



Fudan Series in Graduate Textbooks



Nonlinear Optics

非线性光学

钱士雄 朱荣毅 编著

Shixiong Qian Rongyi Zhu

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Abstract

This textbook discusses the basic principles and some applications of a rapidly developing branch, nonlinear optics (NLO), in the optics science. Nonlinear optics emerged in 1961 and has got well matured in past forty years. As the volume of this textbook is limited, we will only talk about the most important principles and developments in nonlinear optics.

Chapter 1 is the introduction to the graduates about the forty years development of nonlinear optics. Chapter 2 relates to the basic theory of the nonlinear optical susceptibility. Chapters 3 and 4 deal with the second-order nonlinear optical phenomena. Chapter 5 is an overview of the third-order NLO effects. From Chapter 6 to chapter 10, four-wave mixing, stimulated scattering, resonant interaction, optical fiber the ultrashort lasers and ultrafast processes are discussed in order.

For the real teaching program, more materials should be added. This textbook can act as a general description of nonlinear optics for people who are interesting in this branch.

PREFACE

In the past three hundred years, from seventeenth century to the middle of twentieth century, many famous scientists, including Isaac Newton, Augustin Jean Fresnel, James Clerk Maxwell and Albert Einstein have made great contributions to the development of the optics science.

Their research achievements let people know the concepts of refraction, diffraction and interference of the light, and thus the geometrical optics and the wave theory of light were successfully established. The wave theory of the light could nearly explain all the optical phenomena observed at that period and has dominated the optics science for more than two hundred years. It came to be perfect when Maxwell developed the electromagnetic wave theory in the middle of nineteenth century.

However, the wave theory of light faced a great challenge in the beginning of last century after the discoveries of photoelectric effect and atomic spectra. The establishment of the quantum theory of light soon led to the concept of wave-particle duality of the light, which unified the wave theory and the photon concept. After that, the optics science was thought to be fully explored. In comparison to the great progress later in other fields of physics science, such as condensed matter, semiconductor physics, nuclear physics and high energy physics, the optics came to a period without encouraging success.

Nevertheless, the scientists in optics field did not stop their research paces at all, they devoted themselves to explore new and undiscovered areas in the optics science. Since 1950, many scientists considered the possible application of the stimulated emission and pointed out

that there would exist very interesting phenomena, *i. e.*, microwave amplification of the stimulated emission radiation (MASER) and light amplification of the stimulated emission radiation (LASER). These new ideas were successfully carried into realities in 1960's.

The proposal of laser device and the invention of the ruby laser were the mark of new milestone in the development of the optical science, which opened a door for the human being to have a very powerful and wonderful tool to study the nature in deeper insight and to create many novel devices.

Since then, a lot of different laser systems have been built using gases, vapors, liquids, glasses, crystals and semiconductors as operating media, and lasers using He-Ne, CO₂, dye, Nd-doped glass, Nd:YAG and Ti:sapphire have been widely used as laboratory instruments and utilized for the industrial applications. Employing the new and marvelous laser sources, people have found a great number of interesting phenomena, which were unbelievable before the invention of the laser. We are now in the new epoch of the optics science.

Among these achievements, nonlinear optics is a very attractive and wonderful branch. It has thoroughly changed the ideas existed in old optics science and has demonstrated many new unimaginable effects. Due to the interaction between light and matter, the frequency of the light wave could be changed and tuned, the absorption constant and the refraction index of the medium would not be kept as constants, many high lying energy states of the atoms and molecules could be detected by using laser beam with high intensity, the pulse duration of the laser beam could reach femtosecond (10^{-15} second) and even shorter, *etc.* The fruitful research successes in nonlinear optics branch make us so exciting that we can do what we want to do.

Over the past forty years, the research progress in the nonlinear optics achieved abroad and in China have been widely touching the human society and our daily life. Nowadays, people are no longer unfamiliar with the nonlinear optical effects. The course of nonlinear optics has been becoming a popular professional course in the universities and colleges for either undergraduates or graduate students in the Department of Physics and Department of Optical science and engineering. It is of course an obligatory course for the students who major in optics.

Based on the teaching experiences of the course "Nonlinear optics" for years in Fudan University, we feel that an appropriate textbook in English is indispensable. This motivates us to write this book. The book contains mainly the fundamental of the nonlinear optics, including the basic principles of nonlinear optics, the recent progresses and the important applications of nonlinear optics in modern science and technology. It is not possible and necessary to include all aspects of nonlinear optics in a concise text like this. The content of this book consists of ten chapters to be taught in one semester with three class hours each week. The chapters 2, 3 and 4 of this book were written by Rongyi Zhu, and the remaining chapters were written by Shixiong Qian.

The authors

June, 2005

前 言

非线性光学作为近代光学学科中最为活跃和发展最为迅猛的分支之一,在新型的高强度光源——激光器问世后,脱颖而出,成为光学研究领域中的一个热点。

非线性光学的出现极大地改变了人们对于光学的认识。以前人们采用线性光学的理论处理和解释了几百年来所观察到的光学现象,包括光的折射、衍射和干涉现象。线性光学的理论认为,光在传播过程中保持它的频率不变,各种物质有着固定的光学常数,如吸收系数、折射率和色散关系等。但利用激光器所进行的研究工作却揭示光学现象远非全是线性的。

采用非线性光学的理论可以解释人们所观测到的大量新现象和新效应,说明了在极强的光场下光和物质相互作用的基本规律。

本书将这个新领域介绍给读者,旨在使他们了解非线性光学的基本原理和最新的研究成果,教材着重于最基本的物理概念和处理方法,并对非线性光学的几个主要研究领域给出简单扼要的综述介绍,使他们在学完本课程后对非线性光学有一个基本的了解。

本书共十章,第一、第五至第十章由钱士雄执笔,第二至第四章由朱荣毅执笔。

谨将本书献给复旦大学 100 周年校庆。

编者

2005 年 6 月

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1.1 *From Linear Optics to Nonlinear Optics*

Nonlinear optics (NLO), as a fruitful and interesting branch in optics, emerged soon after the operation of the first ruby laser. Based on the great efforts done by many scientists, great successes have been achieved in establishing the basic principles, discovering new NLO effects and materials, and exploiting applications of NLO.

Before the invention of the laser, the optics was basically a field of linear optics. During the propagation, the light keeps its frequency wherever it propagates, and several light beams do not interact on each other in the overlapping area. The optical constants of the matter, such as absorption constant, refractive index and dielectric constant, are always kept unchanged at a given wavelength and are independent on the intensity of the input light. The light can only be generated from various light sources or from the excited states of the matter, and intense light could not be generated by the interaction between the light beam and the non-absorbing materials. These concepts have been accepted by people for several hundred years and were taken as the basic rules in the optics.

In the frame of linear optics, the electric polarization of the medium is linearly proportional to the amplitude of the incident electric field. Usually, the linear electric susceptibility $\chi^{(1)}$ is a complex tensor, and the relation between the electric polarization $\mathbf{P}$, the amplitude of electric field $\mathbf{E}$ and the susceptibility is expressed as

$$\mathbf{P}(\mathbf{r}, t) = \varepsilon_0 \chi^{(1)} \mathbf{E}(\mathbf{r}, t), \quad (1.1-1)$$

where ε_0 is the permittivity of free space, $\mathbf{r}$ is the position vector, and t is the time. Starting from Eq. (1.1-1) and combining the famous Maxwell's equations, we get the following wave equation of the light field:

$$\nabla^2 \mathbf{E}(\mathbf{r}, t) - \mu_0 \sigma \frac{\partial \mathbf{E}(\mathbf{r}, t)}{\partial t} - \mu_0 \varepsilon_0 \frac{\partial^2 \mathbf{E}(\mathbf{r}, t)}{\partial t^2} = \mu_0 \frac{\partial^2 \mathbf{P}(\mathbf{r}, t)}{\partial t^2}, \quad (1.1-2)$$

where μ_0 is the magnetic permeability of free space, and σ is the conductivity of the matter. All propagation processes, such as absorption, refraction and dispersion of the light, can be well treated based on the linear optics. In general, the real part of the linear electric susceptibility is related to the refraction and dispersion, while the imaginary part of the susceptibility produces the absorption.

However, the basic concepts of linear optics have been greatly changed due to the emergence of laser, and the principles of the traditional linear optics could not explain many new optical effects discovered after 1960.

In fact, before the invention of laser, several unusual phenomena have been found, for example, the dependence of the refractive index of the matter on the applied static electric field. In these electro-optic phenomena, when the change of the refractive index of the matter is proportional to the amplitude of the applied static electric field, the effect is called Pockels effect. While in another phenomenon called Kerr effect, the change of the refractive index is proportional to the square of the amplitude of the applied static electric field. Both effects were found in nineteenth century and have been widely used for the modulation and the switching of the light. In addition, there appeared two effects. One is Raman effect found in 1928, where new emissions with the frequency shift related to the vibrational modes of the molecule were detected. Another one is the fluorescence emission known pretty early, which was produced by the radiative relaxation of the electron from the excited state to the ground state. However, the signals generated in these two effects do depend linearly on the amplitude of the applied optical field, so they are actually not the NLO effects, but are assigned as spontaneous Raman scattering and spontaneous emission, respectively.

As the laser beam has much higher power than the normal light sources, such as sun light, mercury lamp and other lamps, new effects

could be easily generated. Even at the early stage of laser epoch, the power density of the laser beam at the focal point could reach the order of GW/cm^2 , thus many NLO effects were observed. Under the radiation of laser beam, intense light beams with frequencies different from that of the incident laser beam can be really generated from the transparent crystals and materials, and the light beams can interact in the overlapping area accompanying with the energy transfer and/or the phase transfer from one beam to another. The absorption constant and the refractive index of the matter are found being not constant, but are intensity-dependent. The light intensity transmitted through the matter depends not only on the input intensity but also have several output values at a fixed input intensity. The above effects are all NLO effects, such as second harmonic generation (SHG), sum frequency generation (SFG), stimulated Raman scattering (SRS), and two-photon absorption (TPA).

All the newly discovered phenomena could not be explained by the linear treatment and they have resulted in a rapid development of the NLO theory. The expression used for explaining the linear optical effects can no longer be used to interpret these new phenomena, so Eq. (1.1-1) must be modified by taking the higher-order terms of the electric polarization into consideration as following:

$$\mathbf{P}(\mathbf{r}, t) = \epsilon_0 \chi^{(1)} \mathbf{E} + \epsilon_0 \chi^{(2)} : \mathbf{E}\mathbf{E} + \epsilon_0 \chi^{(3)} : \mathbf{E}\mathbf{E}\mathbf{E}. \quad (1.1-3)$$

In this expression, the first term in the right side is the linear term, while the second term corresponds to the second-order NLO effects and the third term can be used to explain the third-order NLO effects. $\chi^{(2)}$ and $\chi^{(3)}$ are the second-order and third-order nonlinear electric susceptibilities of the medium, respectively. It can be expected that even the higher-order NLO effects could be possibly observed with the intense laser beam. In fact, the high-order harmonic generations (HHG) up to several hundred orders were generated in 1989.

1.2 The First Ten Year's Evolution

The first NLO phenomenon was discovered soon after 1960 by P. A. Franken when he took a spectrograph of the emission spectrum from the quartz plate illuminated by the ruby laser beam, and he found that there was a new emission line at 374 nm just half of the wavelength of the laser beam. It was soon realized that this is a second harmonic wave generated from the quartz plate. Although the birefringence of the quartz

could not fulfill the phase matching condition for SHG, this observation really opened the door of the NLO field. SHG was soon becoming a very important technique in transforming the long wavelength laser beam to the coherent radiation at shorter wavelength. This was a very essential significance in both the theoretical research and applications in NLO field. After that, a lot of NLO effects were quickly discovered. E. J. Woodbury observed another new emission line from the nitrobenzene liquid which was used as the Q-switching material in the laser cavity. This observed 766 nm signal was interpreted as the SRS emission by the pumping of ruby laser beam. Since that, SRS effect has been observed from different kinds of materials, including gases, liquids and crystals, and was used as another method to produce the coherent radiations at various wavelengths. By using high resolution spectroscopic technique, people also observed new emission lines with very small frequency shift from the incident laser beam. These new lines were assigned to the stimulated Brillouin scattering (SBS).

Besides SHG, other second-order NLO effects including SFG, optical parametric oscillation (OPO), optical parametric amplification (OPA) and difference frequency generation (DFG) were also observed. Since the second-order NLO effects have many potential applications in the wavelength extension of the coherent radiation and can only occur in the materials without inversion symmetry, the exploration of NLO crystals with high SHG conversion efficiencies becomes an important task in NLO field. The use of SHG, SFG, DFG and OPO techniques has extended the coherent radiation from ultraviolet to the near infra-red regions, improving greatly the experimental capabilities.

In general, the third-order electric polarization is proportional to the cubic of the amplitude of the incident optical electric field, thus the occurrence of the third-order NLO effects needs higher power density. Due to the progress in mode-locking technique, picosecond (ps) lasers built in the 60s of the last century have been used to study more NLO effects, including the third-order NLO effects.

Two-photon absorption phenomenon was first observed in crystals, where two photons were simultaneously absorbed by one molecule, resulting in the excitation of the molecule to a high excited state which could not be reached by one photon absorption. As two counter-propagating laser beams could greatly reduce the Doppler broadening, the discovery of two-photon absorption stimulates greatly the development of the high-resolution laser spectroscopy.

The saturable absorption (SA) is another phenomenon that reveals the change of absorption constant of the matter under the action of high power beam. Within the inhomogeneous absorption band, an absorption dip is produced at the band center. This new technique was soon employed to make the frequency standard by using atomic vapor and gas, such as Rb, Cs and Ne. One of its important applications is to improve the accuracy of the measurement of the light velocity.

Relying on the short duration of the laser pulses, a new kind of optical phenomena, *i. e.*, the transient optical phenomena, were observed and developed progressively. All these phenomena, including optical echo, optical nutation, self-induced transparency and free-induced decay, could be well explained by the semiclassical theory in the laser physics.

Based on the great progress in the experimental researches during this period, N. Bloembergen and P. Butcher wrote their famous books in NLO field, named as "Nonlinear optics" and "Nonlinear optical phenomena", respectively, which summarized the up-to-date developments of NLO in that period and used the density matrix theory to derive the theoretical formulation for the nonlinear electric susceptibility. These two books have greatly deepened our understanding on the observed NLO experimental evidences.

1.3 Rapid Development Period of Nonlinear Optics

From 1970 to 1990, accompanying with the great successes in laser science and technology, many new effects were exploited, including the optical bistability (OBIS), soliton, squeezed state, HHG, *etc.* In the domain of laser technique, dramatic progresses in shortening the pulse duration further to femtosecond (fs) scale have greatly stimulated the researches of ultrafast processes in photophysics, photochemistry and photobiology. The applications of advanced laser techniques have led to the great successes in optical fiber communications, laser manufacturing of microstructures, *etc.* All these fruitful achievements have demonstrated the very powerful and marvelous functions of the laser in the development of modern science and technology.

Four-wave mixing (FWM) found in the first decade of the laser epoch has rapidly developed in these twenty years. The unique features of FWM are the recovery of the phase and the correction of the phase aberration, which are very attractive in many applications. A special tech-

nique is degenerate four-wave mixing (DFWM), where two counter-propagating pump beams are used to generate a new beam in the opposite direction of the probe beam. This new beam has the complex conjugate relation with the probe beam. In such case, the phase aberration introduced by the propagation in atmosphere or other media could be effectively corrected, making DFWM to be very promising in the applications for the laser propagation in long-range distance, and the self-tracking system used for the laser fusion. If the resonant DFWM configuration was used, the reflectivity of the probe beam would be larger than 100 %. DFWM has been used to build the phase conjugate mirror in the laser cavity to eliminate the phase distortion caused by the thermal effect in the laser rod. In the optical information processing, DFWM could be used as the devices for deconvolution, Fourier transformation and character recognition. The near degenerate four-wave mixing (NDFWM) has the character of very narrow reflection band, which could act as the filter with very narrow frequency bandwidth.

A special configuration of FWM called coherent anti-Stokes Raman scattering (CARS) was studied during this period. The unique advantage of CARS technique is that it could avoid the interference of the fluorescence in the measurement and improves greatly the signal to noise ratio. One of its important applications is to measure the temperature and the composition content of the combustion system and furnace. CARS technique has also been used in studying the dephasing process of molecules and semiconductors due to its coherent feature.

Two important stimulated processes, SRS and SBS, have been valuably applied in the extension of the wavelength range of the coherent radiation. Due to the rapid improvement of the laser performance, laser pulses with wide range of duration covering nanosecond (ns), ps and even fs scales could be obtained and the pulse energy could be raised to a higher level. By using these powerful laser beams, SRS and SBS processes in the media were carefully studied and well understood. Very intense SRS and SBS radiations were got by intense pumping. The most attractive medium is the high pressure gas, including hydrogen, methane and deuterium. As the hydrogen has the largest Raman shift of 4155 cm^{-1} among the molecules, it is attractive in the application for the large wavelength extension of the coherent radiation. It was found that hydrogen in high pressure cell could generate very efficient Raman conversion. Conversion efficiency of 45 % for first Stokes SRS was achieved in 1980 and multi-order anti-Stokes SRS outputs were also generated. The