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# Coherent Control of Four-Wave Mixing

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高等教育出版社
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SIBO HUNPIN XIANGGAN KONGZHI

With 190 figures

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# Preface

The field of nonlinear optics has developed rapidly since the invention of the first laser exactly 50 years ago. Many interesting scientific discoveries and technical applications have been made with nonlinear optical effects in all kinds of nonlinear materials. There are already several excellent general textbooks covering various aspects of nonlinear optics, including *Nonlinear Optics* by Robert W. Boyd, *Nonlinear Optics* by Yuen-Ron Shen, *Quantum Electronics* by Amnon Yariv, *Nonlinear Fiber Optics* by Govind P. Agrawal, etc. These textbooks have provided solid foundations for readers to understand various (secondand third-order) nonlinear optical processes in atomic gas and solid media. The earlier monograph by the authors, *Multi-wave Mixing Processes*, published last year, has presented experimental and theoretical studies of several topics related to multi-wave mixing processes (MWM) previously done in the authors' group. The topics covered in that monograph include ultrafast polarization beats of four-wave mixing (FWM) processes; heterodyne detections of FWM, six-wave mixing (SWM), and eight-wave mixing (EWM) processes; Raman and Rayleigh enhanced polarization beats; coexistence and interactions of MWM processes via electromagnetically induced transparency (EIT). The monograph shows the effects of high-order correlation functions of different noisy fields on the femto- and atto-second polarization beats, and heterodyne/homodyne detections of ultrafast third-order polarization beats. It has also shown the coexistence of FWM and SWM processes in several multi-level EIT systems, as well as interactions between these two different orders of nonlinearities.

This new monograph builds on and extends the previous works, and presents additional and new works done in recent years in the authors' group. Many newly obtained results, extended detail calculations, and more discussions are provided, which can help readers to better understand these interesting nonlinear optical phenomena. Other than showing more results on controls and interactions between MWM processes in hot atomic media, several novel types of spatial solitons in FWM signals are presented and discussed, which are new phenomena in multi-level atomic systems. Chapter 1 reviews some basic concepts to be used in later chapters, such as the nonlinear susceptibility, coherence functions and doubledressing schemes. Chapter 2 extends the previous results on polarization beats to include both difference-frequency femtosecond and sum-frequency attosecond beats in the multi-level media depending on the specially arranged relative time delays in the multi-colored laser beams. Chapter 3 gives results on Raman, Raman-Rayleigh, Rayleigh-Brillouin, and coexisting Raman-Rayleigh-Brillouin-enhanced polarization beats. Chapter 4 presents multi-dressing FWM processes in confined and non-confined atomic systems with specially-designed spatial patterns and phase-matching conditions for laser beams. Chapter 5 shows enhancement and suppression in FWM processes in multi-level atomic media, generated FWM signals can be selectively enhanced and suppressed via an EIT window. The evolution of dressed effects can be from pure enhancement into pure suppression in degenerate-FWM processes. Chapter 6 demonstrates the modification and control of MWM processes by manipulating the dark-state or EIT windows with polarization states of laser beams via multiple Zeeman sublevels. Chapter 7 shows spatial dispersion properties of the probe and generated FWM beams which can lead to spatial shift and splitting of these weak laser beams. Chapter 8 presents the observations of several novel types of solitons, such as gap, dipole, and vortex solitons, for generated FWM beams in different experimental parametric regions.

Authors believe that this monograph treats some special topics of coherent controls of FWM and MWM and can be useful to researchers interested in related nonlinear MWM processes. Several features presented here are distinctly different and advantageous over previously reported works. For example, authors have shown evolutions of enhancement and suppression of FWM signals due to various dressing schemes by scanning the dressing field detuning. Also theoretical calculations are in good agreement with experimentally measured results in demonstrating enhancement and suppression of MWM processes. Efficient spatial-temporal interference between FWM and SWM signals generated in a four-level atomic system has been carefully investigated, which exhibits controllable interactions between two different (third- and fifth-) order nonlinear optical processes. Such controllable high-order nonlinear optical processes can be used for designing new schemes for all-optical communication and quantum information processing. Authors also experimentally demonstrate that by arranging the strong pump and coupling laser beams in specially-designed spatial configurations (to satisfy phase-matching conditions for efficient FWM processes), generated FWM signals can be spatially shifted and split easily by the cross-phase modulation (XPM) in the Kerr nonlinear medium. Moreover, when the spatial diffraction is balanced by XPM, spatial beam profiles of FWM signals can become stable to form spatial optical solitons. For different input orientations and experimental parameters (such as laser powers, frequency detunings, and temperature), novel gap, vortex, and dipole solitons have been shown to exist in the multi-level atomic systems in vapour cell. These studies have opened the door for achieving rapid responding all-optical controlled spatial switch, routing, and soliton communications.

This monograph serves as a reference book intended for scientists, researchers, advanced undergraduate and graduate students in nonlinear optics.

We take this opportunity to thank many researchers and collaborators who have worked on the research projects as described in this book.

Yanpeng Zhang
Zhiqiang Nie
Min Xiao
October 2010

# Contents

# 1 Introduction

The subjects of this book focus on mainly around two topics. The first topic (Chapters 2 and 3) covers the ultrafast four-wave mixing (FWM) polarization beats due to interactions 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 delays in multi-colored laser beams. The polarization beat signal is shown to be particularly sensitive to the statistical properties of the Markovian stochastic light fields with arbitrary bandwidth. Also, the Raman, Raman-Rayleigh, Rayleigh-Brillouin, coexisting Raman-Rayleigh-Brillouin-enhanced polarization beats due to color-locking noisy field correlations have been studied. Polarization beats between various FWM processes are among the most important ways to study transient properties of media. The second topic (Chapters 4 – 8) relates to the frequency domain and spatial interplays of FWM processes induced by atomic coherence in multi-level atomic systems. FWM processes with different kinds of dual-dressed schemes in ultra-thin, micrometer and long atomic cells, selectively enhanced and suppressed FWM signals via an electromagnetically induced transparency (EIT) window are described, co-existing FWM and six-wave mixing (SWM) processes, especially temporal and spatial interference between them in multi-level EIT media are presented in Chapter 4. Furthermore, These effects of spatial displacements and splitting of the probe and generated FWM beams, as well as the observations of gap soliton trains, vortex solitons of FWM, stable multicomponent vector solitons consisting of two perpendicular FWM dipole components induced by nonlinear cross-phase modulation (XPM) in multi-level atomic media, are shown and investigated in Chapters 5 – 8. Experimental results will be presented and compared with the theoretical calculations throughout the book. Also, emphasis will be given only to the works done by the authors' groups in the past few years. Some of the works presented in this book are built upon our previous book (*Multi-wave Mixing Processes* published by High Education Press & Springer 2009), where we have mainly discussed the co-existence and interactions between efficient multi-wave mixing (MWM) processes enhanced by atomic coherence in multilevel atomic systems. Before starting the main topics of this book, some basic physical concepts and mathematical techniques, which are useful and needed in the later chapters, will be briefly introduced and discussed in this chapter.

## 1.1 Nonlinear Susceptibility

For over a decade, one of useful nonlinear optical techniques is the so-called "noisy" light spectroscopy. The ultrashort time resolution of material dynamics has been accomplished by the interferometric probing of wave mixing with broadband, nontransform-limited noisy light beams. The time resolution is determined by the ultrafast correlation time of noisy light and not by its temporal envelope, which is typically a few nanoseconds [1–6]. Such the noisy light source is usually derived from a dye laser modified to permit oscillation over almost the entire bandwidth of the broadband source. The typical bandwidth of the noisy light is about 100/cm, and has a correlation time of 100 fs [7]. In fact, the multimode broadband light has an autocorrelation time similar to the autocorrelation time of the transform-limited femtosecond laser pulse of equivalent bandwidth, although the broadband light can, in principle, be a continuous wave (cw).

In order to describe more precisely what we mean by optical nonlinearity, let us consider how the dipole moment per unit volume, or polarization $\boldsymbol{P}$, of a material system depends on the strength $\boldsymbol{E}$ 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 $\boldsymbol{P} = \boldsymbol{P}_L + \boldsymbol{P}_{NL}$ [8, 9]. Here, $\boldsymbol{P}_L = \boldsymbol{P}^{(1)} = \varepsilon_0 \boldsymbol{\chi}^{(1)} \cdot \boldsymbol{E}$ and $\boldsymbol{P}_{NL} = \boldsymbol{P}^{(2)} + \boldsymbol{P}^{(3)} + \cdots = \varepsilon_0(\boldsymbol{\chi}^{(2)} : \boldsymbol{E}\boldsymbol{E} + \boldsymbol{\chi}^{(3)} \vdots \boldsymbol{E}\boldsymbol{E}\boldsymbol{E} + \cdots)$.

When we only consider the atomic system (which are isotropic and have 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 terms with even-order powers in the applied field strength vanish [8]. The lowest order 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 the third- and fifth-order nonlinear optical susceptibilities, respectively.

FWM refers to nonlinear optical processes with four interacting electromagnetic waves (i.e., with three applied fields to generate the fourth field). In the weak interaction limit, FWM is a pure third-order nonlinear optical process and is governed by the third-order nonlinear susceptibility [8]. 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 $\boldsymbol{k}_1, \boldsymbol{k}_2$, and $\boldsymbol{k}_2'$. $E_1(\omega_1), E_2(\omega_2)$, and $E_2'(\omega_2)$ denote the three input laser fields. Here, $\omega_i$ and $\boldsymbol{k}_i$ are the frequency and propagation wave vectors of the $i$th beam. We can choose to have a small angle $\theta$ between the input pump laser beams $\boldsymbol{k}_2$ and $\boldsymbol{k}_2'$. The probe laser beam (beam $\boldsymbol{k}_1$) propagates along a direction that is almost opposite to that of the beam $\boldsymbol{k}_2$ (see Fig. 1.1). The corresponding nonlinear atomic polarization $P^{(3)}(\omega_1)$ along the $i(i = x, y)$ direction, from first-order perturbation theory, is given by [10]

$$P_i^{(3)}(\omega_1) = \varepsilon_0 \sum_{jkl} \chi_{ijkl}^{(3)} E_{1j}(\omega_1) E_{2k}'^{*}(\omega_2) E_{2l}(\omega_2), \tag{1.1}$$

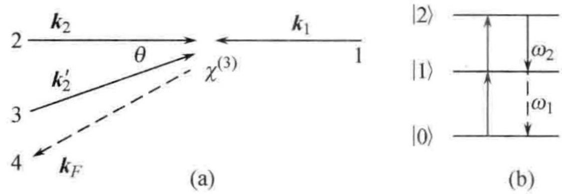


**Fig. 1.1.** (a) Schematic diagram for the phase-conjugate FWM process. (b) Energy-level diagram for FWM in a close-cycled three-level cascade system.

where the third-order susceptibility contains the microscopic information about the atomic system. The susceptibility of the nonlinear tensor $\chi_{ijkl}^{(3)}(\omega_F; \omega_1, -\omega_2, \omega_2)$ is also related to polarization components of incident and generated fields. For an isotropic medium, as in the atomic vapor, only four elements are not zero, and they are related to each other by $\chi_{xxxx} = \chi_{xxyy} + \chi_{yxxy} + \chi_{yxyx}$. For the generated SWM signal $E_S$ (fields $E_2$ and $E_3$ propagate along the direction of beam 2 and $E_2'$ and $E_3'$ propagate along beam 3), the fifth-order nonlinear polarization $P^{(5)}(\omega_1)$ along the $i(i = x, y)$ direction is then given by

$$P_i^{(5)}(\omega_1) = \varepsilon_0 \sum_{jklmn} \chi_{ijklmn}^{(5)} E_{1j}(\omega_1) E_{2k}'^{*}(\omega_2) E_{2l}(\omega_2) E_{3m}'^{*}(\omega_3) E_{3n}(\omega_3), \tag{1.2}$$

where $\chi_{ijklmn}^{(5)}$ is the fifth-order nonlinear susceptibility. For an isotropic medium, there are sixteen nonzero components and only fifteen of them are independent because they are related to each other by

$$\begin{aligned} 3\chi_{xxxxxx} = {} & \chi_{yyxxxx} + \chi_{yxyxxx} + \chi_{yxxyxx} + \chi_{yxxxyx} + \chi_{yxxxxy} + \\ & \chi_{xyyxxx} + \chi_{xyxyxx} + \chi_{xyxxyx} + \chi_{xyxxxy} + \chi_{xxyyxx} + \\ & \chi_{xxyyxx} + \chi_{xxyxyx} + \chi_{xxyxxy} + \chi_{xxxyxy} + \chi_{xxxxyy}. \end{aligned} \tag{1.3}$$

## 1.2 Coherence Functions

Lasers are inherently noisy devices, in which both phase and amplitude of the field can fluctuate. There are many different stochastic models to describe laser fields. However, since many models of fluctuating laser fields have identical second-order field correlations, differences among them will become important only if observable effects depend on higher-order field correlations. Noisy light can be used to probe atomic and molecular dynamics, and it offers an unique alternative to the more conventional frequency-domain spectroscopies and ultrashort time-domain spectroscopies [1, 11 – 14]. When the laser field is sufficiently intense that multi-photon interactions occur, the laser spectral bandwidth or spectral shape, obtained from the second-order correlation function, is inadequate to characterize the field. Rather than using higher-order correlation functions explicitly, three different Markovian fields are considered: i.e., (a) the chaotic field, (b) the phase-diffusion field, and (c) the Gaussian-amplitude field.

If laser sources have Lorentzian line shape, we have the second-order coherence function $\langle u_i(t_1)u_i^*(t_2)\rangle = \exp(-\alpha_i|t_1-t_2|)$ (i.e., $\langle|u_i(t)|^2\rangle = 1$ when $t = t_1 = t_2$). Here, $\alpha_i = \delta\omega_i/2$, with $\delta\omega_i$ being the linewidth of the laser with frequency $\omega_i$. On the other hand, if laser sources are assumed to have Gaussian line shape, then we have

$$\langle u_i(t_1)u_i^*(t_2)\rangle = \exp\Big\{-\Big[\alpha_i(t_1-t_2)/2\sqrt{\ln 2}\Big]^2\Big\}.$$

In the following, we only consider the former case. In fact, the form of the second-order coherence function shown above, which is determined by the laser line shape, is the general feature of stochastic models [15].

In this section, three Markovian noise stochastic models, the chaotic field model (CFM), the phase-diffusion model (PDM), and the Gaussian-amplitude model (GAM) are considered at a high enough intensity level to fully appreciate the subtle features of FWM spectroscopy [16, 17].

First, in CFM, we assume that the pump laser is a multimode thermal source and $u(t) = a(t)e^{i\phi(t)}$, where $a(t)$ is the fluctuating modulus and $\phi(t)$ is the fluctuating phase. In this case, $u(t)$ has Gaussian statistics with its fourth-order coherence function satisfying [18] $\langle u_i(t_1)u_i(t_2)u_i^*(t_3)u_i^*(t_4)\rangle_{CFM} = \langle u_i(t_1)u_i^*(t_3)\rangle\langle u_i(t_2)u_i^*(t_4)\rangle + \langle u_i(t_1)u_i^*(t_4)\rangle\langle u_i(t_2)u_i^*(t_3)\rangle = \exp[-\alpha_i(|t_1-t_3| + |t_2-t_4|)] + \exp[-\alpha_i(|t_1-t_4| + |t_2-t_3|)]$. In fact, all higher order coherence functions can be expressed in terms of products of second-order coherence functions. Thus any given $2n$th-order coherence function may be decomposed into the sum of $n!$ terms, each consisting of the product of $n$ second-order coherence functions. The general expression can be obtained as, $\langle u_i(t_1)\cdots u_i(t_n)u_i^*(t_{n+1})\cdots u_i^*(t_{2n})\rangle_{CFM} = \sum_\pi \langle u_i(t_1)u_i^*(t_p)\rangle\langle u_i(t_2)u_i^*(t_q)\rangle \cdots \langle u_i(t_n)u_i^*(t_r)\rangle$, where $\sum_\pi$ denotes a summation over the $n!$ possible permutations of $(1, 2, \ldots, n)$.

Second, in PDM the dimensionless statistical factor can be written as $u(t) = e^{i\phi(t)}$ (i.e., $|u(t)| = 1$) with $\dot{\phi}(t) = \omega(t)$, $\langle\omega_i(t)\omega_i(t')\rangle = 2\alpha_i\delta(t-t')$, $\langle\omega_j(t)\omega_j(t')\rangle = 2\alpha_j\delta(t-t')$ and $\langle\omega_i(t)\omega_j(t')\rangle = 0$. The second-order coherence function for a beam with Lorentzian line shape is given by [19]

$$\begin{aligned}\langle u(t_1)u^*(t_2)\rangle &= \langle \exp i\Delta\phi\rangle \\ &= \exp\left[-\int_0^{t_1-t_2}(t_1-t_2-t)\langle\omega(t_1)\omega(t_1-t)\rangle \mathrm{d}t\right] = \exp(-\alpha|t_1-t_2|).\end{aligned}$$

Here, $\Delta\phi = \phi(t_1) - \phi(t_2) = \int_{t_2}^{t_1}\omega(t)\mathrm{d}t$ has Gaussian statistics, so that $\langle \exp i\Delta\phi\rangle = \exp(-\sigma^2_{\Delta\phi}/2)$. By the classical relation of linear filtering, we have

$$\sigma^2_{\Delta\phi} = L(t_1-t_2) = 2\int_0^{t_1-t_2}(t_1-t_2-t)\langle\omega(t_1)\omega(t_1-t)\rangle \mathrm{d}t.$$

Now, the fourth-order coherence function can be calculated, which can be written as

$$\begin{aligned}&\langle u_i(t_1)u_i(t_2)u_i^*(t_3)u_i^*(t_4)\rangle_{PDM} \\ &\quad= \exp\{-[L(t_1-t_3)+L(t_1-t_4)+L(t_2-t_3)+L(t_2-t_4)- \\ &\qquad L(t_1-t_2)-L(t_3-t_4)]\} \\ &\quad= \exp[-\alpha_i(|t_1-t_3|+|t_1-t_4|+|t_2-t_3|+|t_2-t_4|)]\times \\ &\qquad \exp[\alpha_i(|t_1-t_2|+|t_3-t_4|)] \\ &\quad= \frac{\langle u_i(t_1)u_i^*(t_3)\rangle\langle u_i(t_2)u_i^*(t_4)\rangle\langle u_i(t_1)u_i^*(t_4)\rangle\langle u_i(t_2)u_i^*(t_3)\rangle}{\langle u_i(t_1)u_i^*(t_2)\rangle\langle u_i(t_3)u_i^*(t_4)\rangle}.\end{aligned}$$

Furthermore, we have the general expression for the second-order coherence function as

$$\begin{aligned}&\langle u_i(t_1)\cdots u_i(t_n)u_i^*(t_{n+1})\cdots u_i^*(t_{2n})\rangle_{PDM} \\ &\quad= \frac{\prod_{p=1}^{n}\prod_{q=1}^{n}\langle u_i(t_p)u_i^*(t_{n+q})\rangle}{\prod_{p=1}^{n}\prod_{q=p+1}^{n}\langle u_i(t_p)u_i^*(t_q)\rangle\langle u_i(t_{n+p})u_i^*(t_{n+q})\rangle}.\end{aligned}$$

Finally, in GAM, one has $u(t) = a(t)$, where $a(t)$ is real and Gaussian, and fluctuates about a mean value of zero. The fourth-order coherence function of $u(t)$ satisfies [20]

$$\begin{aligned}&\langle u_i(t_1)u_i(t_2)u_i(t_3)u_i(t_4)\rangle_{GAM} \\ &\quad= \langle u_i(t_1)u_i(t_3)\rangle\langle u_i(t_2)u_i(t_4)\rangle + \langle u_i(t_1)u_i(t_4)\rangle\langle u_i(t_2)u_i(t_3)\rangle + \\ &\qquad \langle u_i(t_1)u_i(t_2)\rangle\langle u_i(t_3)u_i(t_4)\rangle \\ &\quad= \langle u_i(t_1)u_i(t_2)u_i(t_3)u_i(t_4)\rangle_{CFM} + \langle u_i(t_1)u_i(t_2)\rangle\langle u_i(t_3)u_i(t_4)\rangle \\ &\quad= \exp[-\alpha_i(|t_1-t_3|+|t_2-t_4|)] + \exp[-\alpha_i(|t_1-t_4|+|t_2-t_3|)] + \\ &\qquad \exp[-\alpha_i(|t_1-t_2|+|t_3-t_4|)].\end{aligned}$$

In fact, according to the moment theorem for real Gaussian random variables, we have the general expression for the 2$n$th-order coherence function, as

$$\langle u_i(t_1)\cdots u_i(t_n)u_i(t_{n+1})\cdots u_i(t_{2n})\rangle_{GAM} = \sum_P \prod_{j\neq k'}^{2n} \langle u_i(t_1)u_i(t_k)\rangle,$$

where $\sum_P$ indicates the summation over all possible distinct combinations of the $2n$ variables in pairs.

## 1.3 Suppression and Enhancement of FWM Processes

In presence of a strong dressing field $G_2$, the dressed states $|+\rangle$ and $|-\rangle$ can be generated with the separation $\Delta_\pm = 2|G_2|$, as shown in Fig. 1.2. When scanning the frequency of the dressing field, we can obtain the EIT for the probe field and a suppressed FWM signal [Fig. 1.2 (b)], or electromagnetically induced absorption (EIA) for the probe field and an enhanced FWM [Fig. 1.2 (c)]. For the probe field propagating through the medium, we define the baseline versus the dressing field detuning $\Delta_2$ to be at the probe field intensity without the dressing field. Thus, this baseline is just the Doppler-broadened absorption signal of the material. With $G_2$ beam on, we can obtain one EIT peak at $\Delta_1 + \Delta_2 = 0$, where the transmitted intensity is largest comparing to the baseline. Since there is no energy level in the original position of $|1\rangle$ and the probe field is no longer absorbed by the material, the degree of transmission (or suppression of absorption) of the probe field is highest. An EIA dip is obtained at $\Delta_1 + \Delta_2 = |G_2|^2/\Delta_1$, where the transmitted intensity is smallest compared to the baseline. The reason is that the dressed state $|+\rangle$ or $|-\rangle$ is resonant with the probe field which is absorbed by the material and the degree of transmission (or enhancement of absorption) of the

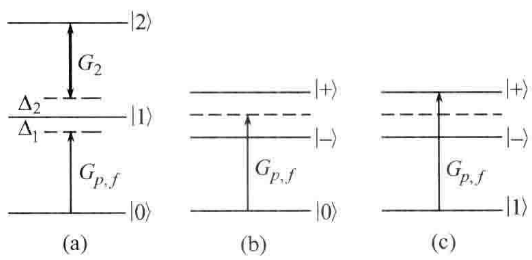


**Fig. 1.2.** (a) The diagram of the three-level ladder-type system with a dressing field $G_2$ (and detuning $\Delta_2$). The dressed-state pictures of the (b) suppression (EIT) and (c) enhancement (EIA) of FWM $G_f$ (or probe field $G_p$ with detuning $\Delta_1$) for the two-level system, respectively.