1.2 MAJOR EVENTS IN HISTORY OF NONLINEAR OPTICS

In this section we shall briefly review some (not all) major events that brought significant or considerable influences on the formation and growth of nonlinear optics.

As we mentioned in the preceding section, the formation of nonlinear optics originated in the early 1960's. The discovery of the optical second-harmonic generation (1961) was commonly recognized as the first milestone of the formation of nonlinear optics.[3] Shortly after that, several other optical frequency-mixing effects were sequentially demonstrated based on the use of laser radiation, which included the optical sum-frequency generation (1962),[5] optical third-harmonic generation (1962),[6] optical rectification (1962),[7] optical difference-frequency generation (1963),[8,9] and the optical parametric amplification and oscillation (1965).[10,11] These experimental demonstrations not only verified the validity of nonlinear polarization theories[4] but also provided an alternative approach to generate coherent optical radiation.

During the same time period, another important event was the discovery of stimulated Raman scattering (1962),[12,13] which can be recognized as the second milestone in the history of nonlinear optics. The significance of this discovery is that, for the first time, the 'stimulated' nature of light scattering excited by an intense laser beam was revealed. The stimulated scattering is an alternative physical approach to generate the coherent optical radiation without the need of population inversion. Later, researchers reported the observation of another major nonlinear optical effect, stimulated Brillouin scattering (1964),[14] which arose from the interaction of an intense monochromatic optical field with the induced hypersonic field in a scattering medium through the so-called optical electrostriction mechanism. Since then, the stimulated Brillouin scattering has become an efficient technique to generate or amplify the coherent optical radiation with a small frequency-shift or fine tunability. In principle, this effect can also be employed to generate or amplify a coherent hypersonic field in the optical medium.

Another major subject of nonlinear optics is related to the refractive-index change induced by an intense laser beam as well as the impact of this change on the laser beam itself. An important article focused on this issue was published in 1964 with a conceptual discussion and a semi-quantitative description of the self-focusing (self-trapping) behavior of an intense optical beam propagating in a nonlinear medium,[15] although the similar concept of self-trapping was mentioned in another early paper.[16] Immediately, studies of the self-focusing effect attracted a great deal of attention because (i) this effect was often related to the optical damage occurring in solid-state lasing materials or optical elements and (ii) it was responsible for the observed 'anomalous' threshold decrease of some nonlinear processes (such as stimulated Raman scattering), owing to the dramatic increase of the local beam intensity inside a nonlinear medium. Further studies of dynamic self-focusing processes for short laser pulses revealed special properties of the moving-focus as well as the new phenomena, self-phase modulation and spectral self-broadening.[17-21] Now it is well known that the self-phase modulation and spectral self-broadening effects are among the basic mechanisms for generating ultra-short laser pulses and the continuum radiation with a super-broad spectral band.

Other kinds of nonlinear optical phenomena were reported in the 1960's, the so-called transient coherent optical effects, including photon echoes (1964),[22] self-induced transparency (1965,1967),[23,24] and optical nutation (1966,1968).[25,26] These effects are related to the transient-response behavior of a resonant optical medium interacting with short optical pulses or a fast-switched optical field. Some of them are the optical analog of the corresponding effects in nuclear magnetic resonance studies. The studies of transient optical effects can provide a new approach to investigate the relaxation behavior of resonant transitions in absorptive media.

In addition, there are other two fundamental nonlinear optical effects, the principles of which were already known before the invention of lasers. One is the so-called saturable absorption effect observed early in radio(microwave)-frequency spectroscopy. The logical inspiration from these observations was the idea of population inversion that led to the invention of masers and lasers.[27,28] Shortly after the invention of the first laser device, the optical saturable absorption in organic dye solutions and other materials were well studied and soon applied to the *Q*-switching and mode-locking of laser devices.[29-32] The other effect is two-photon absorption (TPA). Although the theoretical description of this effect was reported in the early 1930's,[33] the earliest experimental demonstrations of TPA-induced fluorescence were achieved with the use of laser radiation (1961).[34,35] The initial research work of laser-based TPA and the related processes stimulated an extensive investigation on two-photon and multi-photon induced absorption, fluorescence, ionization and dissociation. Later, all these kinds of studies have formed the important part of nonlinear optics and laser spectroscopy.

The 1970's was the second decade for the booming growth of nonlinear optics. During this period of time, numerous new nonlinear optical effects and novel techniques were further reported and could be summarized in the following three main areas. The first area was related to the invention of various novel nonlinear spectroscopic techniques, such as coherent anti-Stokes Raman spectroscopy (CARS),[36-39] Doppler-free saturation spectroscopy (1971),[40,41] Doppler-free TPA spectroscopy (1970,1974),[42-46] inverse Raman spectroscopy,[47-49] Raman gain spectroscopy,[50-52] and laser polarization spectroscopy (1976).[53-57] All these nonlinear spectroscopy techniques require two laser beams, and at least one of them should be tunable. In general, one laser beam (as a pump beam) is used to excite a selected group of atoms or molecules, and the second laser beam (as a probe beam) is used to detect the specific change of optical properties of samples owing to the select excitation. For gas samples, the longitudinal (first-order) Doppler broadening can be eliminated by the use of two counter-propagating laser beams. In this case, the spectral resolution is mainly limited by the transit-time broadening and the transverse (second-order) Doppler broadening. The former is limited by the transit time of a moving atom across the laser beam of a finite beam size; the latter is limited by the nonzero translational velocity of atoms. To eliminate the transit-time broadening, the so-called optical Ramsey fringes technique was theoretically proposed (1972, 1976)[58,59] and experimentally demonstrated (1976,1977).[60-63] The principle of this new method is based on the interaction of a directional atom/molecule beam with a periodically-spaced multi-beam coherent optical field or, alternatively, with a periodically-repeated coherent optical pulsed field. In both cases the effective interaction time of an atom system with the coherent optical field is increased; therefore the spectral resolution can be considerably improved according to the uncertainty principle. The other ultimate solution to eliminate both the transit-time broadening and transverse Doppler broadening is to reduce the translational velocity of atoms as low as possible. For this purpose the optical cooling or trapping spectroscopic technique was proposed.[64-67] In principle, there are two optical approaches that can be used to reduce the velocity of atoms and to trap them in a fixed position. One is based on the ponderomotive (or electrostrictive) force of a light field exerting on an atom system, which is proportional to the gradient of the square of the applied optical electric field. By using this method, a slow atom may be trapped in the nodes or antinodes of a three-dimensional optical standing wave field.[64,65] The other is based on the resonance radiation pressure force exerting on a resonant absorptive atom system;[66,67] this idea was supported by preliminary experimental results (1975,1978).[68-70] Studies on all of these novel spectroscopic effects have greatly enriched our knowledge and offered us many new spectroscopic techniques with a much higher spectral, spatial, and temporal resolution than the conventional spectroscopic techniques.

The second area was related to the studies of optical phase conjugation. In 1972, Russian researchers reported the experimental observation of the wave-front reversal property of backward stimulated Brillouin scattering.[71,72] Although this was the first observation of an optical phase-conjugation phenomenon, the physical understanding of this observed effect was not very clear for quite a long time. Therefore this observation did not attract enough attention until the theoretical suggestions of using special three-wave mixing and four-wave mixing to generate phase-conjugate waves were proposed (1976, 1977).[73,74] These two proposed methods were soon experimentally accomplished (1977).[75,76] After that, a great deal of efforts have been made in the research area of optical phase conjugation. Through the studies of optical phase-conjugation we have had a comprehensive understanding about various physical mechanisms that can be used to generate optical phase-conjugate waves. On the other hand, optical phase-conjugation techniques are useful for many special applications, such as the high-brightness lasing or amplification through a distorting gain medium, the aberration compensation in a disturbing propagation medium, the real-time optical holographic wave-front reconstruction, and the optical data storage and processing.

The third area is related to the studies of optical bistable effects. Although the original idea of an optical bistable device was reported in 1969 based on a saturable absorber in a F-P etalon,[77] researchers' attention really started to focus on this subject only after the first experimental demonstration of a real optical bistable device reported in 1975.[78] In this case, the mechanism of optical bistability was the optical field-induced refractive-index change of a nonlinear medium inside a F-P etalon,[79] which is called the nonlinear dispersion type of intrinsic optical bistable device. Another type of device, the so-called hybrid type of optical bistable device was demonstrated in 1977.[80] It was based on a second-order nonlinear electro-optical crystal placed inside a F-P etalon; the refractive-index change of the crystal was controlled by an external electric field that was proportional to the optical feedback from the F-P cavity. The significance of optical bistable studies is to explore possible ways to control light with light. The principles of optical bistability can be employed to accomplish a variety of vital functions such as optical switching, transistor, limiter, stabilizer, clipper, etc. In other words, optical bistable devices may be the key elements for optical fiber telecommunications, optical logical circuits, and optical computers in the future.

In the 1980's, nonlinear optics had been well established already. Although no many new effects or novel phenomena were reported, the momentum of nonlinear optics continuously increased in this decade. The research efforts focused on those areas such as nonlinear spectroscopic techniques with ultra-high spectral resolution or high temporal resolution, nonlinear optical applications for generation and control of ultra-short laser pulses, the optical phase-conjugation based on the degenerate four-wave mixing and backward stimulated scattering, optical bistability studies, photorefractive-effect studies, special techniques of second harmonic generation, and optical soliton studies.

1.3 FEATURES OF INTERACTION OF INTENSE LIGHT WITH MATTER

Regarding the interaction of light radiation with matter, the total number of newly discovered effects and phenomena after the advent of lasers is even larger than that of the optical effects and phenomena known before the invention of lasers. One may ask why so many new things can be found in a so short time period (only three to four decades). To answer this question, we should consider the essential differences between the light beams from laser devices and that from ordinary light sources. Only based on these differences, we can realize how powerful the laser radiation could be when it interacts with matter.

It is well known that the laser radiation is generated based on stimulated emission from a population inversion system, whereas the ordinary light is based on spontaneous emission from conventional light sources. Consequently, these two