Module 4 : Third order nonlinear optical processes

Lecture 22 : Saturation of absorption

Objectives

In this lecture we will study the following

- Saturation behavior of a homogeneously broadened two-level system.
- Hole burning in inhomogeneously broadened two-level system.
- Lamb dip spectroscopy.
- Its application to the frequency stabilization of laser.

Saturation of absorption

According to Einstein, a monochromatic electromagnetic field interacting with an atom can either induce a transition from its lower energy state to the excited state or from an excited state to the lower energy state depending upon its initial state of residence. In the former process, known as absorption (which can also be called induced absorption), a transition is accompanied by loss of a photon from the electromagnetic field and an increase in the occupation number of the excited state. In the latter case, called stimulated emission, a photon coherent with the electromagnetic field (e.m. field) is added to the field and the occupation numbers of the excited state goes down by 1.

The two processes are complementary in the sense they take and give photons in identical manner. Apart from these two processes, an atom in its excited state can spontaneously emit a photon within the characteristic mean life time of the excited state and relax to the lower state.

The induced absorption coefficient is given by the product of the absorption cross-section and the population difference of the two levels. Since the induced absorption redistributes the population difference, one expects it to reduce with increasing intensity and correspondingly the absorption coefficient of the medium will decrease. This is called the saturation of absorption.

Alternatively, we can understand the saturation of absorption due to the increased detuning between light and the matter at higher intensities due to the intensity dependent increase of the energy separation between the two arising from the dynamic Stark effect. To be able to fully appreciate the consequences of this phenomenon, we will develop a simple theoretical model.

We consider light interacting with a two-level system (see figure 22.1) with excited and ground states energies E_2, E_1, $E_2 - E_1 = \hbar\omega_0$. Let their occupation numbers be ρ_{22} and ρ_{11}, respectively. The rate equations for the occupation numbers of the excited and ground states in a closed 2-level system can be written as

$$\frac{\mathrm{d}}{\mathrm{d}t}\rho_{11} = \frac{\rho_{22}^{eq}}{T_1} - W_{12}\Delta\rho + \frac{\rho_{22}}{T_1} \tag{22.1}$$

And

$$\frac{\mathrm{d}}{\mathrm{d}t}\rho_{11} = -\frac{\rho_{22}^{eq}}{T_1} + W_{12}\Delta\rho - \frac{\rho_{22}}{T_1} \tag{22.2}$$

where $\Delta\rho = \rho_{11} - \rho_{22}$, $W_{12} = B_{12}u(\omega) = \frac{\pi\Omega_{12}^2}{\varepsilon_0}g(\omega)$ is the transition rate; $\Omega_{12} = \frac{1}{\hbar}\langle 1|e\hat{r}\cdot\hat{E}|2\rangle$ is the is the Rabi oscillation Frequency; $B_{12} = B_{21}$ is Einstein's B coefficient; T_1, is the population relaxation time or the longitudinal relaxation time;

$$g(\omega) = \frac{\gamma}{(\omega-\omega_0)^2 + \gamma^2}$$

is the line shape function of the transition and the superscript *eq* refers to the respective quantities in thermal equilibrium γ is the spectral width of the absorption line.

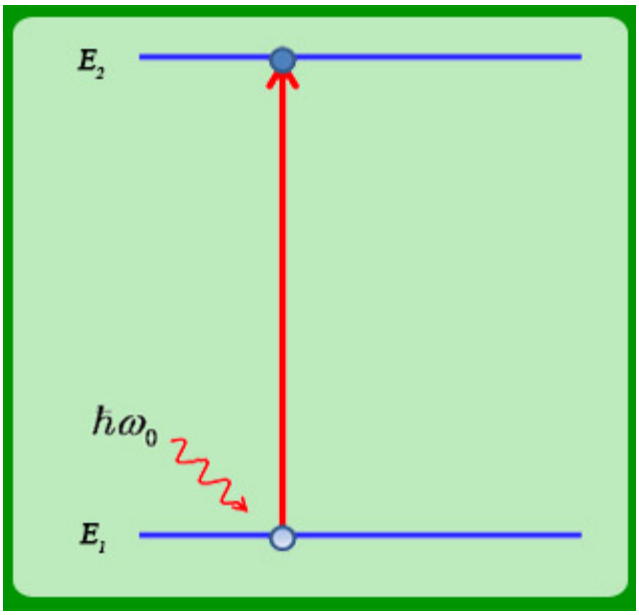


Figure 22.1 Resonant excitation of a two-level-system

In a closed 2-level system the total population density

$$\rho_t = \rho_{11} + \rho_{22} = constant \tag{22.3}$$

Using this equation we can write

$$\rho_{22} = \frac{\rho_t - \Delta\rho}{2} \tag{22.4}$$

While writing equations (22.1) and (22.2), we have ignored the coherence effects.

Subtracting the equations (22.2) from (22.1) and using equation (22.4), we get the rate equation for the population change

$$\left(\frac{\mathrm{d}}{\mathrm{d}t} + \frac{1}{T_1}\right)\left(\Delta\rho - \Delta\rho^{eq}\right) = -\frac{2\pi}{\hbar\epsilon_0}\Omega_{12}^2 \tag{22.5}$$

In steady state one can therefore, write

$$\left(\Delta\rho - \Delta\rho^{eq}\right) = \frac{\gamma^2\left(\frac{I}{I_s}\right)\Delta\rho^{eq}}{\left(\omega - \omega_0\right)^2 + \gamma^2\left(1 + I/I_s\right)} \tag{22.6}$$

where I is the intensity of the e. m. field and $I_s = \frac{1}{2}nc\varepsilon_0\frac{\gamma}{T_1}\left(\frac{|E|^2}{\Omega_{12}}\right)$ is the saturation intensity. It is obvious from equation(22.6) that as the intensity of the interacting field increases, the population difference decreases. At $I = I_s$, it reduces to half its initial value. The absorption coefficient of the pump beam $\alpha \propto W_{12}\Delta\rho$

Hence

$$\alpha(\omega) = \frac{\alpha_0\gamma^2}{\left(\omega - \omega_0\right)^2 + \gamma^2\left(1 + \frac{I}{I_s}\right)} \tag{22.7}$$

where α_0 is the linear absorption coefficient at the peak of the Lorenzian profile $(\omega = \omega_0)$ under the condition $I / I_s \to 0$. It can be seen from equation (22.7) that under saturation effect the unsaturated homogeneous line width γ broadens to

$$\gamma_s = \gamma\left(\sqrt{\left(1+\frac{I}{I_s}\right)}\right) \tag{22.8}$$

As $I / I_s \to \infty$, the absorption coefficient $\alpha(\omega) \to 0$ and the medium is completely bleached.In weak field limits, $I / I_s \ll 1$

$$\alpha(\omega) = \frac{\alpha_0 \gamma^2}{(\omega-\omega_0)^2+\gamma^2}\left[1-\frac{\gamma^2 I / I_s}{(\omega-\omega_0)^2+\gamma^2}\right] \tag{22.9}$$

Thus $\Delta\alpha = \alpha_0 - \alpha(\omega) \propto |E|^2$ and hence, saturation of absorption is a third order nonlinear effect. One can also notice that saturation effect is strongest at the line center.

In a gaseous medium spectral lines are inhomogeneously broadened due to the Doppler effect.When a laser beam of frequency w passes through a gaseous medium, a molecule moving with velocity v_z along the laser propagation direction its absorption frequency in the lab frame is shifted to

$$\omega_a = \omega_0 + kv_z \tag{22.10}$$

where ω_0 is the resonant frequency of the molecule in the rest frame.

Then only those molecules which are moving with velocity v such that the laser frequency falls within homogeneous width i.e. $\omega = \omega_a \pm \gamma$ will contribute to absorption of laser light.The population difference for this group of molecules in the two states is

$$\Delta\rho(v_z) - \Delta\rho^{eq}(v_z) = \frac{\gamma^2\left(\frac{I}{I_s}\right)\Delta\rho^{eq}(v_z)}{(\omega-\omega_0+kv_z)^2+\gamma^2\left(1+\frac{I}{I_s}\right)} \tag{22.11}$$

The equilibrium population difference $\Delta\rho^{eq}(v_z)$ is given by a Maxwell –Boltzmann distribution.

$$\Delta\rho^{eq}(v_z) \propto \frac{1}{\sqrt{\pi}v_p} e^{-\frac{v_z^2}{v_p^2}} \tag{22.12}$$

where v_p is the most probable speed.

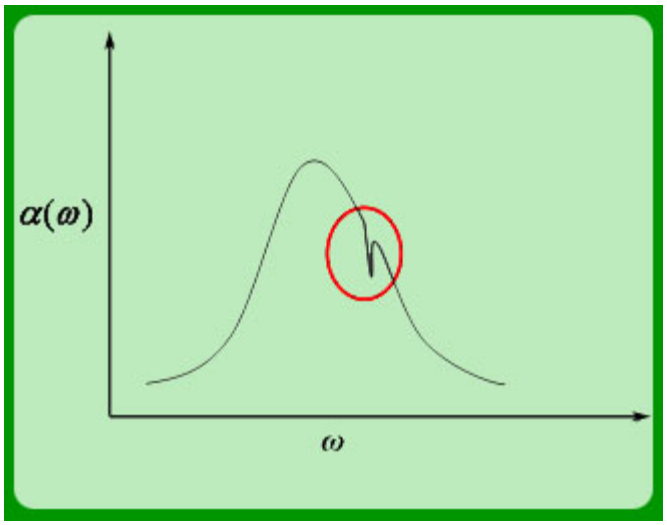


Figure 22.2 Hole burning in inhomogeneously broadened absorption line profile

Thus the appreciable pumping of population to an excited state will occur only for this group of molecules i.e

$$v_z = \frac{\omega - \omega_0}{k} \tag{22.13}$$

and consequently a dip appears in the population distribution and hence in the absorption profile as sketched in the figure 22.2. This is called the hole burning. The width of the hole is given by

$$\frac{\gamma}{k}\left(\sqrt{1+\frac{I}{I_s}}\right) \tag{22.14}$$

Lamb dip spectroscopy :

Let us consider the standing wave field created by two counter propagating laser beams of frequency ω in a gaseous medium so that their wave vectors are $\vec{k_1} = -\vec{k_2}$.

We will neglect the coherent effects in our discussion. These two beams burn two holes at $v_z = \pm\frac{(\omega - \omega_0)}{k}$ in the Doppler broadened profile. The corresponding population difference can be written as using equation (22.11)

$$\Delta\rho - \Delta\rho^{eq} = \left[\frac{\gamma^2}{(\omega - \omega_0 + kv_z)^2 + \gamma_s^2} - \frac{\gamma^2}{(\omega - \omega_0 - kv_z)^2 + \gamma_s^2}\right] \tag{22.15}$$

using I_s is the saturation intensity due to one beam, corresponding saturated absorption coefficient is written

$$\alpha(\omega) = \alpha_0(\omega)\left[1 - \frac{I_s}{2}\left(1 + \frac{\gamma_s^2}{(\omega - \omega_0)^2 + \gamma_s^2}\right)\right] \tag{22.16}$$

For $\omega \neq \omega_0$, incident wave is absorbed by molecules with velocity components

$$v_z = (\omega - \omega_0) / k \tag{22.17}$$

and

$$v_z = -(\omega - \omega_0) / k \tag{22.18}$$

For $\omega = \omega_0$ both waves are absorbed by molecules having $v_z = 0 \Rightarrow$ dip at $\omega = \omega_0$ in the Doppler broadened absorption profile as shown in figure (22.3).

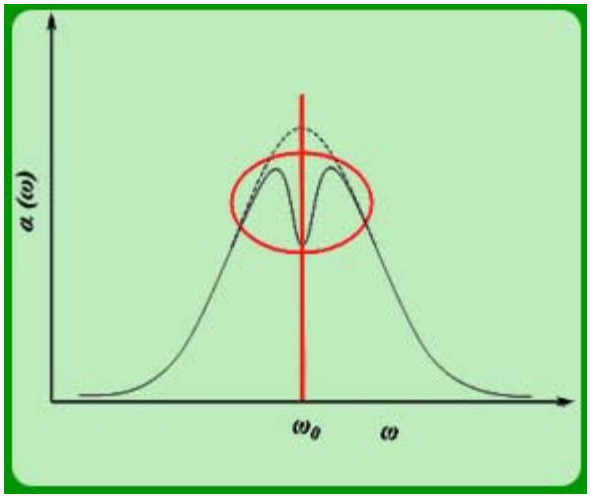


Figure 22.3 Lamb Dip

Doppler free absorption dip in the inhomogeneously broadened line profile is called the Lamb dip. Consider two absorption transitions from $|i\rangle$ to two closely spaced levels $|j\rangle$ and $|k\rangle$ so that their Doppler profiles overlap, if $\omega_{ij} - \omega_{ik} = \Delta\omega > 2\gamma_s$ then their Lamb dips can be completely separated.

- From the observed saturation dip, we can measure the homogeneous line width and hence the longitudinal relaxation time T_1.
- The Lamb dip is much narrower than the Doppler width and hence can be used to resolve the closely spaced lines hidden under the Doppler broadened profile.

Thus one can use the saturation phenomenon in inhomogeneous broadened systems to achieve high resolution Doppler free saturation spectroscopy.

For the experimental realization, a tunable laser beam is split into two counter propagating pump and probe beams. The pump beam is modulated for the measurement of transmission of the probe beam using a phase sensitive detection technique. This eliminates the Doppler profile background from the saturation of absorption signal.

The Doppler free absorption dip

$$\alpha_0 - \alpha_s = \frac{\alpha_0}{2}\frac{I}{I_s}\left(\frac{\gamma_s^2}{(\omega-\omega_0)^2+\gamma_s^2}\right) \qquad (22.19)$$

is thus obtained. A typical experimental set up is shown in figure 22.4. Coherent effects can affect the width and depth of the hole. These can be avoided by keeping incident laser intensity in the weak field limit.

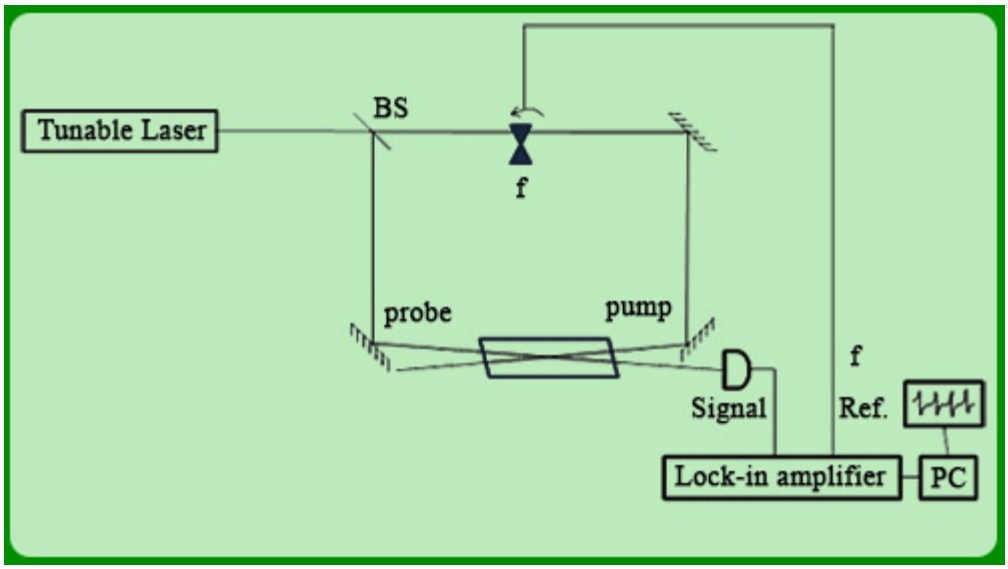


Figure 22.4 Lamb dip spectroscopy setup

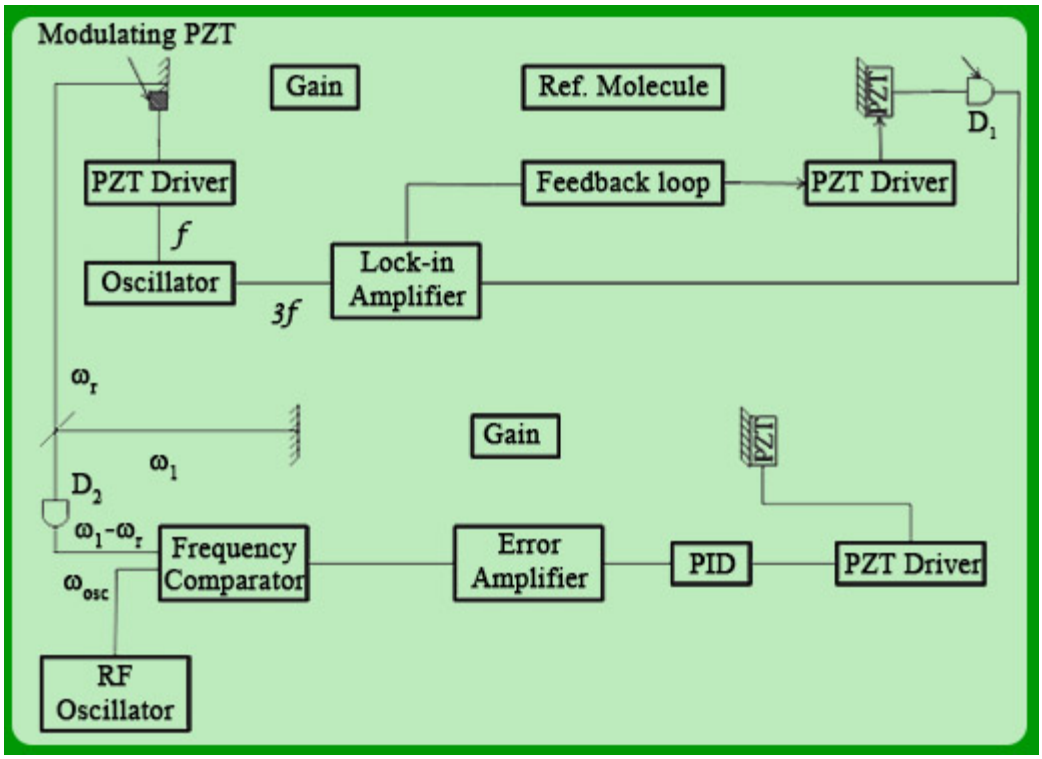
High spectral resolution possible by saturation spectroscopy can easily resolve hyperfine structure of the spectral lines which can provide valuable insight on fundamental interactions. Such high resolution spectroscopy, demands laser sources generating precise frequency radiation. Frequency drifts due to mode hopping in usual laser sources renders them inadequate for such applications. Special techniques are then required to lock the frequency of easers with precision and high stability.

In the next part we describe the use of saturation of absorption phenomenon itself for this purpose. Frequency stabilized lasers are also required for the length and time standards.

Laser frequency stabilization:

An atomic or molecular transition provides a convenient and stable reference for locking the frequency of a laser. High resolution intracavity lamb dip spectroscopy of an appropriate atomic or molecular sample can locate the molecular transition in the Doppler broadened profile to which the oscillation frequency of the laser can be locked. The output power of the laser depends upon the gain and Doppler broadened intracavity saturable absorber profiles. Inverted Lamb dip signal appears riding on the broad background in the laser output. Broad background can be eliminated by taking the third derivative of the Lamb dip signal and the frequency of transition peak can be located very precisely from the zero crossing of the third derivative. However a laser stabilized to a molecular transition will emit at fixed frequency only. Such a laser, though may be useful for the standards, is grossly inadequate for spectroscopic application due to lack of tunability.

To overcome this limitation one can combine this scheme with frequency offset locking technique. A typical scheme for this is shown in figure 22.5. Cavity of the reference laser is weakly modulated at some frequency, say *f*, through a piezoelectric actuator. The laser cavity is first tuned to the desired Lamb dip signal of the intra cavity molecular reference sample. Lamb dip is detected at 3*f* using a lock-in amplifier. The recorded signal corresponds to the third derivative of the Lamb dip signatures. Stability achieved by this technique can now be transferred to another tunable laser by the offset-locking technique.



A beat frequency signal is produced by mixing the output of the primary reference laser and the secondary tunable laser in a detector. The beat frequency signal is then compared with frequency ω_{osc} of a tunable and stable RF oscillator. An error signal is generated to control the tunable laser such that $\omega_l = \omega_r - \omega_{osc}$ at all times. This way the frequency of the tunable laser is locked to the offset frequency $\omega_l = \omega_r - \omega_{osc}$ which can thus be tuned simply by tuning the RF oscillator frequency, ω_{osc} .

Recap

In this lecture we have studied

1. Effect of radiation field intensity on the population difference, absorption coefficient and line width of a homogeneously broadened two-level system. It has been shown that the saturation of absorption is a third order nonlinear optical process and under saturation the absorption coefficient decreases with intensity and absorption line profile broadens.
2. Hole burning phenomenon in a inhomogeneously broadened two-level system.
3. High resolution Doppler free Lamb dip spectroscopy and
4. Its application to laser frequency stabilization.