

CHAPTER THREE

Investigation of absorption-dispersion relation with dipole-dipole interaction (on-resonance case)

3.1 Introduction

In recent years there has been a lot of interest in studying the absorption-dispersion relation for various atomic systems [3.1-3.5]. Ling and Barbay [3.4] have shown that the index of refraction can be enhanced at vanishing absorption by incorporating Doppler broadening. Quang and Freedhoff [3.6] have shown that a high index of refraction can be accompanied by vanishing absorption for a certain range of Rabi frequencies from coherently driven two level atoms. As a result, preparation of matter in such a state would provide us with a new type of optical material with many applications in fundamental and applied physics [3.2]. In particular, enhancement of index of refraction at zero absorption has a potential application with respect to laser particle acceleration [3.7], optical microscopy [3.8] and atomic tests of electroweak physics [3.9].

In this chapter, we investigate the absorption-dispersion relation from two collective atoms with the inclusion of dipole-dipole interaction driven by a resonant laser and damped by a normal vacuum.

3.2 Master equation with dipole-dipole interaction

The model that we consider is the two-atom coherently driven Dicke system [3.10] in which the atomic separations is assumed to be much smaller than the resonant wavelength and we take into account the dipole-dipole interaction between the two atoms. The master equation for the reduced atomic density operator which describes the atoms coherently driven and damped by a normal vacuum in a rotating frame at the laser frequency ω_L [3.11] is

$$\begin{aligned} \frac{\partial \rho}{\partial t} = & -i\Delta[S^z, \rho] - i\Omega[S^+ + S^-, \rho] - i\alpha_+[S^+S^-, \rho] \\ & + \frac{\gamma}{2}(2S^- \rho S^+ - S^+ S^- \rho - \rho S^+ S^-) \end{aligned} \quad (3.2.1)$$

where S^\pm, S^z are the usual collective spin operators, γ is the Einstein A coefficient and 2Ω is the Rabi frequency. α_+ is the static dipole-dipole interaction potential such that [3.12]

$$\alpha_+ \equiv \frac{3\gamma}{2(kr_{ij})^3} \left[1 - 3(\hat{\mu} \cdot \hat{r}_{ij})^2 \right] \quad (3.2.2)$$

where $\hat{\mu}$ and \hat{r}_{ij} are unit vectors along the transition electric dipole moment and the interatomic distance \hat{r}_{ij} , respectively; $k = \frac{2\pi}{\lambda_0}$ where λ_0 is the single-atom

resonance wavelength. The dipole-dipole interaction arises from the virtual photon exchange between pairs of atoms and becomes important only when the spacing between atoms becomes comparable with the transition wavelength [3.13]. The significant changes can be expected in the structure of the absorption-dispersion spectra. $\Delta = \omega_A - \omega_L - \alpha_+$, where ω_A is the atomic transition frequency. Clearly, if $\Delta = -\alpha_+$, the laser is tuned to the atomic transition frequency.

We now project equation (3.2.1) into the Hilbert space of the energy eigenstates $|S, m\rangle$ with $S = 1$ and $m = 0, \pm 1$, i.e. the Dicke states [3.10]. The collective operators S^2 and S^z satisfy

$$S^2|S, m\rangle = S(S+1)|S, m\rangle \quad (3.2.3)$$

$$S^z|S, m\rangle = m|S, m\rangle \quad (3.2.4)$$

where $m = -S, -S+1, \dots, S-1, S$. The atomic polarization operators S^+ and S^- are given by

$$S^+|S, m\rangle = \sqrt{V_{m+1}}|S, m+1\rangle \quad (3.2.5)$$

$$S^-|S, m\rangle = \sqrt{V_m}|S, m-1\rangle \quad (3.2.6)$$

where

$$V_{m+1} = (S+m+1)(S-m) \quad (3.2.7)$$

$$V_m = (S-m+1)(S+m) \quad (3.2.8)$$

The following relations have been used to obtain a complete set of coupled differential equations for the matrix elements of the atomic density operator (see later).

$$\begin{aligned}
|m\rangle &\equiv |S, m\rangle & , \quad \langle m|[S^z, \rho]|n\rangle &= (m-n)\rho_{m,n} \\
\langle m|S^+ \rho|n\rangle &= V_m^{\frac{1}{2}} \rho_{m-1,n} & , \quad \langle m|S^- \rho|n\rangle &= V_{m-1}^{\frac{1}{2}} \rho_{m-1,n} \\
\langle m|\rho S^+|n\rangle &= V_{n+1}^{\frac{1}{2}} \rho_{m,n+1} & , \quad \langle m|\rho S^-|n\rangle &= V_n^{\frac{1}{2}} \rho_{m,n-1} \\
\langle m|S^- \rho S^+|n\rangle &= (V_{m+1} V_{n+1})^{\frac{1}{2}} \rho_{m+1,n+1} & , \quad \langle m|S^+ \rho S^-|n\rangle &= (V_m V_n)^{\frac{1}{2}} \rho_{m-1,n-1} \\
\langle m|S^- \rho S^-|n\rangle &= (V_{m+1} V_n)^{\frac{1}{2}} \rho_{m+1,n-1} & , \quad \langle m|S^+ \rho S^+|n\rangle &= (V_m V_{n+1})^{\frac{1}{2}} \rho_{m-1,n+1} \\
\langle m|S^- S^+ \rho|n\rangle &= V_{m+1} \rho_{m,n} & , \quad \langle m|S^+ S^- \rho|n\rangle &= V_m \rho_{m,n} \\
\langle m|S^- S^- \rho|n\rangle &= (V_{m+1} V_{m+2})^{\frac{1}{2}} \rho_{m+2,n} & , \quad \langle m|S^+ S^+ \rho|n\rangle &= (V_m V_{m-1})^{\frac{1}{2}} \rho_{m-2,n} \\
\langle m|\rho S^- S^-|n\rangle &= (V_n V_{n-1})^{\frac{1}{2}} \rho_{m,n-2} & , \quad \langle m|\rho S^+ S^+|n\rangle &= (V_{n+1} V_{n+2})^{\frac{1}{2}} \rho_{m,n+2} \\
\langle m|\rho S^- S^+|n\rangle &= V_{n+1} \rho_{m,n} & , \quad \langle m|\rho S^+ S^-|n\rangle &= V_n \rho_{m,n}
\end{aligned}
\tag{3.2.9}$$

The master equation in terms of the matrix elements $\rho_{m,n}(t)$ of the reduced atomic density operator satisfy [3.14]

$$\begin{aligned}
\frac{\partial \rho_{m,n}}{\partial \tau} &= -i \frac{\Delta}{\gamma} (m-n) \rho_{m,n} - i \frac{\alpha_z}{\gamma} (V_m - V_n) \rho_{m,n} \\
&+ i \frac{\Omega}{\gamma} \left[V_m^{\frac{1}{2}} \rho_{m-1,n} + V_{m+1}^{\frac{1}{2}} \rho_{m+1,n} - V_{n+1}^{\frac{1}{2}} \rho_{m,n+1} - V_n^{\frac{1}{2}} \rho_{m,n-1} \right] \\
&+ \frac{1}{2} \left[2(V_{m+1} V_{n+1})^{\frac{1}{2}} \rho_{m+1,n+1} - (V_m + V_n) \rho_{m,n} \right]
\end{aligned}
\tag{3.2.10}$$

where $\tau = \gamma t$ is dimensionless time describing the evolution of the system and $\rho_{m,n} = \langle m|\rho|n\rangle$. The matrix elements $\rho_{m,n}(t)$ of the atomic density operator satisfy the matrix equation.,

where $\tau = \gamma t$ is dimensionless time describing the evolution of the system and $\rho_{m,n} = \langle m | \rho | n \rangle$. The matrix elements $\rho_{m,n}(t)$ of the atomic density operator satisfy the matrix equation.,

$$\frac{d\vec{\psi}}{d\tau} = L\vec{\psi} + \vec{Y} \quad (3.2.11)$$

where L is a non-singular 8×8 matrix and \vec{Y} is a column matrix; $\vec{\psi}$ is an eight-dimensional vector composed of the density matrix elements

$$\begin{aligned} \rho_{1,1} &\equiv \psi_1 & , & & \rho_{0,1} &\equiv \psi_4 & , & & \rho_{0,-1} &\equiv \psi_7 \\ \rho_{0,0} &\equiv \psi_2 & , & & \rho_{1,-1} &\equiv \psi_5 & , & & \rho_{-1,0} &\equiv \psi_8 \\ \rho_{1,0} &\equiv \psi_3 & , & & \rho_{-1,1} &\equiv \psi_6 & , & & \rho_{-1,-1} &\equiv \psi_9 \end{aligned} \quad (3.2.12)$$

where $\psi_9 = 1 - \psi_1 - \psi_2$. Equation (3.2.11) can be decomposed into eight coupled differential equations

$$\begin{aligned} \dot{\psi}_1 &= -2\psi_1 + \frac{i\Omega}{\gamma} \sqrt{2}(\psi_4 - \psi_3) \\ \dot{\psi}_2 &= -2\psi_2 + 2\psi_1 + \frac{i\Omega}{\gamma} \sqrt{2}(\psi_8 - \psi_7 - \psi_4 + \psi_3) \\ \dot{\psi}_3 &= -\frac{i\Omega}{\gamma} \sqrt{2}(\psi_1 - \psi_2 + \psi_5) - (2 + \frac{i\Delta}{\gamma})\psi_3 \\ \dot{\psi}_4 &= \frac{i\Omega}{\gamma} \sqrt{2}(\psi_1 - \psi_2 + \psi_6) + (\frac{i\Delta}{\gamma} - 2)\psi_4 \\ \dot{\psi}_6 &= \frac{i\Omega}{\gamma} \sqrt{2}(\psi_4 - \psi_8) - (1 - (\frac{2i\alpha_+}{\gamma} + \frac{2i\Delta}{\gamma}))\psi_6 \end{aligned}$$

$$\begin{aligned}\dot{\psi}_7 &= \frac{i\Omega}{\gamma} \sqrt{2} (1 - \psi_1 - 2\psi_2 + \psi_5) - \left(\frac{i\Delta}{\gamma} + 1 + 2 \frac{i\alpha_+}{\gamma} \right) \psi_7 + 2\psi_3 \\ \dot{\psi}_8 &= \frac{i\Omega}{\gamma} \sqrt{2} (2\psi_2 - \psi_6 + \psi_1 - 1) + \left(\frac{i\Delta}{\gamma} - 1 + 2 \frac{i\alpha_+}{\gamma} \right) \psi_8 + 2\psi_4\end{aligned}$$

(3.2.13)

The matrix L is

$$L = \begin{bmatrix} -2 & 0 & -\beta\sqrt{2} & \beta\sqrt{2} & 0 & 0 & 0 & 0 \\ 2 & -2 & \beta\sqrt{2} & -\beta\sqrt{2} & 0 & 0 & -\beta\sqrt{2} & \beta\sqrt{2} \\ -\beta\sqrt{2} & \beta\sqrt{2} & -\left(2 + \frac{i\Delta}{\gamma}\right) & 0 & -\beta\sqrt{2} & 0 & 0 & 0 \\ \beta\sqrt{2} & -\beta\sqrt{2} & 0 & -\left(2 - \frac{i\Delta}{\gamma}\right) & 0 & -\beta\sqrt{2} & 0 & 0 \\ 0 & 0 & -\beta\sqrt{2} & 0 & -\left(1 + 2\frac{i\Delta}{\alpha} + 2\frac{i\alpha_+}{\gamma}\right) & 0 & \beta\sqrt{2} & 0 \\ 0 & 0 & 0 & \beta\sqrt{2} & 0 & -\left(1 - 2\frac{i\Delta}{\alpha} - 2\frac{i\alpha_+}{\gamma}\right) & 0 & -\beta\sqrt{2} \\ -\beta\sqrt{2} & -2\beta\sqrt{2} & 2 & 0 & \beta\sqrt{2} & 0 & -\left(1 + \frac{i\Delta}{\alpha} + 2\frac{i\alpha_+}{\gamma}\right) & 0 \\ \beta\sqrt{2} & 2\beta\sqrt{2} & 0 & 2 & 0 & -\beta\sqrt{2} & 0 & -\left(1 - \frac{i\Delta}{\alpha} - 2\frac{i\alpha_+}{\gamma}\right) \end{bmatrix}$$

(3.2.14)

where $\beta = \frac{i\Omega}{\gamma}$, $\bar{\psi}$ and \bar{Y} are

$$\bar{\psi} = \begin{bmatrix} \psi_1 \\ \psi_2 \\ \psi_3 \\ \psi_4 \\ \psi_5 \\ \psi_6 \\ \psi_7 \\ \psi_8 \end{bmatrix} \quad \text{and} \quad \bar{Y} = \begin{bmatrix} 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ \beta \\ -\beta \end{bmatrix} \quad (3.2.15)$$

Laplace transforming equation (3.2.11) gives

$$\bar{\psi}(z) = M\bar{\psi}(0) + z^{-1}M\bar{Y} \quad (3.2.16)$$

where $M = (z - L)^{-1}$ and z is the Laplace transform variable. The steady-state solutions is thus

$$\begin{aligned} \bar{\psi}(\infty) &= \lim_{z \rightarrow 0} z\bar{\psi}(z) \\ &= -L^{-1}\bar{Y} \end{aligned} \quad (3.2.17)$$

The elements of the inverse of a non singular 8 X 8 matrix can be obtained by using a symbolic manipulating software like Mathematica and we have done this in the Appendix .

The exact steady-state solution of the master equation for two coherently driven identical two-level atoms with the inclusion of dipole-dipole interaction and

damped by a normal vacuum in the steady state can be solved exactly by setting the left hand side of the equations (3.2.13) to be zero [3.15]. It simply involves solving eight simultaneous equations. The steady-state expressions for the components of ψ are

$$\begin{aligned}
 \psi_1 &= \frac{4\Omega^4}{D} \\
 \psi_2 &= \frac{2\Omega^2}{D} (2\Omega^2 + 4\Delta^2 + 8\Delta\alpha_+ + 4\alpha_+^2 + \gamma^2) \\
 \psi_3 &= \psi_4^* = -\frac{2^{\frac{3}{2}}i\Omega^3}{D} (2i\Delta + 2i\alpha_+ - \gamma) \\
 \psi_5 &= \psi_6^* = \frac{2\Omega^2}{D} [2\Delta^2 - 3i\Delta(2i\alpha_+ - \gamma) + 4\alpha_+(\alpha_+ + i\gamma) - \gamma^2] \\
 \psi_7 &= \psi_8^* = \frac{\sqrt{2}}{D} i\Omega \left\{ i\Delta [-4\Delta^2 + i\Delta(16i\alpha_+ - 4\gamma) - 20\alpha_+^2 - 8i\alpha_+\gamma - \gamma^2] \right. \\
 &\quad \left. + (-8i\alpha_+^3 + 4\alpha_+^2\gamma - 2i\alpha_+\gamma^2 + \gamma^3) - 2\Omega^2(2i\Delta + 2i\alpha_+ - \gamma) \right\}
 \end{aligned} \tag{3.2.18}$$

where

$$\begin{aligned}
 D &= 2\Omega^2 [6\Omega^2 + 2(4\Delta^2 + 8\alpha_+ + 4\alpha_+^2 + \gamma^2)] + \Delta [4\Delta^3 + 24\Delta^2\alpha_+ + \Delta(52\alpha_+^2 + 5\gamma^2) \\
 &\quad + 48\alpha_+^3 + 12\alpha_+\gamma^2] + 16\alpha_+^4 + 8\alpha_+^2\gamma^2 + \gamma^4
 \end{aligned}$$

3.3 Absorption and dispersion spectrum

We now study the absorption and dispersion spectra of two two-level atoms in a normal vacuum. Absorption and dispersion spectra are represented by the real

We now study the absorption and dispersion spectra of two two-level atoms in a normal vacuum. Absorption and dispersion spectra are represented by the real and imaginary parts of a complex linear susceptibility [3.16]:

$$\Gamma(\omega) = \int_0^{\infty} d\tau \exp(-i\omega\tau) \lim_{t \rightarrow \infty} \langle [S^+(\tau+t), S^-(t)] \rangle \quad (3.3.1)$$

The two-time correlation function in equation (3.3.1) can be evaluated by using the quantum regression theorem [3.17]. For a quantum mechanical system which is Markovian, the one-time expectation values

$$\langle A(t) \rangle = \sum_{\mu} f_{\mu}(t, t') \langle A_{\mu}(t') \rangle \quad (3.3.2)$$

where $f_{\mu}(t, t')$ is a c-number function and $t > t'$, leads to the two-time correlation function

$$\langle A(t)B(t') \rangle = \sum_{\mu} f_{\mu}(t, t') \langle A_{\mu}(t')B(t') \rangle. \quad (3.3.3)$$

where A, B are arbitrary operators. In the present case, the one-time expectation values of $\langle S^+(t) \rangle$ can be expressed as

$$\begin{aligned} \langle S^+(t) \rangle &= \text{Tr}\{\rho(t)S^+\} = \exp(i\omega L t) \text{Tr}\{\tilde{\rho}(t)S^+\} \\ &= \sqrt{2}[\psi_4(t) + \psi_8(t)] \exp(i\omega L t) \end{aligned} \quad (3.3.4)$$

Applying the quantum regression theorem to equation (3.3.4) we can evaluate $\langle S^+(t+\tau)S^-(t) \rangle$, and in a similar manner $\langle S^-(t)S^+(t+\tau) \rangle$ which in turn results in equation (3.3.1) taking the form

$$\begin{aligned} \Gamma(\omega) = & \left[(M_{4,4} + M_{8,4}) (\psi_2(\infty) - \psi_1(\infty)) + (M_{4,6} + M_{8,6}) (\psi_8(\infty) - \psi_4(\infty)) \right. \\ & + (M_{4,2} + M_{8,2}) (\psi_7(\infty) - \psi_3(\infty)) + (M_{4,8} + M_{8,8}) (1 - 2\psi_2(\infty) - \psi_1(\infty) \\ & \left. - (M_{4,7} + M_{8,7}) \psi_5(\infty) + (M_{4,1} + M_{8,1}) \psi_3(\infty) + (M_{4,3} + M_{8,3}) \psi_5(\infty) \right]_{Z=i(\omega-i\gamma_L)} \end{aligned} \quad (3.3.5)$$

Note that $M_{4,4}$ etc. are the elements of the matrix M in equation (3.2.17) and are given in the Appendix . Equation (3.3.5) can be written as

$$\Gamma(\omega) = R(\omega) + i\chi(\omega) \quad (3.3.6)$$

The absorption spectrum is given by

$$R(\omega) = \text{Re} \int_0^\infty d\tau \exp(-i\omega\tau) \lim_{t \rightarrow \infty} \langle [S^+(\tau+t), S^-(t)] \rangle \quad (3.3.7)$$

The dispersion spectrum on the other hand is given by

$$\chi(\omega) = \text{Im} \int_0^\infty d\tau \exp(-i\omega\tau) \lim_{t \rightarrow \infty} \langle [S^+(\tau+t), S^-(t)] \rangle \quad (3.3.8)$$

3.4 Results and discussions

By using equation (3.3.7), we can calculate the absorption spectrum from two atoms with the inclusion of dipole-dipole interaction driven by a resonant laser. We begin with a coherent driving field with $\frac{\Omega}{\gamma} = 40$. In figure 3.4.1, the absorption spectrum without dipole-dipole interaction is symmetrical. In the presence of dipole-dipole interactions, (e.g. $\frac{\alpha_+}{\gamma} = 20$), we observe a splitting of the Rabi sideband of the absorption spectrum into symmetric doublets (see figure 3.4.2). The splitting is caused by the dipole-dipole interaction between the atoms. As we increase the dipole-dipole interaction (e.g. $\frac{\alpha_+}{\gamma} = 40$) the widths of the lines become modified as in figure 3.4.3. The negative values of the absorption spectrum mean amplification of the probe beam.

We now use the expression (3.3.8) to briefly study the dispersion spectrum from two atoms resonantly driven by a laser and damped by normal vacuum. When a strong coherent field is applied (e.g. $\frac{\Omega}{\gamma} = 40$) in the absence of dipole-dipole interaction, we observe a symmetrical dispersion spectrum as shown in figure 3.4.4. This spectrum belongs to the Lorentzian type. Large dispersions can be obtained on the right-hand side and the left-hand side of the central frequency. Note that the refractive index vanishes at the central frequency. In the presence of dipole-dipole interaction (e.g. $\frac{\alpha_+}{\gamma} = 20$), we observe the splitting of the Rabi side band of the

dispersion spectrum into doublets. We also observe additional Rabi side bands at both sides of the central frequency (see figure 3.4.5). All these effects are due to the dipole-dipole interaction between the atoms. Since, regarding figure (3.4.6) as compared with figure (3.4.5) we see that in both figures the higher peak is $O(10^{-3})$ (compared with $O(10^{-2})$ in case of no dipole-dipole of figure (3.4.4) but the second peak in figure (3.4.6) reduces in height and the third peak increases as a result of increasing the dipole-dipole strength, compared with figure (3.4.5).. Again, the index of refraction vanishes at the central frequency.

Now we report behaviour of the absorption-dispersion relation (see figures 3.4.7-3.4.10). Clearly for strong coherent driving fields (e.g. $\frac{\Omega}{\gamma} = 40$) with and without dipole-dipole interaction when the laser is tuned to the atomic transition frequency (i.e. $\Delta = -\alpha_+$), the possibility to obtain a large index of refraction accompanied by vanishing absorption is evident (see figures 3.4.7-3.4.9). However, interestingly when the laser is not tuned to the atomic transition frequency (i.e. $\Delta \neq -\alpha_+$), (e.g. $\frac{\alpha_+}{\gamma} = 40, \frac{\Delta}{\gamma} = -20$) such as in figure 3.4.10, we do not find large refractive index accompanied by vanishing absorption. We can therefore conclude that large dispersion accompanied by zero absorption is a feature unique for a strongly driven atomic system with dipole-dipole interaction only when the laser is tuned to the atomic transition frequency.

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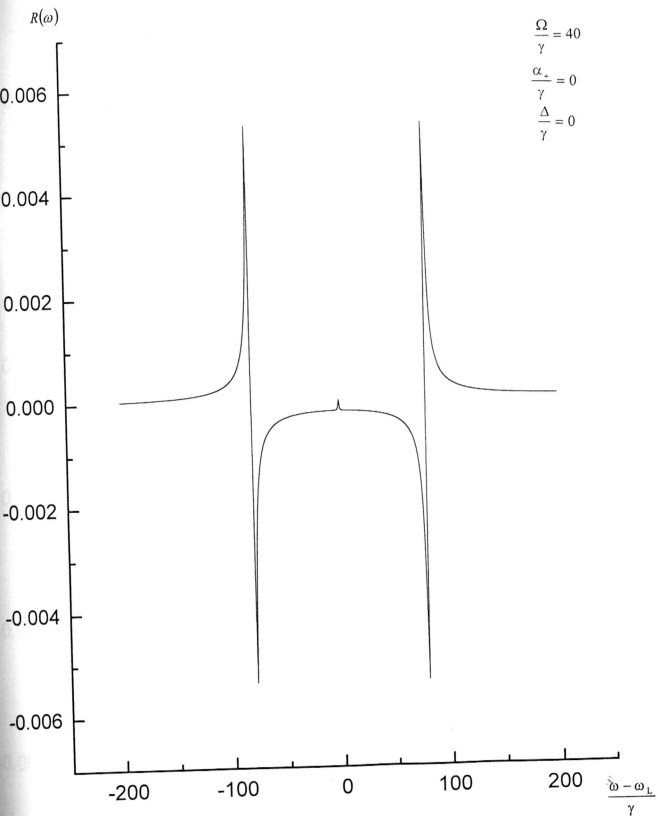


Figure 3.4.1 Plot of absorption spectrum for two atoms driven by a resonant laser.

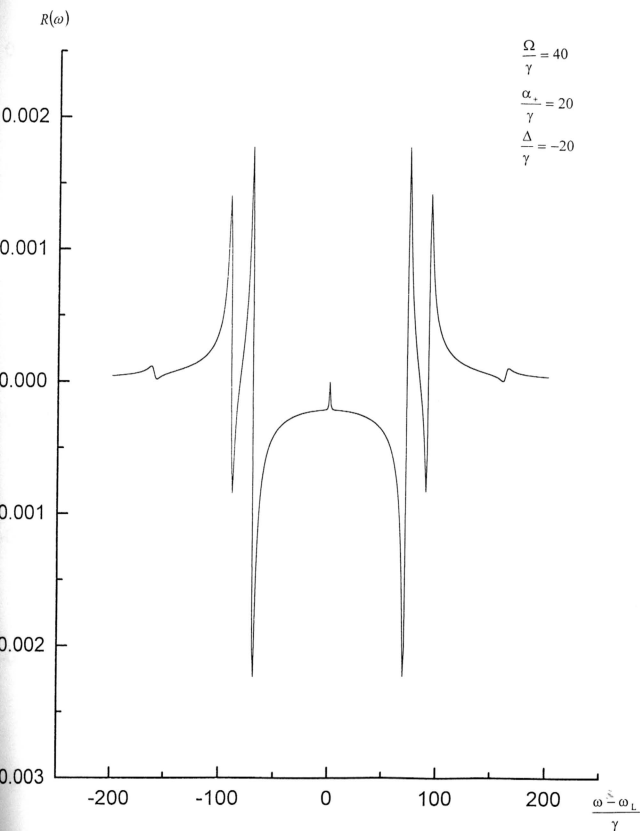


Figure 3.4.2 Plot of absorption spectrum for two atoms driven by a resonant laser.

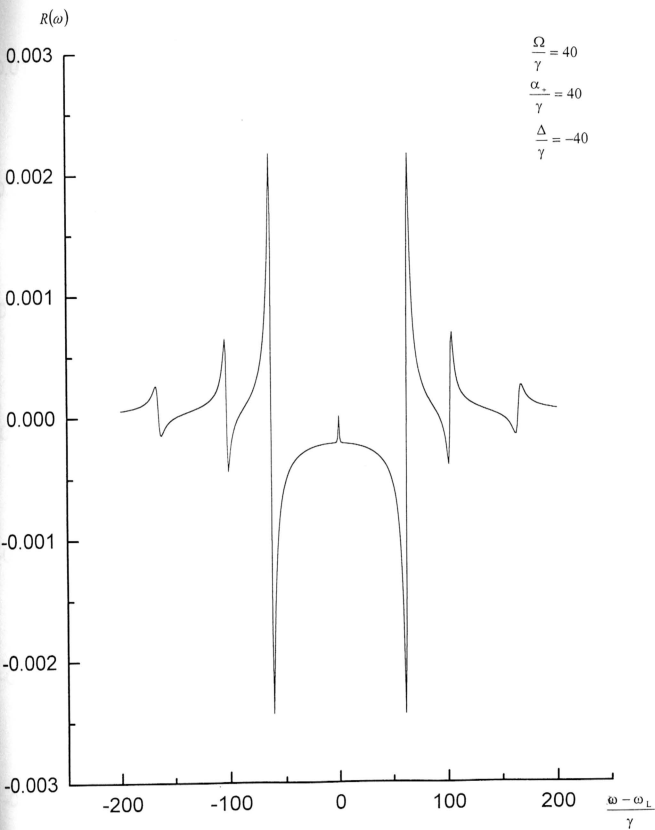


Figure 3.4.3 Plot of absorption spectrum for two atoms driven by a resonant laser.

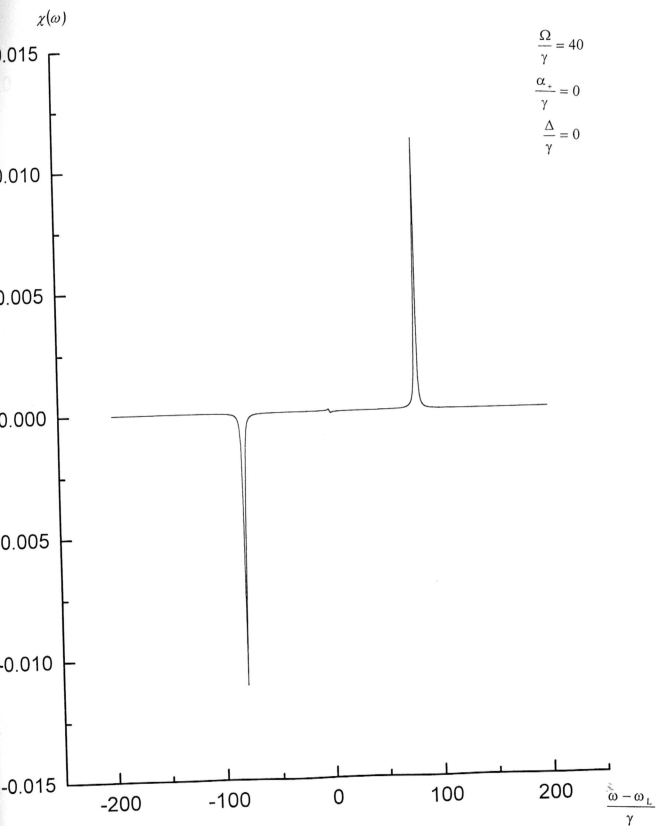


Figure 3.4.4 Plot of dispersion spectrum for two atoms driven by a resonant laser.

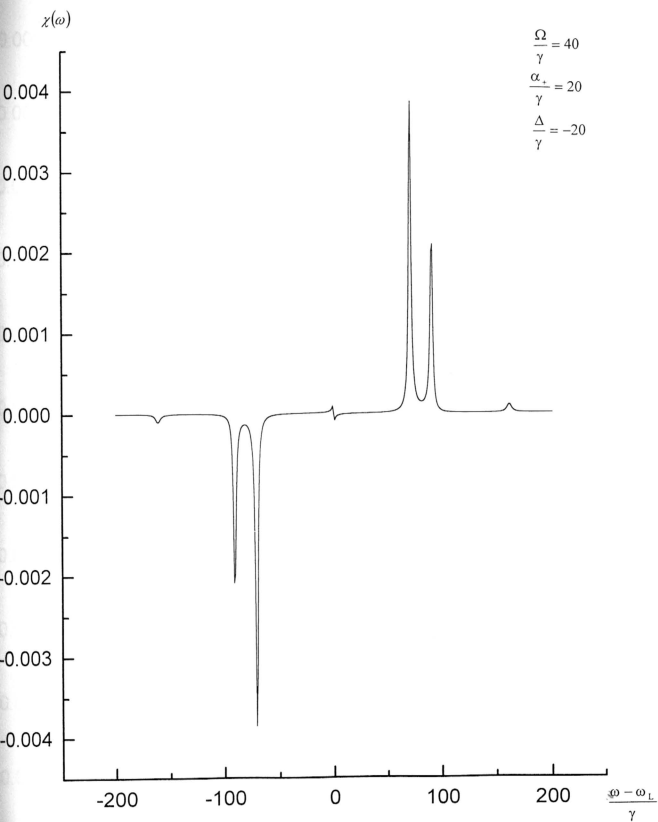


Figure 3.4.5 Plot of dispersion spectrum for two atoms driven by a resonant laser.

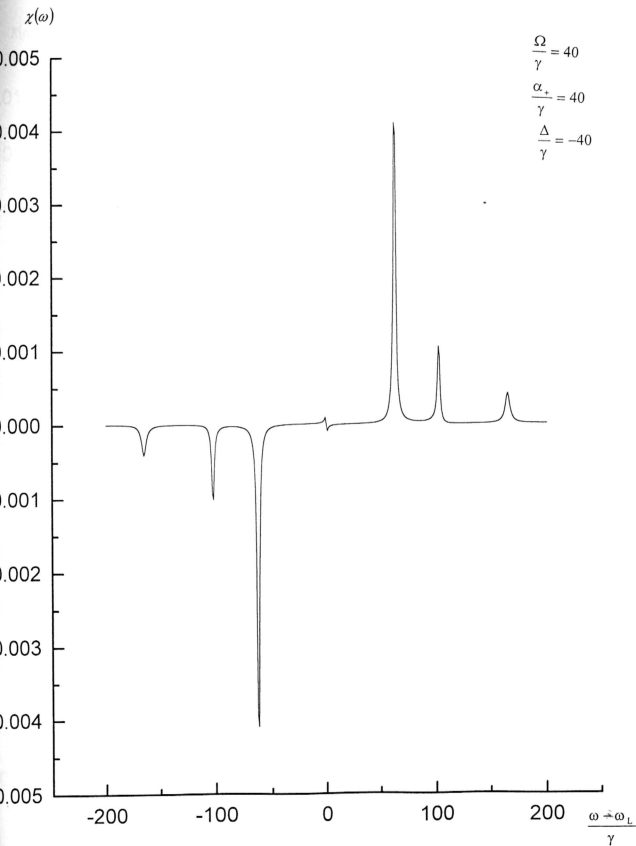


Figure 3.4.6 Plot of dispersion spectrum for two atoms driven by a resonant laser.

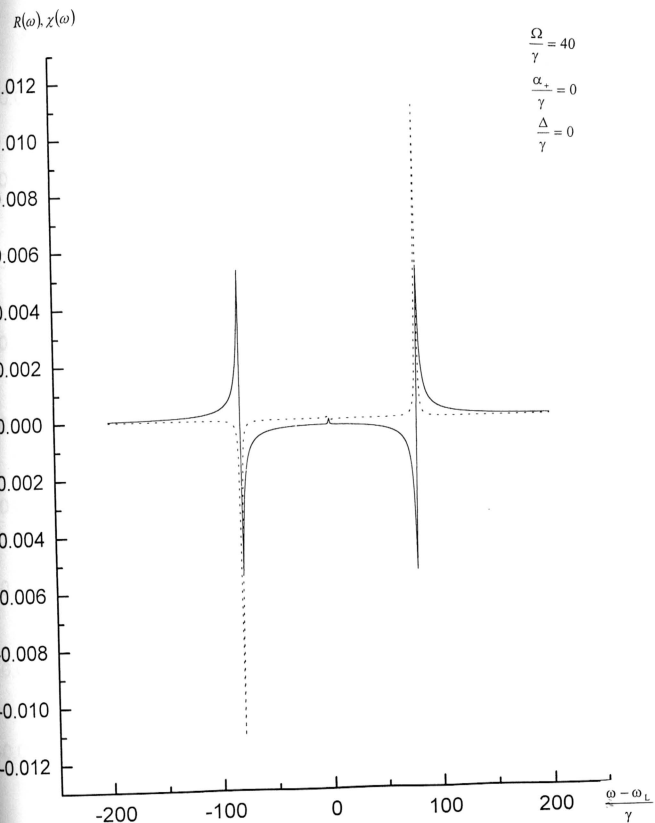


Figure 3.4.7 Plot of absorption-dispersion spectrum for two atoms driven by a resonant laser. The solid and dashed lines represent the absorption and dispersion spectrum respectively.

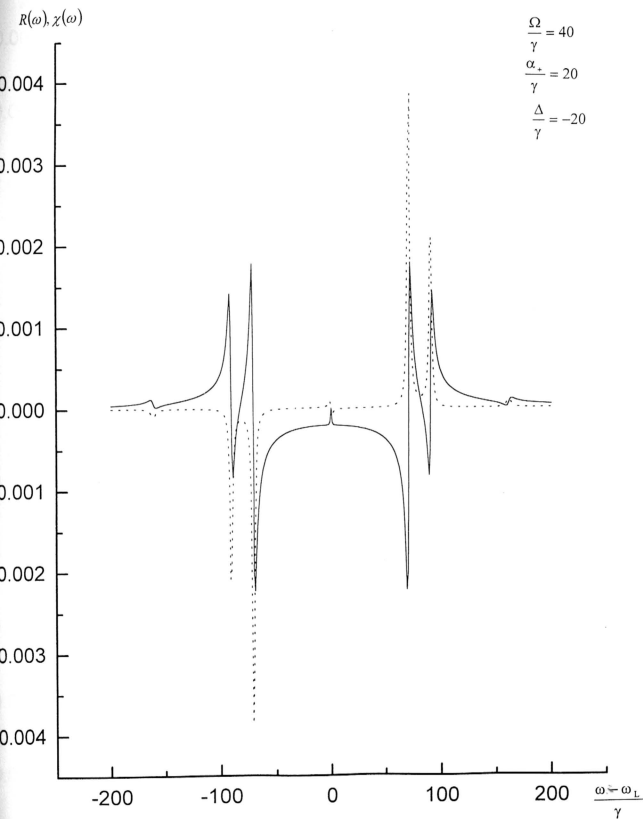


Figure 3.4.8 Plot of absorption-dispersion spectrum for two atoms driven by a resonant laser. The solid and dashed lines represent the absorption and dispersion spectrum respectively.

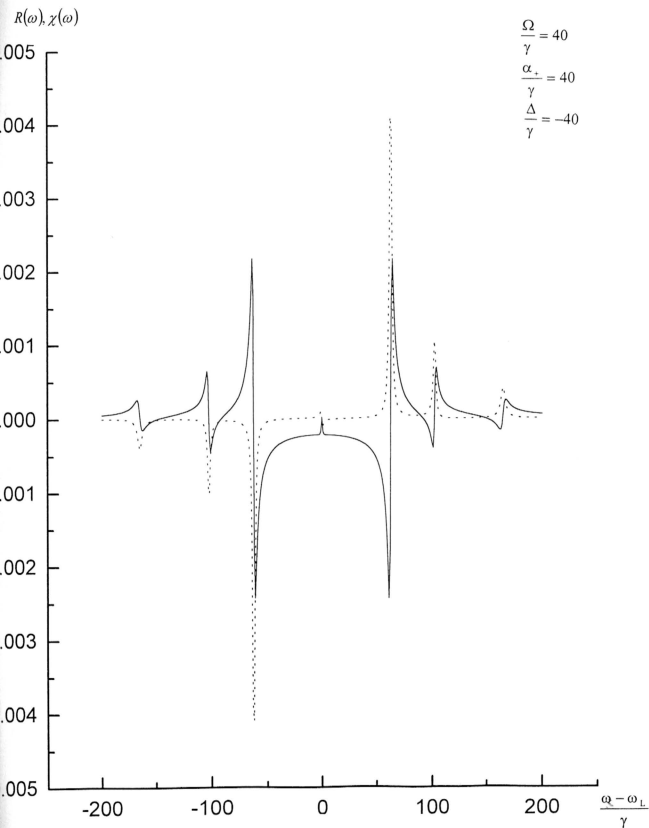


Figure 3.4.9 Plot of absorption-dispersion spectrum for two atoms driven by a resonant laser. The solid and dashed lines represent the absorption and dispersion spectrum respectively.

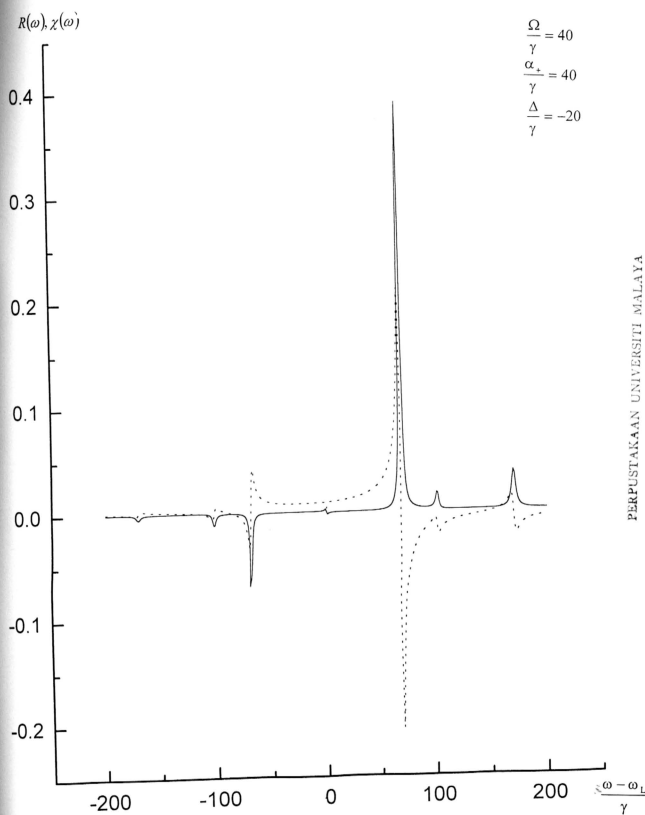


Figure 3.4.10 Plot of absorption-dispersion spectrum for two atoms driven by a **detuned** laser. The solid and dashed lines represent the absorption and dispersion spectrum respectively.