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Multi-Wave Mixing Processes

From Ultrafast Polarization Beats to Electromagnetically Induced Transparency

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——从超快极化拍到电磁诱导透明

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With 134 figures

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Preface

Nonlinear optics covers very broad research directions and has been a very active area of research for about fifty years since the invention of the first laser at the beginning of 1960s. There are several excellent text books devoted to various aspects of nonlinear optics including *Nonlinear Optics* by R. W. Boyd, *Nonlinear Optics* by Y. R. Shen, *Quantum Electronics* by A. Yariv, *Principles of Nonlinear Optical Spectroscopy* by S. Mukamel, and *Nonlinear Fiber Optics* by G. P. Agrawal. Multi-wave mixings in gases, liquids, and solid materials are important parts of the nonlinear optical process. Typically, lower-order nonlinear optical processes always dominate since they are more efficient than higher-order ones. So normally only two-wave, three-wave, and four-wave mixing (FWM) processes, depending on symmetries of nonlinear materials, are studied, and their basic principles are covered in detail in those textbooks. FWM comes from the third-order nonlinearity, which is one of the most popular nonlinear phenomena, and can be easily observed in materials with the inversion central symmetry (in which the third-order nonlinearity is the lowest nonlinear one). In certain specially designed material systems, or in certain phase conjugation configurations, the FWM efficiency can be very high, reaching 100% or even with gain. With newly developed short-pulse high-power lasers, and new materials designed and optimized for certain nonlinearities, higher-order wave-mixing processes, such as the six-wave mixing (SWM, corresponding to the fifth-order nonlinearity) and even the eight-wave mixing (EWM, corresponding to the seventh-order nonlinearity), have been experimentally investigated in recent years. Since typically higher-order nonlinear coefficients are much smaller than lower-order ones, in order to observe higher-order wave-mixing signals, one needs to eliminate (or at least greatly suppress) lower-order signals by various techniques, or to perform a heterodyne detection between the weak signal from a higher-order nonlinear process and a much stronger lower-order signal (as a local oscillator). Of course, very high-order harmonic generations have been used recently to generate UV and even x-ray wavelengths by employing very short and high intensity laser pulses. However, high-order nonlinearities described in this book do not include this region of extreme nonlinear processes.

The authors have worked, both theoretically and experimentally, on nonlinear optics for many years, especially on high-order nonlinear wave-mixing processes in the past few years. They have worked on the electromagnetically induced transparency (EIT)- or atomic coherence-enhanced multi-wave mixing processes in multi-level atomic systems, which have many advantages over traditional multi-wave mixing processes in the nonlinear media. By specially selecting atomic energy levels and phase-matching conditions, they have shown co-existing FWM and SWM signals in several open- and close-cycled four-level atomic systems by making use of the EIT concept of two-photon Doppler-free configurations in Doppler-broadened atomic media. Using uniquely designed spatial laser beam patterns and configurations, generated FWM and SWM signals can fall into a same EIT window with low absorption, and propagate in the same direction with the same frequency. SWM processes can be greatly enhanced by playing with atomic coherences between different energy levels, and relative strengths between FWM and SWM signals can be completely controlled. SWM signals can even be enhanced to have same amplitudes as FWM signals in the same system at the same time. Spatial and temporal interferences, as well as interference in the frequency domain, between FWM and SWM signals have been experimentally demonstrated in four-level atomic systems. An efficient energy exchange during propagation between generated FWM and SWM signals (and with the probe beam) was also observed and studied in detail.

In this monograph, the authors will describe treatments of multi-wave mixing processes using the perturbation approach and show how, by manipulating phase-matching and EIT conditions using various laser beams, co-existing FWM and SWM (or even EWM) processes can be achieved, and how to control their interplays. It has been shown that one can control the relative strength of generated FWM and SWM signals (by using the amplitudes and the frequency detuning of pump beams) and the relative phase between them (by adjusting the time delay of one of the pump beam involved only in the FWM process). This monograph will mainly focus on the relevant work recently done in the authors' group. Other than EIT-enhanced co-existing high-order nonlinear wave-mixing processes in multi-level atomic systems, several other topics will also be discussed, including femtosecond and attosecond polarization beats between two FWM, or two SWM processes, and their heterodyne detections in multi-level atomic systems, as well as Raman- or Rayleigh-enhanced polarization beats in the liquid systems. Some potential applications of the fast polarization beats and EIT-enhanced co-existing high-order nonlinear wave-mixing processes are also discussed in the book.

The authors believe that although several good textbooks on the general topics of nonlinear optics exist, the current book treats a special topic of co-existing multi-wave mixing processes in multi-level systems and will have high values to serve a special group of readers. Especially, the topic of EIT- or atomic coherence-enhanced multi-wave nonlinear optical processes and their interplays have not been touched by any existing books. This monograph serves as a reference book intended for advanced undergraduates, graduate students, and researchers working in the related field of nonlinear optics, nonlinear optical spectroscopy, and quantum optics.

We take this opportunity to thank many researchers and collaborators who have worked on the research projects as described in this book. We specially thank Leijian Shen and Ling Li for their great helps in compiling this book.

Yanpeng Zhang
Min Xiao
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Contents

1 Introduction

The main subject of this book centers around mainly two topics. The first topic (Chapters 2 – 5) covers the ultrafast polarization beat due to the interaction between multi-colored laser beams and multi-level media. Both difference-frequency femtosecond and sum-frequency attosecond polarization beats can be observed in multi-level media depending on the specially arranged relative time delay in multi-colored laser beams. Effects of different stochastic noise models for the lasers on the polarization beat signal are carefully studied. Polarization beats between MWM processes are among the most important ways to study transient property of the medium. The second topic (Chapters 6 – 8) relates to the co-existence and interplay between efficient multi-wave mixing (MWM) processes enhanced by atomic coherence in multi-level atomic systems. The co-existing higher-order nonlinear optical process can be experimentally controlled and becomes comparable or even greater in amplitude than the lower-order wave-mixing process by means of atomic coherence and the multi-photon interference. Furthermore, the spatial-temporal interference and efficient energy exchange during propagation are shown to exist between the generated four-wave mixing (FWM) and six-wave mixing (SWM) signals. The multi-dressed wave-mixing process is also investigated in a multi-level atomic system to enhance or suppress the MWM process. Only the MWM process with multi-colored laser beams and related effects in multi-level media will be covered in this book. Experimental results will be presented and compared with theoretical calculations throughout the book. Also, emphasis will be given only to works done by the authors' group in the past few years. Before starting the main topics of the book, some basic physical concepts and mathematical techniques, which are useful and needed in the later chapters, will be briefly presented in this introduction chapter.

1.1 Nonlinear Susceptibility

Nonlinear optics is the study of optical phenomena that occur in a material system as a consequence of nonlinear response to the input light. Typically, only the coherent laser light is sufficiently intense to provide the nonlinear changes to the material's optical properties. In fact, the beginning of the field of nonlinear optics has often been taken to be at the discovery of the second-harmonic generation (SHG) by Fraken, et al. in 1961 [1], just shortly after the demonstration of the first working laser by Maiman in 1960 [1]. Nonlinear optical phenomena are "nonlinear" in the sense that they occur when the response of a material system to an applied optical field depends on a nonlinear manner to (or high-order power of) the strength of the input optical field. For example, SHG occurs in many optical crystals as a result of the applied strong optical field with the generated second-harmonic signal intensity (at twice the frequency of the applied light) proportional to the square of the applied light intensity. For inversion symmetric materials, such as atoms, the second-order nonlinearity usually does not exist, so the lowest nonlinear effect is the third-order one.

Today, lasers with very high intensities and very short pulse durations are readily available, for which concepts and approximations of the traditional nonlinear optics can no longer apply. In this regime of extreme "nonlinear optics", a large variety of novel and unusual effects arise, such as frequency doubling in inversion symmetric materials or high-harmonic generations in gases, which can lead to attosecond electromagnetic pulses or pulse trains [2]. Other examples of "nonlinear optics" cover diverse areas such as solid-state physics, liquids, atomic and molecular physics, relativistic free electrons in a vacuum, and even the vacuum itself [3, 4]. In this book, we only deal with nonlinear multi-wave mixing processes at relatively lower – orders in multi-level media, in which traditional principles and approximations of nonlinear optics (as described in the textbooks [3, 4]) are still hold.

In order to describe more precisely what we mean by the optical nonlinearity, let us consider how the dipole moment per unit volume, or polarization $\boldsymbol{P}$, of a material system depends on the strength E of the applied optical field. The induced polarization depends nonlinearly on the electric field strength of the applied field in a manner that can be described by the relation $\boldsymbol{P} = \boldsymbol{P}_L + \boldsymbol{P}_{NL}$ [3, 4]. Here,

$$\boldsymbol{P}_L = \boldsymbol{P}^{(1)} = \varepsilon_0 \boldsymbol{\chi}^{(1)} \cdot \boldsymbol{E}$$

$$\boldsymbol{P}_{NL} = \boldsymbol{P}^{(2)} + \boldsymbol{P}^{(3)} + \cdots = \varepsilon_0 (\boldsymbol{\chi}^{(2)} : \boldsymbol{EE} + \boldsymbol{\chi}^{(3)} \vdots \boldsymbol{EEE} + \cdots)$$

When we only consider the atomic system (which is isotropic and has inversion symmetry), we can write the total polarization as $P = \varepsilon_0 \chi E$ in general, where the total effective optical susceptibility can be described by a generalized expression of

$$\chi = \sum_{j=0}^{\infty} \chi^{(2j+1)} |E|^{2j}$$

The lowest term $\chi^{(1)}$ ($j = 0$) is independent of the field strength and is known as the linear susceptibility. The next two terms in the summation, $\chi^{(3)}$ and $\chi^{(5)}$, are known as third- and fifth-order nonlinear optical susceptibilities, respectively.

The index of refractive of many optical materials depends on the intensity of the light due to nonlinear responses, which can be described by the relation

$$n = n_0 + \sum_{j=1}^{\infty} \bar{n}_{2j}|E|^{2j}$$

The nonlinear indices $\bar{n}_{2j}$ are influenced by the intensity of the light [5]. An alternative way of defining the intensity-dependent refractive index is by the equation

$$n = n_0 + \sum_{j=1}^{\infty} n_{2j} I^j$$

where I denotes the intensity of the applied field, given by

$$I = \frac{\varepsilon_0 c}{2} |E|^2$$

Hence, $\bar{n}_{2j}$ and n_{2j} are related by $n_{2j} = \dfrac{2}{\varepsilon_0 c}\bar{n}_{2j}$ [3].

The linear and nonlinear refractive indices are directly related to the linear and nonlinear susceptibilities. It is generally true that $n^2 = 1 + \chi$, and by introducing

$$n = \sum_{j=0}^{\infty} \bar{n}_{2j} |E|^{2j}$$

on the left-hand side and

$$\chi = \sum_{j=0}^{\infty} \chi^{(2j+1)} |E|^{2j}$$

on the right-hand side of this equation, it gives

$$\left(\sum_{j=0}^{\infty} \bar{n}_{2j} |E|^{2j}\right)^2 = 1 + \sum_{j=0}^{\infty} \chi^{(2j+1)} |E|^{2j}$$

Correct to terms of up to the order of $|E|^{2j}$, this general expression gives the following relations, i.e.,

$$n_0^2 = 1 + \mathrm{Re}\chi^{(1)} \Rightarrow n_0 = \sqrt{1 + \mathrm{Re}\chi^{(1)}}$$

$$2n_0\bar{n}_2 = \mathrm{Re}\chi^{(3)} \Rightarrow \bar{n}_2 = \frac{\mathrm{Re}\chi^{(3)}}{2n_0} \Rightarrow n_2 = \frac{2}{\varepsilon_0 c}\frac{\mathrm{Re}\chi^{(3)}}{2n_0}$$

$$2n_0\bar{n}_4 + \bar{n}_2^2 = \mathrm{Re}\chi^{(5)} \Rightarrow \bar{n}_4 = \frac{\mathrm{Re}\chi^{(5)} - \bar{n}_2^2}{2n_0} \Rightarrow n_4 = \frac{2}{\varepsilon_0 c}\frac{\mathrm{Re}\chi^{(5)} - \bar{n}_2^2}{2n_0}$$

$$2\bar{n}_2\bar{n}_4 + 2n_0\bar{n}_6 = \mathrm{Re}\chi^{(7)} \Rightarrow \bar{n}_6 = \frac{\mathrm{Re}\chi^{(7)} - 2\bar{n}_2\bar{n}_4}{2n_0} \Rightarrow n_6$$

$$= \frac{2}{\varepsilon_0 c}\frac{\mathrm{Re}\chi^{(7)} - 2\bar{n}_2\bar{n}_4}{2n_0}$$

Finally the expression

$$n = \sum_{j=0}^{\infty} \bar{n}_{2j} |E|^{2j}$$

becomes

$$n(E) \simeq n_0 + [\mathrm{Re}(\chi^{(3)})/2n_0] |E|^2 + \{\mathrm{Re}(\chi^{(5)}) - [\mathrm{Re}(\chi^{(3)})/2n_0]^2\} |E|^4 / 2n_0 + \cdots$$

where n_0 represents the usual, weak-field (linear) refractive index and the new nonlinear optical constants n_{2j} (sometimes called the second-, fourth-, sixth-... order indices of refraction) give the rate at which the refractive index increases with increasing optical intensity. For a typical nonlinear medium, $\bar{n}_2$ and $\bar{n}_4$ have orders of magnitude of $10^{-7}\mathrm{m}^2/\mathrm{V}^2$ and $10^{-13}\mathrm{m}^4/\mathrm{V}^4$, respectively [5].

The change of optical properties due to the second-order refractive index n_2 (or third-order nonlinear susceptibility) is typically called the optical Kerr effect, by analogy to the traditional Kerr electro-optic effect, in which the refractive index of a material changes by an amount that is proportional to the square of the strength of an applied static electric field (i.e., $n \approx n_0 + \Delta\bar{n}$, $\Delta\bar{n} \approx \bar{n}_2 |E|^2 + \bar{n}_4 |E|^4$, $\bar{n}_4$ is typically neglected in low power range) [3].

Higher-order nonlinear susceptibilities are typically much smaller than the lower-order one by several orders of magnitude. In general, order of magnitude comparison is given by

$$\chi^{(n)}/\chi^{(n-1)} \approx 10^{-7}$$

So, typically the lowest-order non-zero nonlinear term always dominates in studying nonlinear optical properties in a medium, and higher-order nonlinear terms are simply neglected. However, in recent years higher-order nonlinearities have shown to make a significant contribution to nonlinear optical properties, even when the lower-order nonlinear term is not zero, especially at higher optical intensity [3, 4]. For example, in the N-type four-level system [5], as it can be appreciated from $\mathrm{Re}(\chi)$, the real part of the total susceptibility of the medium grows linearly with $|E|^2$ at low powers (due to the effect of only the positive term $\bar{n}_2 |E|^2$, which is the lowest nonlinear term.), but it decreases at high powers (due to the negative term $\bar{n}_4 |E|^4$), while the losses are comparatively small in this range [5]. In such a case, there is a balance for the diffraction plus self-focusing at low field amplitudes and self-defocusing at larger amplitudes. This type of competition can be found in media with the so called cubic-quintic-type nonlinearity, which can be very important in the propagation property of high intensity optical pulse, and optical soliton formation. As we will show in Chapters 6 – 8, different-order nonlinear optical processes can co-exist even with low power cw laser beams, enhanced by atomic coherence in the multi-level atomic systems [6 – 14].

1.2 Four-wave Mixing

The four-wave mixing (FWM) refers to the nonlinear optical process with four interacting electromagnetic waves (i.e., with three applied fields to generate the fourth field). In the weak interaction limit, FWM is a third-order nonlinear optical process and is governed by the third-order nonlinear susceptibility [3]. Unlike the second-order process, the third-order process is allowed in all media, with or without inversion symmetry. Therefore, in many optical media (such as the ones with the inversion symmetry), such third-order nonlinear wave-mixing processes are the lowest-order ones, which are the dominant nonlinear interactions. FWM processes are well studied in many material systems and their general properties can be found in well-written textbooks [3, 4], so we will not review the general topics of FWM processes here. Instead, in the following, we will only discuss some special cases of the FWM process, which are relevant mainly to certain parts of this book (Chapters 2 – 5).

Let us consider a special case of FWM processes. The third-order nonlinear polarization governing the process has, in general, three components with different wave vectors. E_1 (ω_1, $\boldsymbol{k}_1$), E_2 (ω_2, $\boldsymbol{k}_2$) and E_3 (ω_3, $\boldsymbol{k}_3$) denote the three input laser fields. Here, ω_i and $\boldsymbol{k}_i$ are the frequency and the propagation wave vector of the ith beam, respectively. We can choose to have a small angle θ between input pump laser beam $\boldsymbol{k}_2$ and beam $\boldsymbol{k}_1$. The probe laser beam (beam $\boldsymbol{k}_3$) propagates along a direction that is almost opposite to that of beam $\boldsymbol{k}_1$ (see Fig. 1.1). Because of the strong resonant interactions, the third-order nonlinear susceptibility $\chi^{(3)}$ for this FWM process can be very large in certain media (such as multi-level atomic systems). As a result, this third-order nonlinear optical process is often observable with relatively weak continuous-wave (CW) laser beams.

The output of the generated FWM signals can be easily understood from the following physical picture. Two of the three input waves interfere and form either a static grating or a moving grating (depending on the frequency difference between laser beams); the third input wave is then scattered off by this grating to yield the output signal wave. In most cases, contributions from the static gratings should dominate. With three input waves, three different gratings can be formed. The grating formed by the input $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$ waves scatters the $\boldsymbol{k}_3$ wave to yield output signals at $\boldsymbol{k}_3 \pm (\boldsymbol{k}_1 - \boldsymbol{k}_2)$. The one formed by the $\boldsymbol{k}_3$ and $\boldsymbol{k}_2$ waves scatters the $\boldsymbol{k}_1$ wave to yield output signals at $\boldsymbol{k}_1 \pm (\boldsymbol{k}_3 - \boldsymbol{k}_2)$. The one formed by the $\boldsymbol{k}_1$ and $\boldsymbol{k}_3$ waves scatters the $\boldsymbol{k}_2$ wave to yield output signals at $\boldsymbol{k}_2 \pm (\boldsymbol{k}_1 - \boldsymbol{k}_3)$. These processes are illustrated in Fig. 1.1. Altogether, three output signal waves with different wave vectors, $\boldsymbol{k}_{s1} = -\boldsymbol{k}_1 + \boldsymbol{k}_2 + \boldsymbol{k}_3$, $\boldsymbol{k}_{s2} = \boldsymbol{k}_1 - \boldsymbol{k}_2 + \boldsymbol{k}_3$, and $\boldsymbol{k}_{s3} = \boldsymbol{k}_1 + \boldsymbol{k}_2 - \boldsymbol{k}_3$, can be expected. As is common for optical wave-mixing processes, phase-matching condition ($\Delta \boldsymbol{k} = 0$) is of prime importance here, since it greatly enhances the signal output under the phase-matched condition. By carefully considering three output situations, as shown in Fig. 1.1, one can easily see that only the generated output at $\boldsymbol{k}_{s2} = \boldsymbol{k}_1 - \boldsymbol{k}_2 + \boldsymbol{k}_3$ is always phases matched among the three possible output waves, which is the usual case for efficient phase conjugation [4].

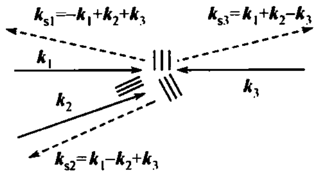

Fig. 1.1. Schematic diagram of phase-conjugate FWM.

More specifically, if beams 1 and 2 have the same frequency (i.e., $\omega_1 = \omega_2$) and a small angle θ is set between them, the coherence length of the generated FWM signal at $\boldsymbol{k}_{s2}$ is given by

$$l_c^f = 2c/[n(\omega_1/\omega_3)|\omega_1 - \omega_3|\theta^2]$$

which is much larger than that of the other two outputs at $\boldsymbol{k}_{s1}$ and $\boldsymbol{k}_{s3}$. However, the nonlinear interactions between laser beam 1 and beam 2 with an absorbing medium can give rise to the molecular-reorientation and the thermal non-resonant static gratings (i.e., Q_M and Q_T in liquid media) [4], respectively. In this case, the FWM signal at $\boldsymbol{k}_{s2} = \boldsymbol{k}_1 - \boldsymbol{k}_2 + \boldsymbol{k}_3$ is the results of diffractions of beam 3 with a frequency ω_3 by these two gratings. On the other hand, the interference pattern generated by beam 2 and beam 3 can move with a phase velocity $|\omega_3 - \omega_2|$. Now, if the frequency difference $\Delta_a = \omega_3 - \omega_2 \approx 0$, two resonant moving gratings Q_{RM} and Q_{RT} with a large angle are formed by the interference between beam 2 and beam 3. Beam 1 is then diffracted by them to enhance the FWM signals. This is the Rayleigh-enhanced FWM with the wave vector $\boldsymbol{k}_{s2} = \boldsymbol{k}_1 - \boldsymbol{k}_2 + \boldsymbol{k}_3$ at the direction of beam 4 (which will be discussed in Chapter 5). However, if the frequency difference $\Delta_b = \omega_3 - \omega_2 - \Omega_R \approx 0$ (where Ω_R is the Raman resonant frequency of the medium), one large resonant moving grating, Q_R, is formed by the interference between beam 2 and beam 3, which will excite the Raman-active vibrational mode of the Kerr medium and enhance the FWM signal (i.e., Raman-enhanced FWM, which will be discussed in Chapter 5.).

FWM interactions are the basic nonlinear optical processes to be discussed in this book. We will present near resonant third-order FWM interactions in two-level and three-level systems with degenerate and non-degenerate configurations. In later chapters, enhancements of FWM processes due to electromagnetically induced transparency (EIT) or atomic coherence in multi-level atomic systems [15–18], as well as competitions between different FWM channels [11], will be discussed. Higher-order wave-mixing processes, such as six-wave mixing (SWM, fifth-order nonlinearity) and eight-wave mixing (EWM, seventh-order nonlinearity), can also be generated and enhanced in multi-level atomic systems via atomic coherence.

1.3 Generalized Resonant MWM in Multi-level Atomic Systems

Multi-level atomic systems can generally be divided into two categories, i.e., close-cycled (such as Λ-type, ladder-type, N-type, double-Λ, and multi-level folded-type) systems and open-cycled (such as V-type, Y-type, inverted Y-type, and K-type) systems. Atomic coherences can be induced in both of such multi-level atomic systems. Enhanced FWM processes have been demonstrated in many of these multi-level atomic systems via atomic coherence, especially in Λ-type [16], double-Λ type [15, 17], ladder-type [6–12], and N-type [18] atomic systems. Typically, in the close-cycled multi-level atomic systems, lower-order nonlinear wave-mixing processes (such as FWM and SWM) can be turned off by special arrangement in laser beams, so the higher-order one (either SWM or EWM) can be observed [15–20]. Recently, new experiments have been designed and implemented, which can generate co-existing FWM and SWM processes in either open-cycled [7–12] or close-cycled [6] multi-level atomic systems by manipulating laser beam configurations and induced atomic coherence among the energy levels (which will be discussed in Chapters 6–8). Simultaneous opening of EIT windows and induced atomic coherence among relevant energy levels are essential in generating co-existing FWM and SWM signals with comparable intensities [7]. Such co-existing wave-mixing processes of different orders (such as FWM and SWM processes), and interplays between them, open the door for new interesting research directions in nonlinear optics. As the details of these systems will be discussed in later chapters, a general example will be given here in the following to illustrate the techniques used to treat such multi-level atomic systems.

Let us consider a close-cycled $(n+1)$-level cascade system, as shown in Fig. 1.2. The transition from state $|i-1\rangle$ to state $|i\rangle$ is driven by two laser fields $E_i(\omega_i, \boldsymbol{k}_i)$ and $E'_i(\omega_i, \boldsymbol{k}'_i)$, with Rabi frequencies G_i and G'_i, respectively. The Rabi frequencies are defined as

$$G_i = \varepsilon_i \mu_{ij}/\hbar$$
$$G'_i = \varepsilon'_i \mu_{ij}/\hbar$$

where μ_{ij} is the transition dipole moment between level i and level $j(or\ i-1)$. Fields E_n and E'_n (with the same frequency) propagate along beam 2 and beam 3, respectively, with a small angle θ between them [see Fig. 1.2(a)]. Fields E_2, E_3 to E_{n-1} propagate along the direction of beam 2, while a weak probe field E_1 (beam 1) propagates along the opposite direction of beam 2. The simultaneous interactions of the multi-level atoms with fields E_1, E_2 to E_n will induce atomic coherence between states $|0\rangle$ and $|n\rangle$ through the resonant n-photon transitions. This n-photon coherence is then probed by the field E'_n and, as a result, a $2n$-wave-mixing ($2n$-WM) signal of frequency ω_1 in beam 4 is generated almost exactly opposite to the direction of beam