CHAPTER 1

INTRODUCTION

Coherent control is one of the leading themes of quantum optics research that is rich in new and counter-intuitive phenomena. The control strategies here are derived from the phenomena of quantum coherence and interference established in multilevel atomic systems driven coherently by two or more electromagnetic fields. In particular, the control of optical response of an atomic/molecular medium and manipulation of light propagation through such a medium has received considerable attention [1-4]. Some of the best known examples of this research are Autler-Townes (AT) splitting [5-9], coherent population trapping (CPT) [10-13], electromagnetically induced transparency (EIT) [14-39], electromagnetically induced absorption (EIA) [40-52] and lasing without population inversion (LWI) [53-79]. These phenomena are characterized by ultra-narrow linewidths, and modified linear and nonlinear susceptibilities. While on one hand these phenomena help to understand the subtle quantum effects in laser-atom interactions, they on the other hand provide useful platform for development of quantum technologies, e.g., frequency stabilizer [80-82], miniaturized atomic clock [83-87], precision magnetometer
In very recent years the research in this area has expanded in several new and exotic directions which include for example, subluminal and superluminal light propagation [96-114] and search for systems exhibiting negative refractive index [115-131].

The main objective of this thesis is to investigate the phenomenon of quantum coherence and interference in optical processes with the objective of achieving control of the interaction between atomic or molecular systems and electromagnetic fields. This chapter discusses briefly the basic physics underlying these optical phenomena and provides a perspective of their applications. The discussion presented here forms the basis for research work presented in the subsequent chapters.

1.1 Coherence and Interference in Atom-Field Interaction

Coherence is fundamental to the quantum optical phenomena. An atomic system interacting with a coherent electromagnetic field retains a distinct phase relationship with the field as long as the incoherent processes, i.e., decays due to spontaneous emission or collisions, do not override the atom-field interaction dynamics. The problem then can be addressed in the framework of quantum mechanics, where superposition and interference play an important role. The essential features of this quantum description are usually discussed by considering a finite-level atom interacting with a classical electromagnetic field. Such idealized $n$–level systems can be realized experimentally by identifying suitable hyperfine levels or Zeeman sublevels in simple atoms, e.g., alkali atoms. Two- and three-level atomic systems are paradigm of these studies, although general $n$–level systems ($n > 3$) provide opportunities to study more complex quantum dynamics as we
see later. The energy levels and atom-field interaction parameters relevant for the discussion of two- and three-level atoms are schematically shown in Fig. 1.1 and 1.2 respectively.

\[ |2\rangle \quad \Delta \quad |1\rangle \]

**Fig. 1.1:** Two-level atom coherently driven by a laser field of Rabi frequency \( \alpha \). \( \Delta \) is the detuning of laser from atomic transition frequency. \( \gamma \) is the radiative decay rate associated with \( |2\rangle \rightarrow |1\rangle \) transition.

\[ |3\rangle \quad \Delta_c \quad |1\rangle \]

\[ |2\rangle \quad \Delta_p \quad \gamma_{s2} \quad |3\rangle \]

**Fig. 1.2:** Level scheme representation of (a) \( \Lambda \) (b) \( V \) and (c) \( \Xi \) systems. Here \( \Delta_c (\Delta_p) \) and \( 2\alpha_c (2\alpha_p) \) are respectively the detuning and Rabi frequency of the pump (probe) laser field and \( \gamma_{ij} \) is radiative decay rate associated with \( |i\rangle \rightarrow |j\rangle \) transition.

The quantum mechanical framework necessary for description of finite-level systems interacting with two or more lasers and with vacuum of radiation field is explicitly developed in Chapter-2. In what follows, the essential results concerning coherence and interference in atomic media and pertaining to the scope of this thesis are reviewed.
1.1.1 Rabi Oscillations and Dressed States

For a two-level atom interacting with classical electromagnetic field, $E = E_0 \cos(\omega t)$, the atom-field dynamics is described by Hamiltonian,

$$ H = H_o + V , $$

where $H_o$ is the field-free Hamiltonian and $V = -d \cdot E$ is the interaction in electric dipole approximation. Here $E$ and $\omega$ are respectively the electric field and frequency of radiation field, and $d$ is the transition dipole moment associated with $|1\rangle \rightarrow |2\rangle$ transition. Atom-field dynamics is then determined by the Rabi frequency $2\alpha$ and detuning ($\Delta$) defined as

$$ 2\alpha = d \cdot E / h , \quad \Delta = \omega_{21} - \omega , $$

where $\omega_{21}$ is the atomic transition frequency. For a loss-less system, the ground and excited level populations exhibit out of phase oscillations, i.e., Rabi oscillations. The oscillation frequency is given by the generalized Rabi frequency defined as

$$ \Omega_r = \left( 4\alpha^2 + \Delta^2 \right)^{1/2} . $$

Incoherent decay ($2\gamma$) results in damping of the Rabi oscillations, and for $\alpha > \gamma$ coherence can persist over several Rabi periods. The model of two-level atom interacting with monochromatic radiation field also permits to introduce the dressed states $|\psi_\pm\rangle$, i.e. the eigen states of the atom + field Hamiltonian $H$, of energies $\varepsilon_\pm$ such that

$$ |\psi_+\rangle = \sqrt{\frac{\Omega_r + \Delta}{2\Omega_r}} |1\rangle \mp \sqrt{\frac{\Omega_r - \Delta}{2\Omega_r}} |2\rangle , \quad \varepsilon_\pm = (\Delta \pm \Omega_r) / 2 . $$

The dressed states can be observed using the techniques of coherent pump-probe spectroscopy. As a part of research work reported in this thesis, suitable experimental
techniques were developed to identify relevant dressed states corresponding to coherent interaction of a strong pump with hyperfine levels of D$_2$ transition in alkali atoms.

### 1.1.2 Autler-Townes Splitting

Autler-Townes (AT) splitting refers to the splitting of the absorption line due to dressing of an atom by a coherent radiation field [5]. Three-level systems as shown in Fig. 1.2 provide the requisite platform for observation of AT splitting. For example, in Fig. 1.2(a), transition $|2\rangle \rightarrow |3\rangle$ is dressed by a strong pump (control) laser of Rabi frequency $2\alpha_c$ and the resulting dressed states are interrogated by a weak probe laser that is scanned in the vicinity of $|1\rangle \rightarrow |3\rangle$ transition. Probe absorption spectrum is then a doublet corresponding to the dressed state transitions $|1\rangle \rightarrow |\psi_{\pm}\rangle$. Frequency separation between these two resonances is given by $\Omega_R = (\Delta^2 + 4\alpha_c^2)^{1/2}$ (cf. Eq. (1.4)) and their half widths are

$$\Gamma_{\pm} = \frac{\gamma_3 + D}{2} \left( \frac{\Delta_{\pm}}{\Omega_R} \right),$$

where $\gamma_3 = \gamma_{31} + \gamma_{32}$ and $D$ is a measure of Doppler width of the medium [6]. One thus observes that for $\Delta = 0$, both resonances have equal linewidth ($\Gamma_+ = \Gamma_-)$, while for $|\Delta| >> \alpha$ one of the resonances can be made of sub-Doppler or even sub-natural linewidth. AT splitting provides a useful way to obtain the properties of an atom/molecule interacting with near resonant radiation [1-6]. Recently AT doublet has also been studied in reference to high order nonlinear processes [7], quantum beats and quantum well structures [8] and in molecular systems [9]. The prospect of obtaining
ultra-narrow linewidth (cf. Eq. (1.5)) has been employed for development of tunable atomic frequency offset locking (AFOL) schemes [80-82].

### 1.1.3 Coherent Population Trapping

Susceptibility ($\chi$) of a two-level atomic medium interacting with a monochromatic field is largely dominated by absorption, i.e., $\text{Im}(\chi)$ [1-4]. Thus this system is unsuitable for applications in nonlinear optics. A three-level system interacting with two coherent fields gives rise to a range of coherent phenomena including CPT and EIT which suppress the resonant absorption [10-39]. The result is a very large dispersive optical nonlinearity which can also be used to control the propagation of light through the medium. The difference between AT and CPT/EIT is closely connected with the difference in the behaviour of two- and three-level systems undergoing resonant excitations. While AT doublet is related only to the development of atomic coherence, EIT and CPT are the results of quantum interference between absorption pathways in a multilevel system [10-19]. It is therefore possible to discriminate AT and EIT on the basis of Fano type interference in the latter mechanism [20] and threshold coupling [21]. A significant feature of EIT and CPT is that they afford sub-natural resolution even in a Doppler broadened medium [22-32].

The basic principle of CPT lies in the use of laser-induced coherences to generate a dark state formed from the coherent superposition of two long lived bare atomic states [10-12]. Consider for example a three-level system in Λ configuration (cf. Fig. 1.2 (a)). When $\alpha_p \sim \alpha_c$, both the fields participate in the dressing of the medium and the
diagonalization of the total Hamiltonian results into the formation of a bright (coupled) state $|C\rangle$ and a dark (non-coupled) state $|NC\rangle$, i.e.,

$$|C\rangle = (\alpha_r / \alpha_p)|1\rangle + (\alpha_c / \alpha_r)|2\rangle,$$

$$|NC\rangle = (\alpha_c / \alpha_r)|1\rangle - (\alpha_p / \alpha_r)|2\rangle,$$

where $\alpha_r = (\alpha_c^2 + \alpha_p^2)^{1/2}$. The dark state is uncoupled from the excited state, i.e. $\langle 3|d \cdot E|NC\rangle \rightarrow 0$ and therefore after being pumped into this dark state, atoms cannot be excited by either of the laser fields. This optical pumping process removes all the population from $|C\rangle$ and traps it into $|NC\rangle$ eventually. This results in the formation of an ultra-narrow ‘dark’ resonance; where the word ‘dark’ is used to denote its non-absorptive nature. These dark resonances are of particular interest for several applications such as efficient nonlinear processes [13], amplification without inversion (AWI) [53-79], atomic frequency standards [80-91], laser cooling [92,93], quantum information processing [94,95] and control of light propagation within a medium [96-114].

1.1.4 Electromagnetically Induced Transparency

EIT is a special case of CPT where the probe field is much weaker compared to the pump field. It represents cancellation of linear susceptibility at the two-photon resonance condition ($\Delta_p = \Delta_c$) via destructive quantum interference; thus rendering an otherwise optically opaque medium transparent [14-18]. The classical analogy of EIT with coupled harmonic oscillators has been demonstrated by Alzar et al. [19]. EIT can be described in terms of two processes that work in tandem to create transparency in the
media: formation of two dressed states by the strong pump and destructive interference in
the probe absorption to these states. For $\alpha_p \ll \alpha_c$, Eq. (1.6) can be simplified to obtain

$$|1\rangle = [\alpha_c |NC\rangle + \alpha_p |C\rangle] / \alpha_T \sim |NC\rangle,$$

(1.7)

implying that the ground state is decoupled from the excited state. Atoms prepared in this
state do not interact with the probe field and hence its absorption in the media vanishes.

Though EIT and CPT appear to be interrelated, there are some distinct differences
between the two processes. CPT is associated with the change in populations only, while
EIT depends on the optical response of the medium. Further EIT is an instantaneous
process (time scale $\sim 1/\alpha_c^2$), while the response time of CPT is much slower i.e. of the
order of several radiative lifetimes or optical pumping timescales [14-16].

In case of a Doppler broadened atomic medium, EIT may be thought of as arising
from the AT doublets corresponding to atoms of velocity $v$ which modifies the pump
detuning $\Delta_c$ to $\Delta_c + k \cdot v$ due to Doppler shift where $k$ is the wave vector. Consider for
example the case when $\Delta_c = 0$. For zero velocity group of atoms, the AT doublet is
symmetric with respect to $\Delta_p = 0$. For all other velocity groups due to Doppler shift one
of the AT components is drawn arbitrarily close to the central frequency while the other
one is pushed away. The averaging of all these AT doublet spectra results into an ultra-
narrow transparency window at $\Delta_p = 0$, which corresponds to EIT resonance. This
representation of EIT is convenient in arriving at the linewidth of EIT in a Doppler
broadened atomic medium as given by Javan et al. [35-37].

For a three-level $\Lambda$ system under weak saturation the half width of EIT is given
by $\Gamma_{EIT} = \alpha_c [2\Gamma_{21}(1 + s) / \gamma_32]^{1/2}$ with $\gamma_31 = \gamma_32$ and $s = \alpha_c^2 \gamma / 2\Gamma_{21}W_D^2$ where $2W_D$ is the
Doppler width of the medium [34-37]. Importance of $\Gamma_{21}$, which determines the coherence lifetime of the system, is clear from this expression. Note here that the $\Lambda$ system has minimum coherence dephasing rate compared to $V$ and $\Xi$ systems, and as a consequence ultra-narrow linewidth EIT can be obtained in a $\Lambda$ system compared to the other configurations [38,39]. Interest in EIT stems from its wide range of applications in enhancement of nonlinear processes [14-16], quantum information control [17], LWI [53-79], AFOL [80-82], time and frequency standards [83], laser cooling and trapping [93], Bose-Einstein condensate [93], super- and sub-radiance [95], slowing [96-98] and storage [99] of light, and realization of negative refraction [126-131].

1.1.5 Electromagnetically Induced Absorption

In contrast to EIT, EIA corresponds to the enhanced absorption of light around resonance due to constructive quantum interference between the excitation amplitudes [40-50]. There are two underlying physical mechanisms for EIA, transfer of coherence (TOC) and transfer of population (TOP) [41]. In a closed system when the pump and probe beams have different polarizations, TOC gives rise to EIA. Here EIA is associated with creation of light induced Zeeman coherences in the excited state and their transfer to ground state by spontaneous emission [40,41]. This happens in the absence of ground state population trapping under the condition that lasers couple two degenerate atomic levels and that the angular moment of the excited state is higher than that of the ground state [41]. EIA can also arise when TOP mediated by collisions from the ground state to a reservoir (a nearby level that does not interact with the pump) is greater than that from the excited state. Such EIA is observed in an open system, when the pump and probe
beams have same polarizations [42]. It is important to note that unlike EIT, EIA can only occur in systems which behave as open $\Lambda$ systems and in the absence of population trapping [41-43]. Such systems can show both positive and negative dispersion. Further, absorption in these systems is reported to have a peak at the line centre accompanied with negative dispersion [43].

Most of the studies on EIA and conversion of EIT to EIA have been done using two-level degenerate systems and N- system [40-44]. In these contexts, the effect of Doppler broadening, coupling powers and temporal evolution of EIA have been studied both theoretically and experimentally [44-49]. EIA phenomenon has also been investigated to realize negative group velocity of light producing superluminal light pulses which may be helpful in storage of light [50-52].

1.2 Role of Incoherence

Incoherence in the laser-atom interaction dynamics is usually introduced by two distinct ways. First is the incoherent processes such as spontaneous emission and collisional decays associated with the medium, while the second is a fallout of intrinsic phase fluctuations associated with the driving field which gives it a finite bandwidth. Generally incoherence leads to destruction of atom-field phase relationship and adversely affects the quantum coherence and interference established in an atomic medium.

1.2.1 Collisional Relaxation

The collisional relaxation processes encountered in a typical vapour cell experiments are of two categories: phase changing and velocity changing collisions. As
the name suggests phase changing collisions cause change in the phase of atomic states thereby preventing the maintenance of coherent excitation. Therefore these collisions adversely affect EIT/CPT linewidths [132-136]. To counter this effect buffer gas and anti-relaxation coatings are used in the experiments. Buffer gas prevents relaxation by slowing down the diffusion of atoms thereby increasing their transit time across the laser beam. In contrast the velocity changing collisions can produce sharper EIT/CPT signals [136]. Velocity changing collisions are elastic collisions which can reduce/increase the velocity of atoms, thereby shuffling them between different velocity groups spanning over the Doppler profile. This increases the transit time and hence the Raman coherence lifetime. Further this causes more atoms to participate in coherence build up thereby aiding optical pumping. The narrowing of spectral lines by these collisions is termed as Dicke narrowing and is more pronounced when the mean free path between velocity collisions is smaller than the wavelength of light [137].

1.2.2 Spontaneously Generated Coherence

Spontaneous emission is a major limiting factor in the observation of coherent processes [1]. However a counter-intuitive phenomenon called spontaneously generated coherence (SGC) occurs in a degenerate or near degenerate level system where the interference between spontaneous emission channels from the same excited level to closely spaced ground levels or from two close lying excited levels to a ground level gives rise to an additional coherence in the medium [138]. SGC arises due to interaction of the closely spaced levels with the vacuum of electromagnetic field and has marked effect on the dynamics of a system. The essential conditions for obtaining SGC are
closely spaced level structure and non-orthogonal dipole matrix elements. SGC has been investigated in context of disappearance of dark state [138], spectral line narrowing and enhancement [139], dynamically controlled photonic band-gap structure [140], enhanced Kerr nonlinearity [141], charged quantum dots [142], AWI [143] etc.

1.2.3 Laser Phase Fluctuations

In coherent laser matter interaction the fields are idealized as monochromatic and pure sinusoid. However, in practice even the most stable laser is not truly monochromatic since random fluctuations in the field are a source of finite bandwidths. Considerable work has been reported in the literature on the effect of finite bandwidths of driving lasers on the coherent dynamics of two- and three-level systems [144-156]. In these works laser phase fluctuations are modeled as Gaussian white noise and theoretical models based on multiplicative stochastic processes have been developed to analyze the effect of bandwidths of lasers and any cross-correlation that may exist between the pump and probe fields [144-160].

For three-level systems it has been observed that the phase fluctuations in general broaden or destroy the coherence established in the medium [144-150]. The cross-correlation between the pump and probe fields can be effectively used to recover the coherent behavior, however, this recovery is dependent on the type of three-level configuration whether Λ, V or Ξ [146]. These studies point to the possibility of observing quantum coherence and interference based phenomena with finite bandwidth lasers provided the pump and probe beams are generated from the same laser source. Similar studies in the context of four-level configurations are scanty [157,158] and that
provides opportunities to investigate these issues in the context of complex quantum dynamics.

### 1.3 Scope and Perspectives

Quantum coherence and interference based phenomena have gone beyond the proof of principles. They have been increasingly employed in the development of quantum devices and also to explore newer frontiers of physics. A brief review of these areas pertinent to the present thesis is covered in this section.

#### 1.3.1 Ultra-Precision Measurements

The narrow dark resonance generated in EIT and CPT provides a useful platform for ultra sensitive measurements which are of great interest in the field of metrology [83-91]. In the context of time and frequency standard, which is defined in terms of the separation between ground hyperfine levels of $^{133}$Cs (9.192631770 GHz), ultra-narrow EIT/CPT resonances generated in room temperature Cs vapour cells provide an excellent reference frequency for the development of miniaturized atomic clock [85-87]. There exist two major advantages in using CPT/EIT for atomic clock development. Firstly, these clocks are passive frequency standards, i.e., they do not require microwave cavity for excitation of the ground hyperfine levels of $^{133}$Cs, unlike the Cs vapour or even cold atom fountain clocks. This passive approach supports substantial miniaturization of the device. Secondly they afford significant reduction in the light shift under appropriate conditions of frequency modulation. EIT/CPT based clocks are compact and portable. Typical frequency stability reported for this type of clocks is $\sim 3 \times 10^{-11}$ (at 1 s of
integration time) [85], which make them useful in communication and in improved global positioning systems (GPS). Also a micro-fabricated atomic clock with a volume of 9.5 mm$^3$, fractional frequency instability of $\sim 2.53 \times 10^{-10}$ (at 1 s of integration time) has been demonstrated by Knappe et al. [87].

The other application is ultra-sensitive magnetometers based on the measurement of Zeeman shifts in atomic spectra and correlation of these shifts with the local magnetic field [88-91]. The typical Zeeman shifts in alkali atoms is $\sim 4$-6 Hz/nT. Experimental measurement of these Zeeman shifts using ultra-narrow dark resonances is central to the development of ultra-sensitive atomic magnetometer [88-91]. A chip scale Rb magnetometer with a sensor of 12 mm$^3$, sensitivity of 50 pT/Hz$^{1/2}$ at 10 Hz bandwidth has been demonstrated by Schwindt et al. [90]. Theoretical limit of sensitivity of such magnetometers is $\sim 1$ fT/Hz$^{1/2}$. Ultra sensitive magnetometers offer numerous applications in medical field, measurement of planetary magnetic field, earthquake detection, tests of the fundamental symmetries of nature and many more [91].

Yet another important application in the domain of frequency standard is atomic frequency offset locking (AFOL) where an ultra-narrow EIT/CPT resonance is generated using a pair of pump and probe lasers satisfying two-photon resonance condition in D$_1$ or D$_2$ transition of alkali atoms and the probe laser is then stabilized on the EIT/CPT resonance. This scheme establishes a fixed frequency offset between probe and pump lasers, and the value of the offset is exactly equal to the transition frequency between two ground levels of the $\Lambda$ system for example. Frequency stability of EIT/CPT based AFOL schemes is much superior to that of the conventional master-slave laser systems [80-82].
1.3.2 Amplification without Inversion

The conventional population inversion condition for achieving lasing action arises from the equilibrium between stimulated absorption and stimulated emission processes. It is in general very difficult to achieve inversion condition at large frequencies, e.g. in extreme UV and X-ray regions, due to cubic dependence of spontaneous emission rate on frequency. The requirement of inversion can be circumvented by coherent preparation of atomic media and utilizing the non absorptive behaviour of EIT and CPT phenomena, and thereby making it possible to achieve AWI and LWI. AWI refers to observation of probe laser amplification in an atomic system where a coherent pump laser acting on one transition circumvents the population inversion condition for an adjoining transition connected by the weak probe [53-79]. LWI refers to the process of AWI plus an additional cavity to achieve lasing action. Interest in AWI and LWI stems from their potential application in generation of low threshold short wavelength lasers [53-59]. Further these radiation sources are expected to have interesting statistical properties such as narrower intrinsic linewidths and amplitude squeezing [60-65]. Also of interest are the issues that include LWI in quantum electrodynamics [66], nanostructures [67], and super- and sub-radiance [68].

Several schemes for the observation of AWI and LWI have been proposed [53-61] and successfully experimented [69-72]. It is widely accepted that inversionless gain in these systems is a consequence of many mechanisms [53-61]. The first one is related to recoil induced lasing where the asymmetry between shifts of stimulated emission and absorption is used to obtain frequency regions where the emission process dominates the absorption in the absence of population inversion. The second one is the inversion in the
dressed state or CPT basis [54-60]. The third mechanism is devoid of any hidden inversion and is a direct consequence of quantum interference. AWI in this situation arises due to the excitation of low frequency coherence in the medium. There exist many studies concerning the role of incoherent pumping [74-76] that compensates for the cavity and other losses, and the effect of homogenous as well as inhomogenous broadening [77-79] of the active medium in the achievement of the inversionless gain.

1.3.3 Slow, Fast and Stopped Light

Quantum interference phenomena give rise to steep change in dispersion of a medium, i.e, Re(χ) in the vicinity of the ensuing resonances. The unusual variations of the refractive index \( n_r(ω) \) of the medium then can be used to modify the group index \( n_g \) of the medium,

\[
 n_g = n_r(ω) + ω[dn_r(ω)/dω],
\]

so that the group velocity, \( v_g = c/n_g \), can be manipulated to achieve fast, slow and stopped light [96-114]. Specifically at the EIT condition, the term \( dn_r/dω \) can be made large and positive thereby giving rise to large group index and generate slowing of a pulse traveling in such a medium. A drastic reduction in speed of light has been demonstrated by Hau et al. [103]. Ultra slow light has promising applications in enhancing the efficiency of nonlinear processes, laser radars, telecommunications, and development of optical buffers and adjustable optical delays [103-107]. It is also possible to stop a light pulse completely when the group velocity is changing with time. In this case the information carried by the pulse is temporarily transferred to the medium.
Pulses can then be “revived” with their original information intact [108-110]. Apart from its application in communications this phenomenon can also be used for storage of light [99], in quantum information and computing as ‘atomic memories’ [111]. The other promising applications of varying the group velocity are in amplification of pondermotive dipole forces [106] and all optical switching [107].

On the other hand if the term $\frac{dn_r}{d\omega}$ is large and negative, for e.g. in an EIA media, the group index ($n_g$) can become negative [50-52]. This implies that the pulse propagation in the medium is much faster than the velocity of light, i.e., $v_g > c$. In other words, the anomalous dispersion region can be used for superluminal light propagation which may be helpful in communications and storage of light [111-114].

1.3.4 Negative Refraction

The fabrication of negative refractive index material, i.e., a medium exhibiting negative permittivity and permeability simultaneously, has attracted extensive attention in recent years [115-131]. These materials are also termed as left-handed materials (LHMs); the name derived from the fact that in such a medium the electric vector, the magnetic vector and the wave vector of a plane monochromatic wave form a left-handed coordinate frame. Since the pioneering work of Veselago [115,116], interest in these systems has grown enormously owing to the possibility of performing unusual and non-intuitive optics. Some of the exotic applications of LHMs are sub-wavelength imaging, reversed Doppler shift, reversed Snell’s law, obtuse angle for Cherenkov radiation, photon tunneling, electromagnetic cloaking and subluminal light propagation [115-121]. Most of the LHMs have been artificially realized in the microwave region by using
metallic split ring resonators and metallic wires, photonic crystals with periodicity much smaller than or of the order of the wavelength of the electromagnetic radiation [122,123].

Coherently driven multi-level atomic systems are promising and simpler candidates for realization of negative refractive index in the optical region [126-131]. With optimum choice of Rabi frequencies and detunings, it is possible to achieve large negative refractive index over a wide probe frequency band. EIT based systems are useful for cancelling the absorption in the medium [126-131]. Further the dispersion properties of such a medium can be used to control the group velocity of the probe beam from subluminal to superluminal [114].

1.3.5 Enhancement of Nonlinear Processes

The growing interest in enhancement of nonlinear processes stems from its several applications such as four-wave mixing, gigantic Kerr nonlinearities, generation of highly efficient optical parametric oscillator and quantum information processing [161-172]. It is interesting to note that while EIT is synonymous with the vanishing of linear susceptibility, the nonlinear susceptibility of the medium can undergo constructive interference which improves the conversion efficiency in four wave mixing [161,162]. The increased efficiency of nonlinear mixing processes is of interest in efficient frequency up-conversion, phase conjugation, control of phase matching and coherent Raman scattering [161-166]. It plays an important role in the generation of squeezed light when the intensity fluctuations in the probe are transferred to fluctuations in conjugate beam, resulting in a high intensity squeezed light [167,168]. It also concerns the area of ultra-cold atoms and Bose-Einstein condensates where standard quantum limit is an
important experimental factor [169,170]. An important nonlinear phenomenon is Kerr nonlinearity where the phase of an optical field is proportional to the intensity of another field. This is directly related to large cross phase modulation (XPM) [171,172]. Kerr nonlinearity offers numerous applications in information processing, generation of optical solitons, nondemolition measurements, quantum logic gates and generation of entangled states [164-166].

The major challenge in the observation of resonant nonlinear processes is that the nonlinear susceptibilities are much weaker than the linear susceptibilities. To this end the phenomena of EIT comes to help, since near the EIT resonance the linear susceptibility is completely cancelled. Consequently there is reduced resonant absorption, optimized phase matching condition due to zero dispersion and constructive interference for nonlinear susceptibility [161-166].

1.4 Organization of the Thesis

The present thesis deals with coherent dynamics of multi-level atomic/molecular systems and its manifestation in the observation of several of the above referred phenomena, i.e., EIT, EIA, AWI and negative refractive index, together with the issues connected with SGC, Kerr nonlinearity and the effect of finite bandwidths of driving fields. While major part of the thesis is concerned with theoretical studies, some work on experimental coherent pump-probe spectroscopy is also reported here. The investigations carried out in this thesis are organized in the following manner:

Chapter-2 provides the discussion on the master equation framework used for addressing the interaction of multi-level system with coherent multi-frequency
electromagnetic field. An explicit derivation of the semi-classical master equation in electric dipole and rotating wave approximations is discussed here for a three-level atomic system interacting with two external coherent fields and vacuum of radiation field. The treatment is generalized for four-level schemes of interest. Further generalization is achieved in the context of three-level molecular systems with permanent dipole moments for examining quantum coherence and interference in such systems.

Chapter-3 deals with coherent pump-probe spectroscopy of three-level molecular $\Lambda$ system with permanent dipole moments. Motivation for these studies is provided by the very recent interest in EIT in the molecular domain. We explicitly show here the absence of amplification in 2+2-photon process for reversal in the signs of permanent moments, as reported earlier [173]. The effect of permanent dipole moments on the observation of EIT and its connection to the issue of subluminal and superluminal light propagation is analyzed. The role of virtual mechanism in 2+1-photon EIT is further examined. This chapter thus provides an integrated view of coherent pump-probe spectroscopy of a medium of dipolar molecules and its comparison with atomic case.

Chapter-4 presents detailed analysis of coherent pump-probe spectroscopy in $\Lambda$ system with an additional adjacent excited level. The level scheme thus consists of two simultaneous $\Lambda$ resonances with common ground levels and excited by the same pair of pump and probe fields, i.e., degenerate double lambda (DDL) resonance. Theoretical results are obtained for probe absorption spectrum and dispersion in the absence/presence of Doppler broadening to observe peculiar interference effects. These are illustrated using model schemes in $D_1$ and $D_2$ transitions of $^{85}\text{Rb}$. The chapter is completed with experimental results on the dressed state spectroscopy in a Doppler broadened medium of
87Rb atoms. The results of this chapter thus provide a realistic description of pump-probe spectroscopy of hyperfine transitions of alkali atoms.

Chapter-5 presents a detailed analysis and discussion on the phenomenon of AWI in the DDL system. It is shown here that a four-level DDL system under specific conditions can exhibit AWI without need of incoherent pumping. The dependence of AWI on atom-field interaction parameters, spontaneous emission rates, low-frequency coherence and Doppler velocity distribution is investigated. Approximate analytical expression for probe absorption is derived to corroborate the numerical results and to discuss the contrasting behavior, i.e., absorption vs. AWI, for the model DDL systems in D1 and D2 transitions of 87Rb. The discussion on AWI is further augmented using quantum jump formalism, which provides useful insight into the underlying mechanism responsible for amplification.

Chapter-6 deals with theoretical analysis of interference effects in general four-level configurations, i.e., tripod system and N-resonance, driven by three coherent fields from the viewpoint of controlling of their coherent dynamics and its manifestations. Tripod system is studied to demonstrate the observation of ultra-narrow double dark resonances. Some specific issues addressed in context of N system include switching between EIT and EIA, role of SGC and enhancement of the Kerr nonlinearity. Also reported here are the experimental results on EIT in N system and its comparison with a Λ system in a medium of Doppler broadened 87Rb atoms.

Chapter-7 deals with the investigations of laser phase fluctuations on the coherent dynamics of four-level systems with N-resonance as an example. The problem is formulated in the framework of master equation and multiplicative stochastic processes
and the effect of finite bandwidths of lasers and their cross-correlation on three-photon and 2+1-photon resonance is examined. It is observed that the phase fluctuations tend to broaden and destroy the sharp resonances, and dampen the Rabi oscillations; however the extent of this effect is critically dependent on the phase fluctuations and cross-correlations associated with the three fields. The effect of phase fluctuations on the EIT and EIA resonances is also reported here.

Chapter-8 is devoted to exploratory investigations on the observation of negative refractive index in four-level systems interacting with three coherent fields, a probe, a control and a \( rf \) field. In the framework of master equation and Classius-Mossotti relation, we obtain relative permittivity and permeability for a dense medium of such atoms to show the existence of probe frequency domains where permittivity and permeability can become simultaneously negative. The use of the dispersion property of the negative refractive index to control the group velocity of the probe beam from subluminal to superluminal is also discussed.

Finally the important conclusions of the present study and scope for future work are briefly summarized in Chapter-9.