .

(2.7) The lifetime of a particular excited argon level varies with pressure as  $\tau = 10^{-8}/(1 + P) \text{ s}$  at 2000 K, where  $P$  is pressure measured in atmospheres. At what pressure will the collisional broadening be as great as the Doppler broadening? Neglect the effect of the lower level lifetime. Take  $\lambda_0 = 488 \text{ nm}$ .

(2.8) A homogeneously broadened gas laser amplifier with  $\Delta\nu = 1 \text{ GHz}$  working at 300 nm has a small-signal gain coefficient at line center of  $1 \text{ m}^{-1}$ . Its susceptibility as a function of frequency varies as

$$\chi''(\nu) \propto \frac{1}{1 + [2(\nu - \nu_0)/\Delta\nu]^2}$$

and

$$\chi'(\nu) = \frac{2(\nu - \nu_0)}{\Delta\nu} \chi''(\nu).$$

Both  $\chi'$  and  $\chi'' \ll 1$ . The refractive index of the gas is  $n = 1$ . Calculate the slowest phase velocity that will be observed for waves travelling through the amplifier.

- (2.9) A gaseous medium has a Lorentzian broadened lineshape of FWHM 1 GHz. Other parameters of the medium are:  $N_2 = 5 \times 10^{16} \text{ m}^{-3}$ ,  $N_1 = 5 \times 10^{15} \text{ m}^{-3}$ ,  $g_2 = g_1$ ,  $A_{21} = 10^8 \text{ s}^{-1}$ ,  $\tau_2 = 5 \text{ ns}$ ,  $\tau_1 = 1 \text{ ns}$ ,  $\lambda_0 = 1 \mu\text{m}$ . Calculate: (i)  $I_S(\nu_0)$ , (ii)  $\chi = \chi' - i\chi''$ , (iii) the change in phase velocity of a weak signal propagating 1 FWHM from line center relative to  $c_0$ , and (iv) what would happen if  $\tau_1 = 1 \mu\text{ s}$  and the medium was pumped from  $N_2 = 0$ ,  $N_1 = 0$  at time 0 at a rate  $R_2 = 10^{20} \text{ m}^{-3} \text{ s}^{-1}$ ,  $R_1 = 0$ ?
- (2.10) How is the result given in Eq. (2.126) modified if  $|\chi(\omega)|$  is not much smaller than  $\epsilon_r$ ? Derive new expressions for the phase velocity and gain coefficient.
- (2.11) Write a computer program to plot the variation in phase velocity as a function of gain one FWHM on the low frequency side of line center as the gain increases from 0 to  $1/\lambda \text{ m}^{-1}$  for a medium with  $\epsilon_r = 1.5$ , and  $\lambda = 1 \mu\text{m}$ .

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