Polarization response of matter: quantum theory of the constitutive law

Density-matrix approach to the description of the interaction of a light wave with an atomic ensemble

In the previous chapter, we have used time-dependent perturbation theory for deriving the probability of atoms undergoing transitions from one eigenstate of their Hamiltonian into another. This formalism led to Fermi's golden rule and the atomic rate equations, the most frequently used formalism for the description of light-induced atomic transitions and lasers. However, the formalism leaves open several questions that are of crucial importance for the operation of lasers and light-matter interactions:

- Where does the broadening of atomic transitions originate from?
- What are the limits of the rate-equation approximation and what is beyond?
- Is light amplification resulting from stimulated emission a coherent process?

The quantum theory of the polarization response of an atomic system to a light field *connects polarization with the electric field (constitutive law)* and – by doing so – provides answers to the above questions in a natural manner. The microscopic theory of the constitutive law derives from the quantum-mechanical motion of electrons

• the linear electric susceptibility (and hence refractive index)

as well as

• the nonlinear susceptibilities,

which were input parameters in previous models of light phenomena. The microscopic theory also establishes the limits of validity of the perturbative expansion of the constitutive law and relates the polarization to the electric field beyond the limits of these approximation (*non-perturbative or strong-field regime*).



The quantum theory of the light-induced polarization of an atomic system approach is based on the density matrix formalism developed in Chapter V-1. Again, we assume a near-resonant interaction between the incident light field and the atomic system

$$\omega \approx \omega_0 = (E_2 - E_1)/\hbar$$

so that other non-resonant levels (shown by dashed lines in Fig. V-18 play no role in the interaction except in determining the equilibrium populations N_1^e and N_2^e .

Fig. V-18

As a result, the density matrix describing the state of the atomic ensemble is reduced to a 2x2 matrix

$$\hat{\rho} = \begin{pmatrix} \rho_{11} & \rho_{12} \\ \rho_{21} & \rho_{22} \end{pmatrix} \tag{V-150}$$

which is represented here in terms of the eigenfunctions $|u_1\rangle$ and $|u_2\rangle$ of the unperturbed Hamiltonian \hat{H}_0 of the atoms. The Hamiltonian of the atoms interacting with the incident light field is again written as

$$\hat{H} = \hat{H}_0 + \hat{H}'; \qquad \hat{H}' = -\frac{1}{2}e^{\mathbf{r}}\mathbf{E}_0 e^{-i\omega t} - \frac{1}{2}e^{\mathbf{r}}\mathbf{E}_0^* e^{i\omega t}$$
 (V-151)

which in the same representation takes the matrix form

$$\hat{H} = \begin{pmatrix} E_1 & H'_{12} \\ H'_{21} & E_2 \end{pmatrix} \tag{V-152}$$

where

$$H_{12} = -\mathbf{E}_{0} \langle u_{1} | e^{\mathbf{r}} | u_{2} \rangle = -\mathbf{E}_{0} \, \mathbf{\mu}_{12} ;$$

$$H_{21} = -\mathbf{E}_{0}^{*} \langle u_{2} | e^{\mathbf{r}} | u_{1} \rangle = -\mathbf{E}_{0}^{*} \mathbf{\mu}_{21} = -\mathbf{E}_{0}^{*} \mathbf{\mu}_{12}^{*} = H_{12}^{'*}$$
(V-153)

and we have assumed the states $|U_1\rangle$ and $|U_2\rangle$ to be of definite parity¹, which implies

$$\mu_{11} = \mu_{22} = 0 \tag{V-154}$$

as a consequence of $\mathbf{\mu}_{km} = \int U_k^*(\mathbf{r}) e \mathbf{r} \ U_m(\mathbf{r}) \ d^3 r$.

For the time being we assume the field to be linearly polarized along the *x* axis and the atoms to be fully aligned with the field. Without the loss of generality we assume the electric field amplitude $E_x(t)$ to be real and choose the phases of the eigenfunctions $|U_1\rangle$ and $|U_2\rangle$ such that

$$\mu_{21} = \mu_{12} = \mu = \int u_1^*(\mathbf{r}) e \, x \, u_2(\mathbf{r}) \, d^3 r \tag{V-155}$$

With these simplifications the Hamiltonian takes the form

¹ The eigenfunction of the Hamilton operator with a potential $V(\mathbf{r})$ that satisfies $V(-\mathbf{r}) = V(\mathbf{r})$ can be shown to obey either $u_k(-\mathbf{r}) = u_k(\mathbf{r})$ or $u_k(-\mathbf{r}) = -u_k(\mathbf{r})$, that is to possess even or odd parity, respectively.

$$\hat{H} = \begin{pmatrix} E_1 & -\mu E_x(t) \\ -\mu E_x(t) & E_2 \end{pmatrix}$$
(V-156)

What we are interested in is the time evolution of the polarization density of the atomic ensemble

$$P_{X}(t) = N \overline{\langle \mu \rangle}$$
 (V-157)

under the influence of the field. Here N is the density of atoms and the ensemble average of the atomic dipole moment is according to (V-83) - given by

$$\overline{\langle \mu \rangle} = \sum_{m,n=1}^{2} \rho_{mn} \mu_{nm} = \mu(\rho_{12} + \rho_{21}) = \mu(\rho_{12} + \rho_{12}^{*})$$
(V-158)

where we made use of Eqs. (V-154) and (V-155). The temporal evolution of $P_x(t)$ is determined by those of the components of the density matrix, which - in turn - is governed by the equation of motion

$$\frac{d\hat{\rho}}{dt} = -\frac{i}{\hbar} [\hat{H}, \hat{\rho}] \tag{V-89}$$

Substituting (V-150), (V-152) and (V-156) into (V-89) yields

$$\frac{d\rho_{12}}{dt} = \frac{i}{\hbar} (\hat{H}\hat{\rho} - \hat{\rho}\hat{H})_{12} = -\frac{i}{\hbar} (E_1 \rho_{12} + H_{12} \rho_{22} - \rho_{11} H_{12} - \rho_{12} E_2) =$$

$$= i\omega_0 \rho_{12} - i \frac{\mu}{\hbar} E_x(t) (\rho_{11} - \rho_{22})$$
(V-159)

$$\frac{d\rho_{11}}{dt} = -\frac{i}{\hbar} (\hat{H}\hat{\rho} - \hat{\rho}\hat{H})_{11} = -i\frac{\mu}{\hbar} E_x(t) (\rho_{12} - \rho_{12}^*)$$
(V-160)

$$\frac{d\rho_{22}}{dt} = -\frac{i}{\hbar} (\hat{H}\hat{\rho} - \hat{\rho}\hat{H})_{22} = i\frac{\mu}{\hbar} E_x(t) (\rho_{12} - \rho_{12}^*)$$
(V-161)

where we utilized $\rho_{21} \,{=}\, \rho_{12}^{*}$.

Energy relaxation and dephasing

By definition, ρ_{kk} is the probability of finding an atom in state $|u_k\rangle$, hence the population density of level 1 and 2 are given by

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$$N_1 = N\rho_{11}; N_2 = N\rho_{22}; \Delta N = N(\rho_{11} - \rho_{22})$$
 (V-162)

According to (V-127), with the interacting field E(1) "turned off", these populations relax towards their equilibrium values due to spontaneous emission and inelastic collisions

$$\frac{dN_1}{dt} = -\frac{N_1 - N_1^e}{\tau_1}; \quad \frac{dN_2}{dt} = -\frac{N_2 - N_2^e}{\tau_2}$$
(V-163)

So that for the corresponding components of the density matrix in the absence of interacting radiation we can write

$$\frac{d\rho_{11}}{dt} = -\frac{\rho_{11} - \rho_{11}^e}{\tau_1}$$
(V-164)

$$\frac{d\rho_{22}}{dt} = -\frac{\rho_{22} - \rho_{22}^{e}}{\tau_2}$$
(V-165)

The diagonal elements of the density matrix hence decay with the energy decay time constants τ_k , which are therefore also referred to as the diagonal or longitudinal relaxation time constants.

Once the interacting field E(t) is switched off, the off-diagonal elements of the density matrix also decay, but for a completely different reason. By definition (V-78)

$$\rho_{12} = \overline{C_2^* C_1} = \overline{\left| C_2 \right| \left| C_1 \right| e^{-i\Delta\phi}}$$
(V-166)

where $\Delta \phi = \phi_1 - \phi_2$ is the phase difference between the two components of the wavefunction in its expansion in terms of $|u_1\rangle$ and $|u_2\rangle$. After the external perturbation leading to a non-zero value of ρ_{12} ceases, the ensemble average of this phase term approaches zero with a decay time constant T_2

$$\frac{d\rho_{12}}{dt} = i\omega_0 \rho_{12} - \frac{\rho_{12}}{T_2}$$
(V-167)

as the phase coherence of the wave functions of the individual atoms in the ensemble is gradually lost due to effects perturbing the wave function. T_2 is called the *dephasing time*.

The highest-frequency perturbations are in most cases *elastic collisions*, which cause only a random shift of $\Delta \phi$, occur at a rate of $1/\tau_{ph}$ without affecting the magnitude of c_1 and c_2 in Eq. (V-166) and hence the level populations. The phase coherence also decays due to population decay so that

$$\frac{1}{T_2} = \frac{1}{\tau_{ph}} + \frac{1}{2} \left(\frac{1}{\tau_1} + \frac{1}{\tau_2} \right) = \frac{1}{\tau_{ph}} + \frac{\Gamma}{2}$$
(V-168)

As Eqs. (V-157) and (V-158) imply

$$P_{\chi}(t) = N\mu(\rho_{12} + \rho_{12}^{*}) = 2N\mu \operatorname{Re}[\rho_{12}]$$
(V-169)

dephasing described by (V-167) and (V-168) gives rise to a clearly observable phenomenon: the loss of quantum mechanical coherence in an atomic ensemble leads to the decay of coherent macroscopic polarization with a decay rate 1/T₂

$$P_{\chi}(t) = P_{\chi 0} e^{-t/T_2} \cos \omega_0 t \tag{V-170}$$

Figure V-19 illustrates the decay of the coherent macroscopic polarisation resulting from elastic collisions in a collection of oscillating atomic dipoles



In this (hypothetical) case:

$$P_{x}(t) = P_{x0} e^{-t/\tau_{\rm ph}}$$
 (V-171)

In the hypothetical case of pure population (energy) decay, the intensity of spontaneously emitted radiation, which is proportional to P_{χ}^2 , decays with the decay constant Γ , see Eq. (V-168), hence

$$P_{x}(t) = P_{x0} e^{-\Gamma t/2}$$
 (V-172)

Comparison of Eqs. (V-171) and (V-172) sheds light on the origin of the factor $\frac{1}{2}$ in Eq. (V-168).

The density matrix equations of motion for resonant light-atom interaction

With the effect of the interacting field introduced from (V-159), (V-160) and (V-161) into Eqs. (V-164), (V-165) and (V-167), we obtain the density matrix equations of motion for resonant light-atom interaction

$$\frac{d\rho_{11}}{dt} = -i\frac{\mu}{\hbar}E_{x}(t)\left(\rho_{12}-\rho_{12}^{*}\right) - \frac{\rho_{11}-\rho_{11}^{e}}{\tau_{1}}$$
(V-173a)

$$\frac{d\rho_{22}}{dt} = i\frac{\mu}{\hbar}E_{\chi}(t)\left(\rho_{12}-\rho_{12}^{*}\right) - \frac{\rho_{22}-\rho_{22}^{e}}{\tau_{2}}$$
(V-173b)

$$\frac{d\rho_{12}}{dt} = i\omega_0 \rho_{12} - i\frac{\mu}{\hbar} E_x(t) \left(\rho_{11} - \rho_{22}\right) - \frac{\rho_{12}}{T_2}$$
(V-173c)

and for the other off-diagonal term

$$\rho_{21} = \rho_{12}^* \tag{V-173d}$$

The classical electron oscillator model, the resonant dipole equation

Adding its complex conjugate to Eq. (V-173c), multiplying the new equation with μN and using (V-169) we obtain

$$\frac{dP_x}{dt} = i\omega_0 \alpha - \frac{P_x}{T_2} \tag{V-174a}$$

In a similar way, $(V-173c) - (V-173c)^*$ and its multiplication with μ /Vyields

$$\frac{d\alpha}{dt} = i\omega_0 P_x - i\frac{2\mu^2}{\hbar}\Delta N E_x - \frac{\alpha}{T_2}$$
(V-174b)

where $\alpha = \mu N(\rho_{12} - \rho_{12}^*)$. Differentiation of (V-174a) and substitution of α and $d\alpha / dt$ from (V-174a) and (V-174b) leads to

$$\frac{d^2 P_{\chi}}{dt^2} + \frac{2}{T_2} \frac{dP_{\chi}}{dt} + \left(\omega_0^2 + \frac{1}{T_2^2}\right) P_{\chi} = \frac{2\omega_0 \mu^2}{\hbar} \Delta N E_{\chi}$$
(V-175)

Multiplying (V-173a,b) with N, and substituting α from (V-174a) results in

$$\frac{dN_1}{dt} = -\frac{1}{\hbar\omega_0} E_x \left(\frac{dP_x}{dt} + \frac{P_x}{T_2}\right) - \frac{N_1 - N_1^e}{\tau_1}$$
(V-176)

and a similar equation for N₂. Since $\omega_0 >> 1/T_2$, these equations simplify to

$$\frac{d^2 P_x(t)}{dt^2} + \frac{2}{T_2} \frac{dP_x(t)}{dt} + \omega_0^2 P_x(t) = K \Delta N(t) E_x(t)$$
(V-177)

$$\frac{dN_{1}(t)}{dt} = -\frac{1}{\hbar\omega_{0}}E_{x}(t)\frac{dP_{x}(t)}{dt} - \frac{N_{1}(t) - N_{1}^{\theta}}{\tau_{1}}$$
(V-178)

$$\frac{dN_2(t)}{dt} = \frac{1}{\hbar\omega_0} E_x(t) \frac{dP_x(t)}{dt} - \frac{N_2(t) - N_2^{\theta}}{\tau_2}$$
(V-179)

where the coupling constant

$$\mathcal{K} = \frac{2\omega_0 \,\mu^2}{\hbar} \quad \text{or} \quad \mathcal{K} = \frac{2\omega_0 \,\mu^2}{3\hbar} \tag{V-180a,b}$$

for fully-oriented or randomly-oriented (see Eq. V-113) atoms, respectively.

These equations completely describe the measurable consequences of the interaction of light with an ensemble of resonant atoms: the influence of the field on the (quantum) state of the atoms (Eqs. V-178 and V-179) and the back action of the interacting atoms in form of a polarization response (Eq. V-177 or V-180) to be introduced in Maxwell's equations. Eq. (V-177) has been termed the *Resonant Dipole Equation* by A. E. Siegman and served as the basis for a thorough treatment of laser physics in his excellent monograph: *Lasers* (University Science Books, Mill Valley, CA, 1986).

Eq. (V-177) reveals that the atom responds to the radiation field resonant with its quantum transition, $\omega \approx \omega_0 = (E_2 - E_1)/\hbar$ very much like a *damped classical electron oscillator* (Fig. V-20).





$$P_{X}(t) = -Nex(t)$$

which with the motion of a classical oscillator

$$x(t) = x_0 \exp(-t/T_2 - i\omega_0't)$$

of eigenfrequency

$$\omega_0' = \sqrt{\omega_0^2 - 1/T_2^2} \approx \omega_0$$

and damping time constant T_2 yields a solution of Eq. (V-177) in the absence of a driving field $E_x(t)$.

The coupling between the classical oscillator and the driving electric field is determined by the dipole matrix element μ and the population difference ΔN .

Fig. V-20

Atomic susceptibility

In what follows, we shall be concerned with the response of the atomic ensemble

$$P_{X}(t) = \frac{1}{2} P(t) e^{-i\omega t} + C.C.$$
(V-181)

to a (near-)resonant light field

$$E_{\chi}(t) = \frac{1}{2} E(t) e^{-i\omega t} + C.C.$$
 (V-182)

where P(t) and E(t) are complex, generally time-dependent amplitudes.

Let us first scrutinize the stationary response of the atomic system for

$$E(t) = E_0$$
 (V-183)

Using the definition (IV-66) of the complex (frequency-dependent) susceptibility $\chi = \chi' + i\chi''$, the steady-state solution to (V-177) can be written in the form

$$P_{\chi}(t) = \operatorname{\mathsf{Re}}(\varepsilon_0 \,\chi \, E_0 \, e^{-i\omega t}) = E_0(\varepsilon_0 \chi' \cos \omega t + \varepsilon_0 \,\chi'' \sin \omega t) \tag{V-184}$$

where - for randomly-oriented atoms - the atomic susceptibility (Fig. V-21) is given by

$$\chi''(\omega) = \frac{\mu^2 \pi}{3\epsilon_0 \hbar} \Delta N g(\omega - \omega_0) \qquad (V-185)$$

$$\chi'(\omega) = \frac{\mu^2 \pi(\omega_0 - \omega) T_2}{3\varepsilon_0 \hbar} \Delta Ng(\omega - \omega_0)$$

(V-186) and

$$g(\omega - \omega_0) = \frac{T_2}{\pi} \frac{1}{1 + (\omega - \omega_0)^2 T_2^2} = \frac{(\Delta \omega_0 / 2\pi)}{(\Delta \omega_0 / 2)^2 + (\omega - \omega_0)^2}$$
(V-187)

normalized Lorentzian lineshape function, with a line width (full width at half maximum)

$$\Delta\omega_0 = 2/T_2$$

and ΔN is the steady-state population difference.

Rederivation of the rate equations

In order to determine ΔN we substitute

$$E_{X}(t) = \frac{1}{2} E_{0} e^{-i\omega t} + \frac{1}{2} E_{0}^{*} e^{i\omega t}$$

and

$$P_{X}(t) = \frac{1}{2} \varepsilon_{0} \chi E_{0} e^{-i\omega t} + \frac{1}{2} \varepsilon_{0} \chi E_{0}^{*} e^{i\omega t}$$
(V-189)



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(V-188)

into (V-178) and (V-179) and obtain

$$\frac{dN_1}{dt} = -W_{12}(N_1 - N_2) - \frac{N_1 - N_1^e}{\tau_1}$$
(V-190)

$$\frac{dN_2}{dt} = W_{12}(N_1 - N_2) - \frac{N_2 - N_2^e}{\tau_2}$$
(V-191)

where the transition rate is given by

$$W_{12} = \frac{\pi}{6} \frac{\mu^2 |E_0|^2}{\hbar^2} g(\omega - \omega_0)$$
(V-192)

Quantum mechanical origin of homogeneous line broadening

In (V-190) and (V-191) we recognize the rate equations (V-129a-b) derived previously from solving Schrödinger's equation (perturbatively) for an individual atom. The lineshape $g(\omega - \omega_0)$ introduced in (V-115) in an *ad hoc* manner is now given by (V-187). Substitution of (V-187) into (V-121) results in the explicit mathematical expression of the transition cross section between nondegenerate levels in terms of properties of the individual atoms (dipole matrix element μ) and the ensemble (relaxation times τ_1 , τ_2 and T_2)

$$\sigma(\omega) = \frac{\mu^2 \omega T_2}{3\epsilon_0 \hbar nc} \frac{1}{1 + (\omega - \omega_0)^2 T_2^2} = \frac{\sigma_0}{1 + (\omega - \omega_0)^2 T_2^2}$$
(V-193)

One of the major benefits of the density matrix approach is that it sheds light on the origin of homogeneous line broadening and delivers the explicit expression of the transition cross section of a homogeneously-broadened transition.

Connection between the atomic susceptibility and transition cross-section

The atomic susceptibility for a transition between nondegenerate levels can now be reexpressed in terms of $\sigma(\omega)$ by comparing (V-185) and (V-186) with (V-193)

$$\chi''(\omega) = \frac{nc}{\omega} \sigma(\omega) \Delta N \tag{V-185'}$$

$$\chi'(\omega) = \frac{nC}{\omega} (\omega_0 - \omega) T_2 \sigma(\omega) \Delta N$$
(V-186')

In the steady state the left-hand side and hence the right-hand side of Eqs. (V-190) and (V-191) are zero. Subtracting (V-191) from (V-190) leads to

$$\Delta N = \frac{\Delta N^{e}}{1 + W_{12}(\tau_1 + \tau_2)} = \frac{\Delta N^{e}}{1 + I/I_s(\omega)}$$
(V-194)

where

$$I_{s}(\omega) = \frac{\hbar\omega}{\sigma(\omega)(\tau_{1} + \tau_{2})} \tag{V-195}$$

is the *saturation intensity* introduced previously for the limiting case of $\tau_1 \rightarrow 0$ by Eq. (V-143).

Nonlinear constitutive law, power broadening

Substitution of Eqs. (V-195) and (V-194) into (V-185') and (V-186') yields (exercise)

$$\chi''(\omega) = \frac{\mathcal{NC}\sigma_0 \Delta N^e}{\omega} \frac{1}{1 + \frac{I}{I_s(\omega_0)} + (\omega - \omega_0)T_2^2}$$
(V-185")

$$\chi'(\omega) = \frac{\Pi C \sigma_0 \Delta N^e}{\omega} \frac{(\omega_0 - \omega) T_2}{1 + \frac{I}{I_s(\omega_0)} + (\omega - \omega_0) T_2^2}$$
(V-186")

The consequence of saturation implying a decrease of the population difference ΔN with increasing field intensity is that the susceptibility becomes field dependent, resulting in a *nonlinear constitutive law* (V-184). This nonlinearity implies a decreasing susceptibility χ and a broadening of the Lorentzian lineshape function with increasing field intensity. The broadening with respect to the zero-field bandwidth $\Delta \omega_0 = 2/T_2$ amounts to

$$\Delta \omega_{\text{sat}} = \Delta \omega_0 \sqrt{1 + \frac{I}{I_s(\omega_0)}}$$
(V-196)

This phenomenon is called power broadening.

Range of validity of the atomic susceptibility and the rate-equation approximation

The steady-state solution to the density matrix equations allowed us to derive the atomic susceptibility and the rate equations for level populations. Now we address the important question: what conditions have to be met in order for the susceptibility properly accounts for the atoms' response to the interacting field and the rate equations properly describe population dynamics between atomic levels?



induced polarization response:



To answer this question, we first scrutinize the transient response of the atoms to a field oscillating at ω_0 that is turned on abruptly at *t* = 0

$$E_{X}(t) = \begin{cases} \frac{1}{2} E_{0} e^{-i\omega_{0}t} + C.C. & t > 0\\ 0 & t < 0 \end{cases}$$
(V-197)

Solution of (V-177) under the assumption of ΔN constant in time yields the transient response of atomic polarization

$$P_{\chi}(t) \approx i \frac{K \Delta N T_2}{2\omega_0} \left[1 - e^{-t/T_2} \right] \frac{1}{2} E_0 e^{-i\omega_0 t} + C.C. =$$

= $\varepsilon_0 \chi(\omega_0) \left[1 - e^{-t/T_2} \right] \frac{1}{2} E_0 e^{-i\omega_0 t} + C.C.$ _(V-198)

in the form of a forced sinusoidal oscillation the amplitude of which builds up to a steady-state value with a time constant T_2 (Fig. V-22).

Fig. V-22

From this transient response we may conclude that the envelope P(t) of the atomic polarization $P_x(t) = \operatorname{Re}[P(t)\exp(-i\omega t)]$ will follow any amplitude or phase variation in the (complex) envelope E(t) of the driving electric field $E_x(t) = \operatorname{Re}[E(t)\exp(-i\omega t)]$ according to $P(t) = \varepsilon_0 \chi E(t)$ with a transient time delay that is $\approx T_2$.

The quasi-static approximation

$$P(t) = \varepsilon_0 \chi E(t)$$

of the atomic polarization response and its direct implication: the rate-equation approximation (Eqs. V-190, V-191) are therefore valid approximations if

(1) the driving electric field amplitude E(t) is nearly constant over the time scale of T_2

$$\frac{d|E|}{dt} \frac{1}{|E|} \ll \frac{1}{T_2} \tag{V-199}$$

and

(2) the population difference $\Delta N(t)$ is nearly constant over the time scale of T_2

$$\frac{d|\Delta N|}{dt} \frac{1}{|\Delta N|} \ll \frac{1}{T_2} \tag{V-200}$$

which, by subtraction of (V-191) from (V-190), can be reformulated as

$$W_{12} = \frac{\sigma}{\hbar\omega} I \ll \frac{1}{T_2}$$
(V-201)

that is, if the bandwidth of the interacting light field is small compared with the atomic linewidth $\Delta \omega_0$ and its strength is sufficiently small to ensure $W_{12} \ll 1/T_2$ (note that the latter condition also depends on the carrier frequency ω because of $W_{12} = W_{12}(\omega)$.

Is saturation feasible within the range of validity of the rate-equation approach? Only if condition #(2) can be reconciled with that of saturation: $W_{12}(\tau_1 + \tau_2) >> 1$, i.e.

$$\frac{1}{\tau_1} \text{ or } \frac{1}{\tau_2} \ll \frac{\sigma}{\hbar\omega} I \ll \frac{1}{T_2}$$
(V-202)

This condition for saturation within the rate-equation approximation can be satisfied only if

$$\tau_1 \text{ or } \tau_2 \gg T_2$$
 (V-203)

is fulfilled, which is the case in virtually all practically useful laser materials: the energy relaxation rates are almost always slow as compared to the dephasing rate.

Large-signal response: Rabi flopping

Let us now examine what happens if the interacting field is so strong that (V-201) is violated, that is

$$\frac{\sigma(\omega)}{\hbar\omega}I > \frac{1}{T_2}$$

Clearly, this situation can only be accounted for by the original density matrix equations of motion (Eqs. V-177,178,179), because one of the conditions for the validity of the rate equations (V-178,179) are not met.

To shed light on the strong-signal behaviour of resonantly driven atoms at the expense of a minimum of maths, we shall make a few simplifying, though in terms of the essence of the underlying physics – not limiting, assumptions. We assume an on-resonance applied driving field $E_x(t) = \operatorname{Re}[E(t)\exp(-i\omega_0 t)]$ with a real envelope function E(t) and write the polarization response in the form $P_x(t) = \operatorname{Re}[iP(t)\exp(-i\omega_0 t)]$, which will ensure P(t) to be a real function, too. Substituting these expressions into (V-177) we obtain for the polarization amplitude P(t)

$$-\frac{d^2 P(t)}{dt^2} + 2i(\omega_0 - \frac{1}{T_2})\frac{dP(t)}{dt} + 2i\frac{\omega_0}{T_2}P(t) = iK\Delta N(t)E(t)$$
(V-204)

Which, by using the slowly-varying amplitude approximation in time (see IV-133), and neglecting $1/T_2$ with respect to ω_0 , simplifies to

$$\frac{dP(t)}{dt} + \frac{1}{T_2}P(t) = \frac{K}{2\omega_0} \Delta N(t)E(t)$$
(V-205)

We also need an equation for the population difference ΔN , which can be obtained from (V-178) and (V-179). To simplify the analysis, we assume the two energy relaxation time constants to be equal

$$\tau_1 = \tau_2 \equiv T_1 \tag{V-206}$$

which, by subtraction of (V-179) from (V-178), leads to

$$\frac{d\Delta N(t)}{dt} + \frac{\Delta N(t) - \Delta N_0}{T_1} = -\frac{1}{\hbar} E(t) P(t)$$
(V-207)

where $\Delta N_{\rm e} = \Delta N_{\rm e}$ and we neglected quantities oscillating at 200 because they average out to zero on a time scale longer than the optical cycle.

We now solve Eqs (V-205) and (V-207) for the simple case of a constant driving signal abruptly turned on at t = 0 with an amplitude *E*₀, in which case they become a simple pair of coupled linear first-order differential equations. By substituting one of them into the other results in second-order equations for the polarization amplitude and the population difference

$$\left[\frac{d^2}{dt^2} + \left(\frac{1}{T_1} + \frac{1}{T_2}\right)\frac{d}{dt} + \left(\frac{1}{T_1T_2} + \omega_R^2\right)\right]\Delta N(t) = \frac{1}{T_1T_2}\Delta N_0 \qquad (V-208)$$

$$\left[\frac{d^2}{dt^2} + \left(\frac{1}{T_1} + \frac{1}{T_2}\right)\frac{d}{dt} + \left(\frac{1}{T_1T_2} + \omega_R^2\right)\right]P(t) = \frac{KE_0}{2\omega_0T_1}\Delta N_0 \qquad (V-209)$$

where

$$\omega_{R} = \sqrt{\frac{\mathcal{K}E_{0}^{2}}{2\hbar\omega_{0}}} = \begin{cases} \frac{\mu E_{0}}{\hbar} & \text{for fully oriented atoms} \\ \frac{\mu E_{0}}{\sqrt{3}\hbar} & \text{for randomly oriented atoms} \end{cases}$$
(V-210)

is the *Rabi frequency*, which is proportional to the driving field strength E_0 and the dipole matrix element μ .





Let us now suppose that Rabi frequency is small compared to ω_0 (so that the slowly-varying amplitude approximation remains valid) but large compared to the energy and phase relaxation rates $1/T_1$ and $1/T_2$ either because E_0 is extremely high or because the relaxation times are very long. Equations (V-208) and (V-209) then reduce to

$$\frac{d^2 \Delta N(t)}{dt^2} + \omega_R^2 \Delta N(t) = 0 \qquad (V-211)$$

and

$$\frac{d^2 P(t)}{dt^2} + \omega_R^2 P(t) = 0 \qquad (V-212)$$

which have the elementary solutions

$$\Delta N(t) = \Delta N_0 \cos \omega_R t \qquad (V-213)$$

and

$$P(t) = \sqrt{K\hbar/2\omega_0} \Delta N_0 \sin \omega_R t =$$

= $P_0 \sin \omega_R t$ (V-214)

Shown in Fig. V-23 for two different values of E₀.

It is apparent from these solutions, which are valid on a time scale much shorter than T_1 and T_2 , that the atomic behaviour in this strong-signal limit is very different from that in the rate-equation limit. Whereas the population difference ΔN never changes sign in the rate equation limit but merely its magnitude $|\Delta N|$ gets reduced with respect to its equilibrium value ΔN^e due to saturation according to (V-194), it oscillates at ω_R between ΔN_0 and $-\Delta N_0$ in the strong-field limit. At the same time,

the induced polarization amplitude, instead of approaching its steady state value $P_{ss} = \varepsilon_0 \chi E_0$, also oscillates with an amplitude that is independent of the driving field strength E_0 .

This oscillatory behaviour of the population difference ΔN and the polarization amplitude P(t) has been named the *Rabi* flopping behaviour after its inventor and occurs during an initial time interval short compared to either T_1 or T_2 , before they are damped with these time constants and approach their steady state value

$$\Delta N_{ss} = \frac{\Delta N_0}{1 + \omega_R^2 T_1 T_2}; \quad P_{ss} = \varepsilon_0 \chi E_0$$
(V-215a,b)

which have been previously also derived from the rate equation analysis. Comparison of (V-215a) with (V-194) and (V-206) yields the useful relation

$$W_{12} = \frac{\omega_R^2 T_2}{2}$$
 (V-216)

Coherent light amplification by stimulated emission

The mathematical treatment of stimulated emission on the basis of the rate equation approach [in particular: Eqs. (V-129c) or (V-128c)] does not provide answer to the important question whether stimulated emission allows coherent amplification of a light wave.

By coherent amplification we mean a process that connects the complex amplitudes of a light wave

$$E(z,t) = \frac{1}{2}E(z)e^{i(kz-\omega t)} + c.c.$$

0

at the input (z = 0) and the output (z = L) of the gain medium according to

$$\frac{E(z=L)}{E(z=0)} = |A| e^{i\varphi}; \quad |A| > 1$$
(V-217)

ensuring *both* |E(Z = L)| > |E(Z = 0)| *and* a constant phase relationship between these amplitudes.

In the rate-equation approach, we had to assume that the photons emitted as a result of stimulated emission have the same properties (momentum, energy, polarization) as the photons inducing the downward atomic transition to obtain Eq. V-122b.

However, even this assumption leads "only" to relationship $\frac{F_{out}}{F_{in}} = e^{\gamma L_g}$, which merely implies that

$$\frac{|E(Z=L)|^2}{|E(Z=0)|^2} = e^{\gamma L_g} = |A|^2$$
(V-218)

with no information being provided about the phase relationship between the input and output wave. The density matrix analysis resolves these shortcomings of the previous modelling in a natural way: it deliverers the polarization response of the

atomic medium (constitutive law); introducing the atomic susceptibility into Maxwell's wave equation allows to determine the back-effect of the atomic transitions on the wave inducing these transitions.

Connection between the gain coefficient, atomic susceptibility and transition cross section

To become more quantitative, let us consider the practically important case of the interacting atoms being imbedded in some condensed-phase medium (most frequently by doping atoms into a solid-state host). The overall polarization can then be decomposed into a resonant component $P_{\text{transition}}$ due to the specific atomic transition and a non-resonant component P_{host} arising from the polarizability of the host medium: $P = P_{\text{host}} + P_{\text{transition}}$, so that the relative permittivity of the laser medium, which enters the wave equation (IV-22), becomes

$$\varepsilon_{r}(\omega) = 1 + \chi_{host}(\omega) + \chi(\omega) = n^{2}(\omega) + \chi(\omega)$$
(V-219)

where $n(\omega)$ is the refractive index of the host medium. The light wave carried at a frequency resonant with the atomic transition

$$E(z,t) = \frac{1}{2} E_0 e^{i(k'z - \omega t)} + C.C.$$
 (V-220)

will then have a wave vector

$$k' = \frac{\omega}{c} \sqrt{\varepsilon_r} = \frac{\omega}{c} \sqrt{n^2(\omega) + \chi(\omega)} \approx k \left(1 + \frac{\chi}{2n^2} \right) = k \left(1 + \frac{\chi'(\omega)}{2n^2} \right) + i \frac{k\chi''(\omega)}{2n^2}$$
(V-221)

where $k = (\omega/c)n(\omega)$ is the wave vector in the absence of the atomic resonance and we have utilized that $|\chi| << 1$, which follows from a concentration of the doping atoms is low as compared with the density of atoms of the host material. Substituting (V-221) into (V-220), we find that in the presence of the atomic resonance, the wave propagates according to

$$E(z,t) = \frac{1}{2}E(z)e^{i(kz-\omega t)} + c.c. \quad ; \qquad E(z) = E_0 e^{\frac{\gamma}{2}z+i\Delta kz}$$
(V-222)

where

$$\Delta k(\omega) = \frac{k\chi'(\omega)}{2n^2}$$
(V-223)

and (by making use of V-185')

$$\gamma(\omega) = -\frac{\omega \chi''(\omega)}{nc} = -\sigma(\omega)\Delta N = \sigma(\omega)(N_2 - N_1)$$
(V-224)

Equation (V-222) clearly satisfies the requirements of coherent amplification: the resonant atomic polarization causes a change of the phase delay per unit length by Δk and – for $N_2 > N_1 \implies \Delta N < 0$ – causes the amplitude of the wave to vary exponentially with distance according to $\exp[(\gamma/2)z]$. Hence we may conclude that **stimulated emission results in coherent amplification of a light wave**. This holds true even if the wave amplitude becomes so strong that saturation occurs and thus the solution of the wave equation can not be given in the simple closed form represented by Eqs. (V-222), (V-223) and (V-224).

Coherent amplification by stimulated emission

$$E(z) = E_0 e^{\frac{\gamma}{2}z + i\Delta kz}$$



Fig. V-24

Saturation of homogeneously- and inhomogeneously-broadened transitions

Energy relaxation and dephasing imply that atomic transitions always possess a finite width; the Lorentzian line shape

$$\chi''(\omega) \propto 1 + \frac{1}{1 + \frac{I}{I_s(\omega_0)} + (\omega - \omega_0)^2 T_2^2}$$
(V-185")

derived from the density matrix analysis has a power-dependent line width

$$\Delta \omega = \Delta \omega_0 \sqrt{1 + \frac{I}{I_s(\omega_0)}}; \quad \text{where } \Delta \omega_0 = \frac{2}{T_2}$$
(V-196)

and the dephasing time T_2 defines the time scale characteristic for the loss of atomic coherence. As the lineshape defined by (V-185") is to be assigned to each individual atom with the atoms being indistinguishable, this type of broadening is referred to as *homogeneous broadening*.

Depending on the sign of the equilibrium population difference ΔN^e the frequency dependence of either absorption or the gain of a laser medium is affected. Here we focus on the effect of line broadening on the behaviour of a laser transition during saturation. For a homogeneously broadened laser transition, the gain coefficient is given by

$$\gamma(\omega) = -\frac{\omega \chi''(\omega)}{nc} = \sigma(\omega) \Delta N^{e} \frac{1}{1 + \frac{I}{I_{s}(\omega)}}$$
(V-225a)

with

$$\sigma(\omega) = \sigma_0 \frac{1}{1 + (\omega - \omega_0)^2 T_2^2} \quad \text{and} \quad I_s(\omega) = \frac{\hbar \omega}{\sigma(\omega)(\tau_1 + \tau_2)} \quad (V-225b,c)$$

and is responsible for the linear output-power-versus-pump-power relationship of a homogeneously-broadened laser, see Eqs. (V-149a-c).

In an inhomogeneous atomic system the individual atoms are distinguishable, with each atom having its own transition frequency $(E_2 - E_1)/\hbar$. The probability of finding and atom with its centre frequency between ω_{ξ} and $\omega_{\xi} + d\omega_{\xi}$, that is with $\omega_0 = \omega_{\xi}$ in (V-225) is $p(\omega_{\xi})d\omega_{\xi}$, resulting in an overall *inhomogeneously broadened* gain coefficient (**exercise**)

$$\gamma_{\text{inh}}(\omega) = \int_{-\infty}^{+\infty} \rho(\omega_{\zeta}) \gamma^{\zeta}(\omega) d\omega_{\zeta} = \frac{\sigma_0 \Delta N^e}{T_2^2} \int_{-\infty}^{+\infty} \frac{\rho(\omega_{\zeta}) d\omega_{\zeta}}{\frac{1}{T_2^2} + \frac{\sigma_0(\tau_1 + \tau_2)I}{\hbar \omega T_2^2} + (\omega - \omega_{\zeta})^2} \quad (V-226)$$

If the width of the distribution $p(\omega_{\xi})$ of transition frequencies is much broader than the homogeneous linewidth (V-196), we may pull $p(\omega_{\xi})_{\omega_{\xi}=\omega} = p(\omega)$ outside the integral sign in (V-226). Furthermore, making use of the identity

$$\int_{-\infty}^{+\infty} \frac{dx}{a^2 + x^2} = \frac{\pi}{a}$$
(V-227)

we obtain the gain coefficient for a laser transition that is dominantly inhomogeneously broadened (exercise)

$$\gamma_{\text{inh}}(\omega) = \frac{\sigma_0 \Delta N^e \pi \rho(\omega)}{T_2} \frac{1}{\sqrt{1 + \frac{I}{I_{s,\text{inh}}}}}$$
(V-228a)

where

$$I_{s,\text{inh}} = \frac{\hbar\omega}{\sigma_0(\tau_1 + \tau_2)} \tag{V-228b}$$

is the saturation intensity of the inhomogeneous line. Comparison of (V-228) with (V-225) reveals to essential differences:

- (1) In contrast with a homogeneously broadened transition the saturation intensity in the inhomogeneous case does not depend on the frequency of the interacting radiation.
- (2) The inhomogeneous atomic ensemble saturates more "slowly" as indicated by the square root in (V-228a). Although the population inversion within a "homogeneous packet" saturates as in (V-225), this saturation is partly compensated by the fact that the number of interacting atoms (contributing to the gain at the frequency of the incident light wave) increases due to power broadening (V-196).
- (3) Gain saturation in the inhomogeneous system *burns a "hole"* in the plot of the gain coefficient as a function of frequency the width of which is dictated by the power-broadened homogeneous linewidth given by (V-196), whereas saturation of the homogeneous line does not affect the gain profile, merely reduces its magnitude. Fig. V-25 compares the spectral profile of saturated gain, γ(ω), with that of unsaturated gain, γ₀(ω), as

"seen" by a weak probing signal of frequency ω ', for a homogeneously-broadened (a) and inhomogeneouslybroadened (b) transition, both saturated with a strong signal at ω .



