

Modern Atomic and Optical Physics II

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Chapter 4

Propagation of light in a resonant medium

We have now developed a formalism to describe coherent and incoherent interactions between atoms and light. These techniques will prove useful in understanding how a resonant light beam propagates through an atomic cloud. In particular, we have previously derived how the applied field induces coherences and transitions in the atomic system; if the system is to be self-consistent, we must also consider the effect of the induced atomic dipole back on the field.

An atom influences the applied field through its dipole moment

$$\langle \hat{d} \rangle = \mu \rho_{21} + \mu^* \rho_{12}. \quad (4.1)$$

A single atom has a very weak effect on a classical field, but a large ensemble of atoms can drastically affect light propagation, so we might seek to identify a macroscopic polarization associated with N atoms in a volume V by

$$P = \frac{N}{V} (\mu \rho_{21} + \mu^* \rho_{12}), \quad (4.2)$$

and solve for the density matrix components using the single-atom optical Bloch equations. However, a large ensemble of atoms may only be treated using our single-atom formalism if the atoms evolve independently. The assumption of independent atoms is valid provided that

(1) The atoms do not interact strongly on the timescales of the processes we wish to describe.

(2) Each atom is damped by its own reservoir. More quantitatively, the atoms must be far enough apart $|r_i - r_j| > \lambda$ that we may neglect interference from two atoms interacting with the same mode of the reservoir.

(3) The applied field is classical.

Mathematically, atoms comprising an atomic ensemble are independent when the ensemble density matrix may be factorized into a direct product of individual atomic density matrices,

$$\hat{\rho}_{ensemble} = \prod_{j=1}^N \hat{\rho}^{(j)} \otimes \dots \otimes \hat{\rho}^{(N)}. \quad (4.3)$$

Since our fields may vary in space as well as time, it is convenient to define a macroscopic polarization,

$$P(r_i) = \frac{N_i}{V_i} \sum_{r_j \in V_i} (\mu \rho_{21}(r_j) + \mu^* \rho_{12}(r_j)), \quad (4.4)$$

which depends on the total dipole moment of the set of N_i atoms at positions $\{r_j\}$ within a small volume V_i centered at position r_i . Using the optical Bloch equations, we can calculate the polarization for a given field; self-consistency requires that this polarization then act as a source term in the Maxwell equations governing light propagation through the atomic cloud:

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} \quad (4.5)$$

$$\nabla \times \mathbf{B} = \frac{1}{c^2} \frac{\partial}{\partial t} \left(\mathbf{E} + \frac{1}{\epsilon_0} \mathbf{P} \right). \quad (4.6)$$

In principle, we could now self-consistently calculate the evolution of the atomic medium and applied light field. In practice, we will typically deal with nearly monochromatic light fields, for which a considerable simplification can be obtained by using the slowly-varying envelope approximation.

4.1 The slowly-varying envelope approximation

We can rewrite the Maxwell equations (in the absence of unbound charges and currents) as a wave equation for \mathbf{E} :¹

$$\nabla^2 \mathbf{E} - \frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \mathbf{P}}{\partial t^2}. \quad (4.10)$$

¹Using $\nabla \times (\nabla \times \mathbf{A}) = \nabla(\nabla \cdot \mathbf{A}) - \nabla^2 \mathbf{A}$ and the curl of Eq. (4.5) we get

$$\nabla \times (\nabla \times \mathbf{E}) = \nabla(\nabla \cdot \mathcal{E}) - \nabla^2 \mathbf{E} \quad (4.7)$$

$$= -\frac{1}{c^2} \frac{\partial^2}{\partial t^2} \left(\mathbf{E} + \frac{1}{\epsilon_0} \mathbf{P} \right). \quad (4.8)$$

The wave equation is second order in time and space, but application of the slowly varying envelope approximation can reduce it to first order. An electric field which is nearly monochromatic can be parameterized by an envelope function and its central frequency $\bar{\nu}$,

$$\mathbf{E}(r, t) = \mathcal{E}(r, t)e^{i\bar{k}z - i\bar{\nu}t} + c.c., \quad (4.11)$$

where \mathcal{E} varies slowly in z and t compared to the optical frequency $\bar{\nu}$ and wavevector $\bar{k} = \bar{\nu}/c$. We also introduce a slowly varying polarization amplitude,

$$\mathbf{P}(r, t) = \mathcal{P}(r, t)e^{i\bar{k}z - i\bar{\nu}t} + c.c. \quad (4.12)$$

Substituting these expressions into Eq. (4.10) and keeping only the lowest order derivatives of the envelopes, we find the evolution equation for the slowly varying amplitudes,

$$\frac{1}{2i\bar{k}}\nabla_{\perp}^2\mathcal{E} + \frac{\partial\mathcal{E}}{\partial z} + \frac{1}{c}\frac{\partial\mathcal{E}}{\partial t} = \frac{i\bar{k}}{2\epsilon_0}\mathcal{P} \quad (4.13)$$

The derivation of Eq. (4.13) governing the slowly varying amplitudes is straightforward. Using the derivatives of the electric field and polarization:

$$\nabla^2\mathbf{E} = \left(\nabla^2\mathcal{E} + 2i\bar{k}\frac{\partial}{\partial z}\mathcal{E} - \bar{k}^2\mathcal{E} \right) e^{i\bar{k}z - i\bar{\nu}t} + c.c. \quad (4.14)$$

$$\frac{\partial^2}{\partial t^2}\mathbf{E} = \left(\frac{\partial^2}{\partial t^2}\mathcal{E} - 2i\bar{\nu}\frac{\partial}{\partial t}\mathcal{E} - \bar{\nu}^2\mathcal{E} \right) e^{i\bar{k}z - i\bar{\nu}t} + c.c. \quad (4.15)$$

$$\frac{\partial^2}{\partial t^2}\mathbf{P} = \left(\frac{\partial^2}{\partial t^2}\mathcal{P} - 2i\bar{\nu}\frac{\partial}{\partial t}\mathcal{P} - \bar{\nu}^2\mathcal{P} \right) e^{i\bar{k}z - i\bar{\nu}t} + c.c. \quad (4.16)$$

we substitute into the wave equation to find

$$\nabla^2\mathcal{E} + 2i\bar{k}\frac{\partial}{\partial z}\mathcal{E} - \bar{k}^2\mathcal{E} - \frac{1}{c^2}\left(\frac{\partial^2}{\partial t^2}\mathcal{E} - 2i\bar{\nu}\frac{\partial}{\partial t}\mathcal{E} - \bar{\nu}^2\mathcal{E} \right) = \quad (4.17)$$

$$\frac{1}{\epsilon_0 c^2}\left(\frac{\partial^2}{\partial t^2}\mathcal{P} - 2i\bar{\nu}\frac{\partial}{\partial t}\mathcal{P} - \bar{\nu}^2\mathcal{P} \right). \quad (4.18)$$

In the absence of unbound charges, $\nabla \cdot \mathbf{E} = 0$, so we obtain the wave equation

$$\nabla^2\mathbf{E} - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{E} = \frac{1}{\epsilon_0 c^2}\frac{\partial^2}{\partial t^2}\mathbf{P}. \quad (4.9)$$

Taking $\bar{\nu} = c\bar{k}$, and noting that in the slowly varying envelope approximation

$$|\bar{\nu}\mathcal{P}| \gg \left| \frac{\partial}{\partial t}\mathcal{P} \right| \quad (4.19)$$

$$|\bar{\nu}\mathcal{E}| \gg \left| \frac{\partial}{\partial t}\mathcal{E} \right| \quad (4.20)$$

$$|\bar{k}\mathcal{E}| \gg \left| \frac{\partial}{\partial z}\mathcal{E} \right|, \quad (4.21)$$

$$(4.22)$$

the relevant terms reduce to

$$\nabla_{\perp}^2 \mathcal{E} + 2i\bar{k} \frac{\partial}{\partial z} \mathcal{E} + \frac{1}{c^2} 2i\bar{\nu} \frac{\partial}{\partial t} \mathcal{E} = -\frac{1}{\epsilon_0 c^2} \bar{\nu}^2 \mathcal{P}, \quad (4.23)$$

or

$$\frac{1}{2i\bar{k}} \nabla_{\perp}^2 \mathcal{E} + \frac{\partial}{\partial z} \mathcal{E} + \frac{1}{c} \frac{\partial}{\partial t} \mathcal{E} = \frac{i\bar{k}}{2\epsilon_0} \mathcal{P}. \quad (4.24)$$

Two properties of this equation should be noted:

(1) In the first term, $\nabla_{\perp}^2 = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}$ leads to transverse effects such as focussing or diffraction which cannot in general be neglected. However, for a plane wave there is no transverse spatial variation so the first term vanishes and we obtain

$$\frac{\partial}{\partial z} \mathcal{E} + \frac{1}{c} \frac{\partial}{\partial t} \mathcal{E} = \frac{i\bar{k}}{2\epsilon_0} \mathcal{P}. \quad (4.25)$$

(2) The slowly varying polarization \mathcal{P} is proportional to the positive-frequency component of the dipole operator in the rotating frame. For example, a two level atom leads to a polarization $\mathcal{P} = (N/V)\mu\rho_{21}(z)e^{-i\bar{k}z}$. (Note that $\rho_{21}(z)$ itself is driven by the local electric field, which contains a fast phase term $e^{i\bar{k}z}$ so that \mathcal{P} is in fact slowly varying in space.)

Generalizations of the SVEA

Our derivation of the slowly varying envelope approximation can be generalized to include multiple optical frequencies. To illustrate this idea, we will consider the example of atomic interactions with an optical frequency comb. This periodic train of short optical pulses can be represented in the Fourier domain by a comb of harmonics, so that the electric field may be written

$$E = \sum_n \mathcal{E}_n(z, t) e^{inkz - i\nu_n t}. \quad (4.26)$$

The spacing between harmonics $\nu_{n+1} - \nu_n$ determines the repetition rate of the pulses, while the bandwidth occupied by the set of harmonics determines

the duration of each pulse. Provided that each harmonic has an envelope $\mathcal{E}_n(z, t)$ which varies slowly in space and time, we can use an extension of the SVEA. Defining the polarization harmonics by

$$P = \sum_n \mathcal{P}_n(z, t) e^{inkz - i\nu_n t}, \quad (4.27)$$

the propagation equation becomes a set of equations for each harmonic component

$$\frac{1}{2i\bar{k}} \nabla_{\perp}^2 \mathcal{E}_n + \frac{\partial}{\partial z} \mathcal{E}_n + \frac{1}{c} \frac{\partial}{\partial t} \mathcal{E}_n = \frac{i\bar{k}}{2\epsilon_0} \mathcal{P}_n. \quad (4.28)$$

This treatment applies to arbitrarily short pulses, since there is no restriction on the number of harmonics included. Note, however, that in general the atomic response will act to couple different harmonics, i.e. \mathcal{P}_n will depend on all components $\mathcal{E}_{n'}$, so the coupled Maxwell-Bloch equations may be quite complicated to solve.

4.2 Linear optical propagation

The simplest possible case of light propagation in a resonant field consists of an applied field sufficiently weak that the atoms only respond to it linearly. We found earlier that when the applied field is weak, perturbation theory may be used to solve exactly for the atomic density matrix components in the Fourier domain. We shall take a similar approach here, where the Fourier components of the polarization may be written in terms of the Fourier components of the off-diagonal elements of the (two-level) atomic density matrix,

$$\mathcal{P}(\delta\nu) = \frac{N}{V} \mu \rho_{21}(\delta\nu) e^{-i\bar{k}z}. \quad (4.29)$$

If the atoms respond linearly, the polarization must be proportional to the applied field. The proportionality constant defines the susceptibility,

$$\chi(\delta\nu) = \frac{\mathcal{P}(\delta\nu)}{\epsilon_0 \mathcal{E}(\delta\nu)}. \quad (4.30)$$

This quantity χ entirely characterizes the atom-photon interaction in the linear regime, and no further information is needed to find an exact solution.

For plane wave propagation (where transverse effects may be disregarded), the SVEA equation reduces to

$$\frac{\partial \mathcal{E}}{\partial z} + \frac{1}{c} \frac{\partial \mathcal{E}}{\partial t} = \frac{i\bar{k}}{2\epsilon_0} \mathcal{P}. \quad (4.31)$$

This first order equation may be trivially solved by moving to the Fourier domain,

$$\frac{\partial \mathcal{E}}{\partial z} = \frac{i\delta\nu}{c}\mathcal{E} + \frac{i\bar{k}}{2} \underbrace{\chi(\delta\nu)}_{\propto \mathcal{P}} \mathcal{E}, \quad (4.32)$$

where the frequency components of the electric field obey

$$\mathcal{E}(\delta\nu, z) = \mathcal{E}(\delta\nu, 0)e^{iz(\delta\nu/c + \bar{k}\chi(\delta\nu)/2)}. \quad (4.33)$$

By transforming back to the time domain, we obtain a general solution for linear propagation,

$$\mathcal{E}(t, z) = \int d(\delta\nu)e^{-i\delta\nu t} \mathcal{E}(\delta\nu, 0)e^{iz(\delta\nu/c + \bar{k}\chi(\delta\nu)/2)}. \quad (4.34)$$

This solution tells us that, given the spectrum of the field at some position in space, we can decompose the field into frequency components, propagate each frequency component separately, and then transform back to the time domain to obtain the full solution. Note that we only need $\chi(\delta\nu)$ to solve the linear propagation problem exactly. This expression encompasses three main effects:

- (1) Absorption arises from the imaginary part of the susceptibility.
- (2) Refraction is caused by the real part of the susceptibility.
- (3) Dispersion results from frequency dependence of the susceptibility.

4.2.1 Absorption and refraction

The effects of absorption and refraction can be illustrated by considering a resonant, continuous-wave field with a delta function frequency distribution, $\mathcal{E}(\delta\nu) \propto \delta(\delta\nu)$, so that

$$\mathcal{E}(t, z) = \mathcal{E}e^{i\bar{k}\chi(0)z/2}. \quad (4.35)$$

In general χ may be a complex quantity, and its real and imaginary parts have qualitatively different effects on the propagating field. In particular, $\text{Im}[\chi]$ leads to exponential attenuation or amplification of the beam intensity with distance at a rate

$$\alpha = \bar{k} \text{Im}[\chi(0)]. \quad (4.36)$$

The real component $\text{Re}[\chi]$ shifts the phase of the field linearly with distance, which can be understood as a modification of the wavevector $\bar{k} \rightarrow \bar{k} + \bar{k}\text{Re}[\chi]/2$, or a change in the index of refraction

$$n = 1 + \frac{\text{Re}[\chi(0)]}{2}. \quad (4.37)$$

Note that these formulae are only valid when the slowly varying envelope approximation holds, i.e. $|\text{Im}[\chi(0)]|, |\text{Re}[\chi(0)]| \ll 1$.

4.2.2 Dispersion

If the applied field has a finite bandwidth, the frequency dependence of the susceptibility begins to matter. For nearly monochromatic fields, the susceptibility may be expanded around its resonant value,

$$\chi(\delta\nu) \approx \chi(0) + \frac{d\chi}{d\nu}\delta\nu + \dots \quad (4.38)$$

Each term leads to different effects: the first corresponds to refraction, the second to group velocity, the third to group velocity dispersion, and so on. For example, if we keep only the second order terms, we find

$$\mathcal{E}(t, z) = \int d(\delta\nu) \mathcal{E}(\delta\nu, 0) e^{-i\delta\nu t} e^{iz(\delta\nu/c + k(\chi(0) + \frac{d\chi}{d\nu}\delta\nu)/2)} \quad (4.39)$$

$$= e^{iz\bar{k}\chi(0)/2} \int d(\delta\nu) \mathcal{E}(\delta\nu, 0) e^{-i\delta\nu(t-z/v_g)} \quad (4.40)$$

$$= e^{iz\bar{k}\chi(0)/2} \mathcal{E}(t - z/v_g, z = 0), \quad (4.41)$$

so the envelope propagates at the so-called group velocity

$$v_g = \frac{c}{1 + \frac{\bar{\nu}}{2} \frac{d\chi}{d\nu}}. \quad (4.42)$$

4.2.3 Two level systems

Consider light propagation through a dilute gas of identical, noninteracting two-level atoms. In the steady state, we can solve the optical Bloch equations for the off-diagonal density matrix elements to obtain the polarization and thus the susceptibility,

$$\chi(\delta\nu) = i \frac{N}{V} \frac{\mu^2}{\hbar\epsilon_0} (\rho_{11}^0 - \rho_{22}^0) \frac{1}{\gamma_{12} - i\delta\nu}. \quad (4.43)$$

Clearly, the susceptibility exhibits a Lorentzian lineshape with linewidth γ_{12} . Identifying the dipole moment with the spontaneous emission rate ($\gamma = \frac{\mu^2 k_0^3}{3\pi\epsilon_0\hbar}$ and $k_0 = 2\pi/\lambda$), we can also express the susceptibility as

$$\chi(\delta\nu) = i \frac{3}{8\pi^2} \frac{N}{V} \lambda^3 (\rho_{11}^0 - \rho_{22}^0) \frac{\gamma}{\gamma_{12} - i\delta\nu}. \quad (4.44)$$

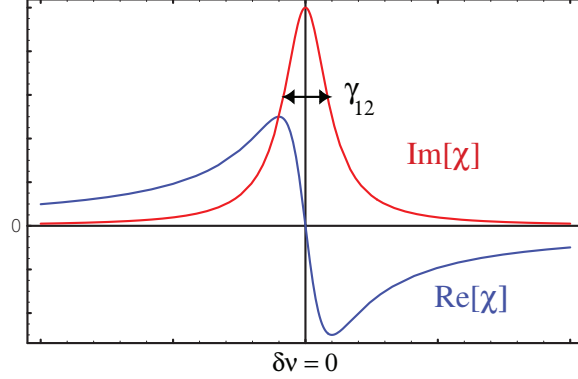


Figure 4.1: The real (refractive) and imaginary (absorptive) parts of the susceptibility for a two-level atom near resonance

Resonant fields

The magnitude of the susceptibility on resonance is

$$\chi(0) = i \frac{3}{8\pi^2} \frac{N}{V} \lambda^3 \frac{\gamma}{\gamma_{12}}. \quad (4.45)$$

Here we have set $\rho_{11}^0 = 1, \rho_{22}^0 = 0$, since it is in the regime where the upper state is negligibly populated that the atom responds linearly to the field. Since $\chi(0)$ is purely imaginary, there is no refraction on resonance, but there is absorption. The electric field amplitude propagates as

$$\mathcal{E}(z) = \mathcal{E}(0) e^{(-\text{Im}[\chi(0)] + i\text{Re}[\chi(0)])\bar{k}z/2} \quad (4.46)$$

and we define the corresponding intensity loss

$$I(z) = I(0) e^{-N\tilde{\sigma}z/V} \quad (4.47)$$

in terms of the absorption cross-section

$$\tilde{\sigma} = \frac{3}{4\pi} \lambda^2 \frac{\gamma}{\gamma_{12}} \quad (4.48)$$

$$\rightarrow \frac{3}{2\pi} \lambda^2 \text{ for purely radiative broadening.} \quad (4.49)$$

Surprisingly, in the case of radiative broadening the resonant cross section is independent of the atomic dipole moment and depends only on λ^2 ; this maximal absorption cross section is known as the “unitary limit”.

Far-detuned fields

Far from resonance, $\delta\nu \gg \gamma_{12}$, the imaginary (absorptive) component $\text{Im}[\chi] \propto 1/\delta\nu^2$ is much smaller than the real (reactive) component $\text{Re}[\chi] \propto 1/\delta\nu$, so the medium is almost entirely refractive and causes little absorption.

4.2.4 Line-broadening mechanisms

The existence of the unitary limit shows that different line broadening mechanisms (e.g. radiative vs non-radiative) can have different effects on the behavior of a system. It is particularly important to distinguish between processes which affects every atom equally and processes which broaden the linewidth of an ensemble but not its constituent atoms. The former “homogeneous” linewidth may not be visible in an ensemble exhibiting the latter “inhomogeneous” broadening. Two important examples of systems exhibiting inhomogeneous broadening are

(1) Optical emitters (e.g. color centers or quantum dots) in a solid state environment. Local fields have a strong influence on the optical transitions, so each emitter will exhibit different transition strengths and frequencies.

(2) Atoms with a thermal velocity distribution. The Doppler effect shifts their resonant frequency, so each atom interacts differently with applied fields.

Doppler broadening

To illustrate how inhomogeneous broadening affects linear optical propagation, we will examine the Doppler broadened atoms in more detail. In particular, we consider atoms in a thermal gas which are illuminated in the z direction by a field of frequency ν . Each atom experiences an apparent frequency $\nu - kv_z + \mathcal{O}(v^2)$ which is Doppler shifted due to the atomic velocity v_z . Equivalently, one can view this as a shift in the atomic resonance frequency $\omega \rightarrow \omega + kv_z$. For a Doppler-broadened medium an incident light beam will only be on resonance with a subset of atoms with the appropriate velocity class, but the absorption spectrum will be very broad, with width on order kv_T where v_T is the characteristic velocity corresponding to the atomic temperature. In order to calculate the susceptibility, we will determine the atomic response of each velocity class, and then sum over all velocity classes. The susceptibility is thus approximately

$$\chi(\delta\nu) \approx \int dv_z \frac{n(v_z)}{\gamma_{12} - i(\delta\nu - kv_z)} \quad (4.50)$$

where the thermal velocity distribution is

$$n(v_z) = \frac{N}{V} e^{-v_z^2/v_T^2} \frac{1}{\sqrt{\pi}v_T}. \quad (4.51)$$

The integral in Eq. (4.50) cannot be solved exactly, but we will consider two limiting cases where it can be approximately evaluated. For very cold atoms, $\gamma_{12} \gg kv_T$, there is no inhomogeneous broadening, and the observed ensemble absorption spectrum is Lorentzian, matching the intrinsic atomic linewidth. In the opposite limit, inhomogeneous broadening dominates, and the ensemble spectrum is Gaussian with width $\sim kv_T$. Note that inhomogeneous broadening also reduces the $\delta\nu = 0$ cross section by $\sim \gamma_{12}/kv_T$, so that

$$\sigma_{inhom.}(0) = \frac{3}{4\pi} \lambda^2 \frac{\sqrt{\pi}\gamma}{kv_T}. \quad (4.52)$$

As a quantitative example, atomic Rb at $T = 300\text{K}$ has an inhomogeneous linewidth of $\sim 500\text{MHz}$, which is far greater than its radiative linewidth of $\gamma \approx 5\text{MHz}$. For a Maxwell-Boltzmann thermal distribution of velocities, the Doppler width scales as the square root of the temperature. By laser cooling atoms (for example in a magneto-optical trap), it is possible to reduce the Doppler broadening to less than the radiative linewidth.

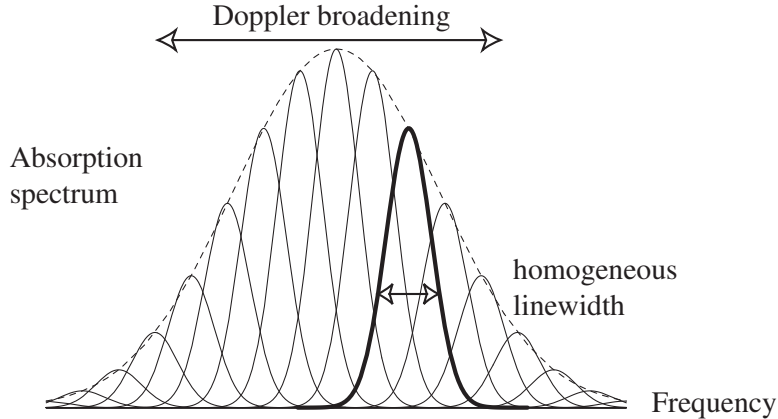


Figure 4.2: The absorption spectrum for a Doppler broadened ensemble of atoms is much broader than the absorption spectrum of a single velocity class, which has a linewidth set by the dephasing rate γ_{12} .

In general, however, it is impossible to tell whether an ensemble is homogeneously or inhomogeneously broadened with linear measurements. Such properties can be probed only by measuring the nonlinear response of the medium.

4.3 Nonlinear atomic response

The perturbative methods used in the linear regime cannot be used to treat strong applied fields. A general approach in the nonlinear regime requires a time-varying solution for the density matrix components as a function of the time-dependent applied field, $\Omega(t)$. As the system complexity increases, such solutions become increasingly difficult to obtain even numerically.

As a special case, we will consider only continuous-wave fields of fixed frequency, and look for the steady-state behavior of the system. To start with, consider a homogeneous ensemble of two-level atoms illuminated by photons with frequency detuned from atomic resonance by $\delta = \bar{\nu} - \omega$. Recall that the steady-state solution for the off-diagonal density matrix components is

$$\rho_{21} = -i \frac{\Omega}{\gamma_{12} - i\delta} (\rho_{11} - \rho_{22}). \quad (4.53)$$

For strong fields, we can no longer neglect changes in the atomic populations, so we should substitute (in the steady state)

$$\rho_{11} - \rho_{22} = \frac{\gamma}{\gamma + 2R_{opt}} \quad (4.54)$$

where

$$R_{opt} = \frac{2|\Omega|^2 \gamma_{12}}{\gamma_{12}^2 + \delta^2}. \quad (4.55)$$

We can use the steady state coherence to find the nonlinear susceptibility in the Fourier domain:

$$\chi_n(\delta) = \frac{\mathcal{P}(\delta)}{\epsilon_0 \mathcal{E}(\delta)} \quad (4.56)$$

$$= \frac{1}{\epsilon_0 \mathcal{E}(\delta)} \frac{N}{V} \mu \rho_{21}(\delta). \quad (4.57)$$

Proceeding in the same manner as before, we find that

$$\chi_n(\delta) = \frac{3}{8\pi^2} \frac{N}{V} \lambda^3 \frac{i\gamma}{\gamma_{12} - i\delta} \frac{\gamma}{\gamma + 2R_{opt}}. \quad (4.58)$$

Note that, whereas the linear susceptibility provided a complete solution to all time-dependent linearly propagating fields, the nonlinear susceptibility $\chi_n(\delta)$ we have just derived is applicable only to continuous wave, steady state solutions. For this special situation we are considering, the susceptibility determines the nonlinear propagation equation for the applied field. Since we are working with continuous wave fields, we may disregard the time derivatives in the wave equation, so the only remaining terms are

$$\frac{\partial \mathcal{E}}{\partial z} = i\bar{k}\chi_n(\delta)\mathcal{E} \quad (4.59)$$

$$= i\bar{k}\left(\frac{3}{8\pi^2}\frac{N}{V}\lambda^3\frac{i\gamma}{\gamma_{12}-i\delta}\frac{\gamma}{\gamma+2R_{opt}}\right)\mathcal{E}. \quad (4.60)$$

The nonlinear susceptibility is itself a function of the strength of the applied field $\Omega = \mu\mathcal{E}/\hbar$, which enters through R_{opt} . As the applied field grows, χ is diminished, an effect known as saturating absorption. In the limit of a weak field, $R_{opt} \ll \gamma$, this expression reduces to the linear susceptibility obtained earlier. We will now examine the nonlinear susceptibility in a few special cases.

Resonant fields

For a perfectly resonant field, $\delta = 0$, the real part of the susceptibility vanishes and the imaginary part becomes

$$\text{Im}[\chi_n(0)] = \frac{3}{8\pi^2}\frac{N}{V}\lambda^3\frac{\gamma}{\gamma_{12}}\frac{1}{1+s} \quad (4.61)$$

where

$$s = \frac{4|\Omega|^2}{\gamma_{12}\gamma}. \quad (4.62)$$

When this saturation parameter s is large, $s \gg 1$, the applied field bleaches the medium, and the transmitted power grows nonlinearly as the incident intensity is increased. In this saturated regime, the spatial profile of absorption changes qualitatively from a linear decay in strong fields to the exponential decay characteristic of weak fields.

Far-detuned fields

It is possible to define a saturation parameter as a function of the detuning,

$$s = \frac{2R_{opt}}{\gamma}. \quad (4.63)$$

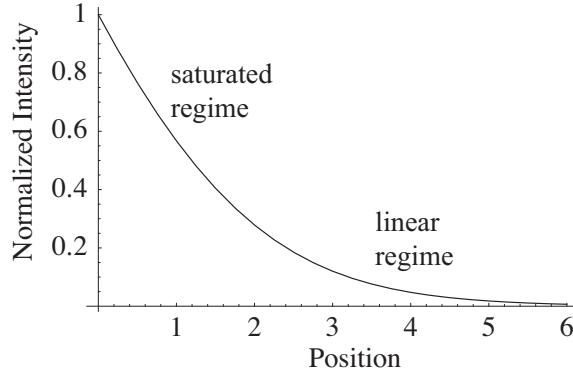


Figure 4.3: An incident field with intensity equal to the saturation intensity initially decays linearly with distance. Once the field is sufficiently weak that nonlinear effects are negligible, the intensity decays exponentially.

Since R_{opt} decreases as detuning increases, greater power is required to saturate an off-resonant transition. For very large detunings, $\delta \gg \Omega, \gamma$, the imaginary part of the susceptibility is negligible, with the real part given by

$$\chi_n \approx -\frac{3}{8\pi^2} \frac{N}{V} \lambda^3 \frac{\gamma}{\delta} \left(1 - \frac{4|\Omega|^2 \gamma_{12}}{\delta^2 \gamma} \right). \quad (4.64)$$

The refractive index of the medium is determined by $\text{Re}[\chi]$, and consequently depends on the intensity. The physics of an intensity dependent refractive index (known as the Kerr effect) may be captured by defining an index which is linear in intensity, $n = n_0 + n_2 I$. Such effects have important physical consequences, including intensity-dependent phase shifts which are often referred to as “self-phase modulation”. In addition, the medium will act as a lens for a beam with finite transverse extent, since the refractive index is a function of power and thus transverse position, leading to so-called self-focussing or de-focussing.

Weakly saturated regime

In some situations we will be working with relatively weak fields, and wish only to calculate the leading order corrections to linear propagation. In such cases, it is useful to expand the susceptibility

$$\chi_n = \chi + \chi^{(3)} |E|^2 + \dots, \quad (4.65)$$

where χ is the linear susceptibility and $\chi^{(3)}$ is the next order correction (at least for a system exhibiting inversion symmetry). In terms of the susceptibility for the two-level system, this expansion looks like:

$$\begin{aligned}\chi_n(\delta) &= \frac{3}{8\pi^2} \frac{N}{V} \lambda^3 \frac{i\gamma}{\gamma_{12} - i\delta} \frac{\gamma}{\gamma + 2R_{opt}} \\ &\approx \frac{3}{8\pi^2} \frac{N}{V} \lambda^3 \frac{i\gamma}{\gamma_{12} - i\delta} \left(1 - \frac{2R_{opt}}{\gamma}\right) \\ &\approx \frac{3}{8\pi^2} \frac{N}{V} \lambda^3 \frac{i\gamma}{\gamma_{12} - i\delta} - \left(\frac{3\lambda^3}{2\pi^2\hbar^2} \frac{N}{V} \frac{i}{\gamma_{12} - i\delta} \frac{|\mu|^2\gamma_{12}}{\gamma_{12}^2 + \delta^2}\right) |E|^2,\end{aligned}$$

where we identify the first term as the linear susceptibility and the second term as the leading nonlinear correction.

Experimental considerations

For practical purposes, it is useful to estimate the intensity needed to saturate a transition at resonance. In a radiatively broadened system, $\gamma_{12} = \gamma/2$, $R_{opt} = 4|\Omega|^2/\gamma$, so the saturation threshold $s = 1$ corresponds to $8|\Omega|^2 \approx \gamma^2$, or an intensity of a few mW/cm² for the alkali atoms. In a Doppler broadened system, $kv_T \gg \gamma_{12}$, a much larger power is required, since an applied field satisfying $8|\Omega|^2 \approx \gamma^2$ will only saturate a velocity class of atoms. However, in this intermediate regime where a small subset of velocity groups are saturated, nonlinear spectroscopy such as pump-probe or hole-burning experiments may be performed with sub-Doppler resolution.

In practice, real systems can saturate much faster than these calculations would predict because the existence of other atomic levels means that the system doesn't necessarily decay back into its original state. For instance, consider an atom with two long-lived hyperfine levels and a third excited level. When a field is applied which is resonant with the transition from one of the hyperfine levels to the excited state, most of the atomic population will end up in the other hyperfine level, an effect known as optical pumping. Now saturation of the resonant transition only requires that R_{opt} exceed the (slow) decay between hyperfine levels, making it much easier to saturate. This example illustrates how the two-level model is often not a very good approximation, especially in situations related to nonlinear interactions of atoms with light.

4.3.1 Saturation spectroscopy

Saturation spectroscopy is an important example of incoherent nonlinear multi-photon interactions. It provides a simple method for probing the natural linewidth of an inhomogeneously broadened medium (unlike linear interactions which cannot distinguish between a homogeneously and inhomogeneously broadened resonance). Saturation spectroscopy is typically performed with two counterpropagating beams at the same frequency ν , near resonance with the atomic transition energy Δ . The beams create an electric field profile in the cloud of thermal atoms,

$$\mathbf{E} = \mathcal{E}_+ e^{-i\nu t + ikz} + \mathcal{E}_- e^{-i\nu t - ikz} + c.c., \quad (4.66)$$

where \mathcal{E}_+ represents a strong field in the forward direction and \mathcal{E}_- represents a weak field in the backward direction.

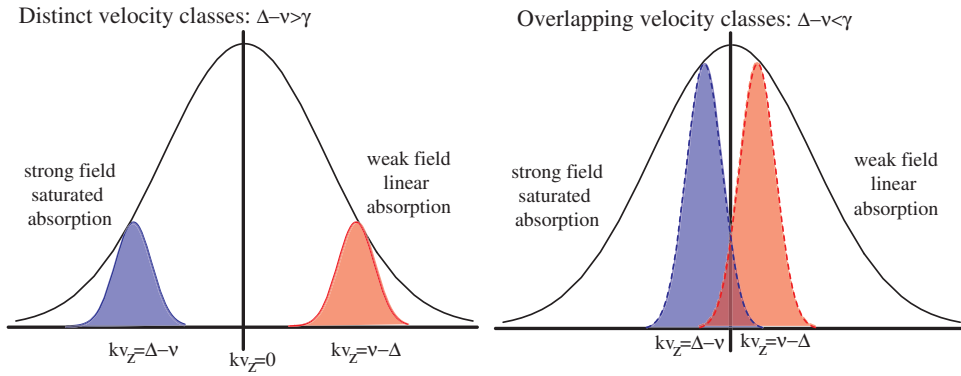


Figure 4.4: The forward and backward propagating beams interact with the velocity class of atoms for which they are resonant. The velocity classes begin to overlap as the optical frequency ν approaches the bare atomic resonance

Due to Doppler shifts in the thermal atoms, these two beams each interact with a certain velocity class. The strong field interacts with atoms satisfying $\nu = \Delta + kv_z$, saturating their transition, while the weak field interacts with atoms Doppler shifted by $\nu = \Delta - kv_z$. For generic detunings, $\nu \neq \Delta$, the two fields do not interact with the same group of atoms. As the frequency approaches the bare atomic resonance, however, $v_z \rightarrow 0$ and the two velocity classes begin to overlap. On resonance, both fields interact with stationary atoms. Consequently, we expect that the absorption

spectrum for the weak field \mathcal{E}_- will show sharp features near $\nu = \Delta$ due to stationary atoms saturated by the strong field.

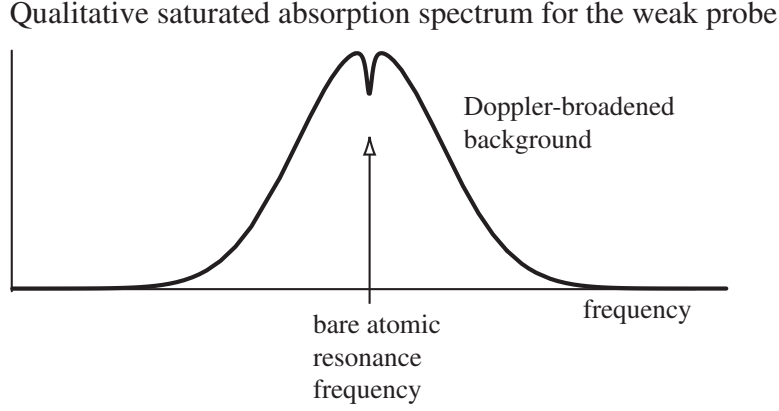


Figure 4.5: The absorption spectrum for the weak probe in the presence of a counterpropagating strong field of the same frequency.

The saturated absorption spectrum

The qualitative picture of saturated spectroscopy may be verified quantitatively by calculating the absorption coefficient for the probe beam, $\alpha_- = k\text{Im}[\chi_-] = k\text{Im}[P_-/\epsilon_0\mathcal{E}_-]$. To separate out the forward and backward propagating fields, we decompose the atomic polarization into two running waves in the $\pm\hat{z}$ direction,

$$P = P_+e^{-i\nu t+ikz} + P_-e^{-i\nu t-ikz} + c.c. \quad (4.67)$$

In order to account for the inhomogeneous Doppler broadening, this total polarization can be further broken down into contributions from atoms with different velocities. A particular velocity group v_z contributes a polarization $P^{(v_z)}$ proportional to the atomic coherence induced by the two counterpropagating beams,

$$\rho_{21}^{(v_z)} = \underbrace{\rho_{21}^{(v_z)+}}_{\sim\mathcal{E}_+} + \underbrace{\rho_{21}^{(v_z)-}}_{\sim\mathcal{E}_-}. \quad (4.68)$$

Once we have calculated the polarization for the appropriate density $n(v_z)$ of atoms, $P_{\pm}^{(v_z)} = n(v_z)\mu^* \rho_{21}^{(v_z)\pm}$, we can sum over velocity classes and divide by $\epsilon_0 \mathcal{E}_{\pm}$ to obtain the probe beam susceptibility.

We can use our previous results from analysis of linear interactions to find this coherence $\rho_{21}^{(v_z)-}$ because the probe beam \mathcal{E}_- is weak. In particular, we find

$$\rho_{21}^{(v_z)-} \approx \frac{i\Omega_-(\rho_{11} - \rho_{22})^{(v_z)}}{\gamma_{12} - i(\delta + kv_z)}, \quad (4.69)$$

where $\Omega_{\pm} = \mu \mathcal{E}_{\pm} / \hbar$. Note that the detuning $\delta = \nu - \omega$ is Doppler shifted due to the atomic velocity along the axis of light propagation. For the probe, atoms propagating to the right see the field at a higher frequency (blueshifted) so the net detuning is $\delta + kv_z$.

Our previous linear analysis required no further calculations because the population distribution $\rho_{11} - \rho_{22}$ was independent of Ω_- for a sufficiently weak field. To accurately model the nonlinear aspects of saturated absorption spectroscopy, however, we must incorporate the effects of the strong field $|\Omega_+|^2$ on $\rho_{11} - \rho_{22}$. We saw earlier that strong, nonlinear interactions can saturate a transition by redistributing the atomic population:

$$(\rho_{11} - \rho_{22})^{(v_z)} = \frac{1}{1 + S(\delta - kv_z)}, \quad (4.70)$$

where the saturation parameter is

$$S(\delta - kv_z) = \frac{4|\Omega_+|^2(\gamma_{12}/\gamma)}{\gamma_{12}^2 + (\delta - kv_z)^2}. \quad (4.71)$$

Note that the sign of the Doppler shift is reversed for Ω_+ because it is propagating in the opposite direction to Ω_- . Substituting into Eq. (4.69), we obtain the backward-propagating coherence induced by Ω_+ and Ω_- in atoms with velocity v_z :

$$\rho_{21}^{(v_z)-} \approx \left(\frac{\gamma_{12}^2 + (\delta - kv_z)^2}{\gamma_{12}^2 + (\delta - kv_z)^2 + 4|\Omega_+|^2(\gamma_{12}/\gamma)} \right) \left(\frac{i\Omega_-}{\gamma_{12} - i(\delta + kv_z)} \right). \quad (4.72)$$

We are now prepared to calculate the absorption coefficient for the probe beam $\alpha_- = k \text{Im}[\chi_-] = k \text{Im}[P_-/\epsilon_0 \mathcal{E}_-]$ by summing over contributions to the polarization from all atomic velocity classes,

$$P_- = \int dv_z \mu^* \rho_{21}^{(v_z)-} n(v_z), \quad (4.73)$$

where each velocity class is weighted by the thermal distribution $n(v_z)$.

$$\chi_- = \frac{|\mu|^2}{\epsilon_0 \hbar} \int dv_z n(v_z) \frac{i}{\gamma_{12} - i(\delta + kv_z)} \left(1 - \frac{S(\delta - kv_z)}{1 + S(\delta - kv_z)} \right) \quad (4.74)$$

The first term describes the usual Doppler-broadened absorption of a weak probe; the second term describes the saturated effect we wish to calculate. By approximating the thermal velocity distribution as constant over the velocities which contribute to the saturated absorption dip, $n(v_z) = n$, we can calculate the integral of the second term in closed form. The resulting expression is rather convoluted, and to elucidate the effects of the strong field we will expand it to lowest order in Ω_+ :

$$\begin{aligned} \chi_- &\approx \frac{|\mu|^2}{\epsilon_0 \hbar} \int dv_z n(v_z) \frac{i}{\gamma_{12} - i(\delta + kv_z)} \left(1 - \frac{4|\Omega_+|^2(\gamma_{12}/\gamma)}{\gamma_{12}^2 + (\delta - kv_z)^2} \right) \\ &= \frac{|\mu|^2}{\epsilon_0 \hbar} \int dv_z n(v_z) \frac{i(\gamma_{12} + i(\delta + kv_z))}{\gamma_{12}^2 + (\delta + kv_z)^2} \left(1 - \frac{4|\Omega_+|^2(\gamma_{12}/\gamma)}{\gamma_{12}^2 + (\delta - kv_z)^2} \right) \end{aligned}$$

The saturated absorption feature is thus given by

$$\begin{aligned} \text{Im}[\chi_-]_{\text{sat}} &\approx -\frac{|\mu|^2}{\epsilon_0 \hbar} \int dv_z n \frac{\gamma_{12}}{\gamma_{12}^2 + (\delta + kv_z)^2} \frac{4|\Omega_+|^2(\gamma_{12}/\gamma)}{\gamma_{12}^2 + (\delta - kv_z)^2} \\ &= -\frac{|\mu|^2 n}{\epsilon_0 \hbar k} \frac{2\pi |\Omega_+|^2(\gamma_{12}/\gamma)}{(\gamma_{12}^2 + \delta^2)}, \end{aligned} \quad (4.75)$$

where we have used $n(v_z) = n$ and

$$\int_{-\infty}^{\infty} \frac{1}{1 + (x - a)^2} \frac{1}{1 + (x + a)^2} dx = \frac{\pi}{2(1 + a^2)}. \quad (4.76)$$

The expression for the saturated absorption feature, in the limit that Ω_+ is weak and $n(v_z)$ is slowly varying, indicates that we should see a sharp dip in the absorption with a characteristic width given by the Doppler-free linewidth γ_{12} .