

Nonlinear Optics and Laser- Matter Interaction: Theoretical and Experimental Approaches

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Chapter - 1

Introduction to Nonlinear Optics

Linear optics is an expansive and intricately woven field that thoroughly investigates, meticulously analyzes, and explores the multifaceted behaviors as well as the essential properties of electromagnetic fields, which are commonly and collectively known as light, as they traverse and propagate through a medium that is specifically characterized by a linear response. This precise and specific type of medium allows the intricate interaction dynamics of light to be finely modulated or adjusted to a very particular extent, contingent on how the light waves engage, interact, and resonate with the medium itself. It is fundamentally crucial to highlight and emphasize that other experimental parameters, which do not exert a significant influence on the overall propagation, scattering, or behavior of the light, remain consistently unchanged and unaffected throughout the entire intricate process of propagation. This particular scenario stands in stark contrast to the behavior typically observed in a nonlinear optical medium, where remarkably different and often unexpected phenomena can manifest in ways that challenge our conventional understanding. In nonlinear contexts, light beams exhibit a remarkable, unexpected, and often surprising ability to interact with one another in relation to all experimental variables that are involved in the complex and sophisticated optical setup. The unique and fascinating property of light interaction in nonlinear media opens the door to a diverse array of striking and captivating phenomena that are not merely limited but are absolutely impossible within the confines and restrictions of linear optics. In the broad and extensive realm of linear optics, marked by its principles, there exists a complete and utter absence of any meaningful interaction between the electromagnetic waveforms and the variations across numerous experimental parameters. This indicates that when distinct mixtures of electromagnetic waves are observed, assessed, or analyzed, the perceptible changes and differences that occur are solely linked, and can be thoroughly attributed, exclusively to modifications in the amplitude of these constituent waves. It is this distinctive and defining feature that serves as a fundamental characteristic of linear optics, and this characteristic distinctly separates it from all the nonlinear interactions that can and do occur within more complex and

multifaceted settings of optical experimentation. Furthermore, it is noteworthy and of great significance that linear optics can be accurately and precisely described through specific phenomenological models that effectively capture the core essence of all interactions involved in the propagation, modulation, and behavior of light waves in a linear medium. This predictability and clarity of interactions within linear optics stand in sharp contrast to the unpredictable and often bewildering nature of nonlinear optics, where the behavior of light does not strictly adhere to or follow any single, definitive law or universally accepted set of governing principles regarding interaction. The greatest challenge encountered in the field of nonlinear optics, however, arises from the intricate and often perplexing mathematical forms that describe the dynamic and complex reactions associated with nonlinearity itself. This level of complexity not only makes it particularly challenging but can frequently result in significant difficulties when attempting to represent these intricate and nuanced interactions in an effective and coherent manner within a unified mathematical framework that could potentially simplify comprehension for researchers and students alike. As a direct consequence of all these inherent complexities within the field, the study of nonlinear optics routinely involves a range of far more elaborate theoretical constructs and approaches, which are characterized by their elevated and heightened complexity when juxtaposed against the relatively straightforward linear counterpart. This greater level of intricacy necessitates a much deeper exploration and understanding of the fundamental physics and mathematics to fully comprehend, analyze, and interpret the captivating phenomena that become apparent when one operates within the challenging and multifaceted nonlinear optical regime. Therefore, the rigorous examination and evaluation of these principles and laws continue to remain a pivotal area of research that holds significant importance within the broader field of optics and photonics. The insights gleaned in this essential, foundational area can lead not only to remarkable advancements in both theoretical understanding and practical applications but also to an enhanced comprehension of light as a foundational aspect of the universe around us, illuminating the very fabric of existence itself [1, 2, 3, 4, 5, 6, 7].

Nonlinear optical phenomena encompass a profoundly rich and diverse array of interactions that involve not only low but also impressively high values of light intensity. These interactions typically take place over both remarkably short and extended time scales, providing a unique arena for exploration. The fascinating nature of these phenomena is explored from a myriad of perspectives that relate to the intricacies of different energy scales, which correlates with inquiries that span from the fundamental quantum mechanics governing electrons to the most recent and innovative applications

observed under intense radiation fields. The intricate and complex interaction between intense laser fields, the localized dynamics of electrons, and foundational theories concerning various states of matter come together, merged with a multitude of diverse concepts. Collectively, they form a vibrant and dynamic research domain that has significant implications. This expansive domain reaches out to all scientific communities that hold a vested interest in promoting advancements within modern science and technology, fostering interdisciplinary collaborations. Furthermore, a crucial component that plays a pivotal guiding role in contemporary research in nonlinear optics is the continuous technological advancement which enables the acquisition, control, and manipulation of intense light sources. These advances span across a remarkably broad spectrum of wavelengths, which is impressively extensive, encompassing yet not limited to X-rays, extreme ultraviolet light, soft X-rays, and delving down to the terahertz frequency range. The systematic investigation of all these emerging discoveries within a cohesive and overarching framework significantly represents one of the primary themes of this specific course, underscoring the importance of a comprehensive understanding. Although nonlinear optics is relatively young as a scientific discipline, numerous pivotal expansions have arisen and taken shape in this specialized field over the years. These expansions span essential areas, including plasma and particle beam physics, astrophysics, alongside various applications that have been witnessed in medicine, biology, and multiple engineering sectors. Many noteworthy technological and scientific innovations have undeniably emerged from the foundational concepts and empirical results that are yielded by ongoing nonlinear optical research endeavors, creating a lively interplay between theory and practice. This particular phenomenon arises as nonlinear interactions frequently facilitate beneficial and requisite energy conversion processes, the establishment of innovative analytical methodologies, non-intrusive measurements of vital physical data under extreme conditions, and highly optimized heating processes applicable to plasma. Each of these factors contributes significantly and meaningfully to the overall growth and continual development of this thrilling and rapidly evolving field of study. The quest to thoroughly understand and effectively manipulate such phenomena holds thrilling potential, not merely from an academic standpoint, but also in practical applications that may redefine the boundaries of what is possible in various scientific and industrial landscapes. This journey into exploration and discovery in nonlinear optics invites dreamers and innovators to push the limits of current knowledge, adhering to an ever-evolving narrative of science and technology [8, 9, 10, 11, 12, 13, 14, 15, 16].

1.1 Basic Principles of Linear Optics

In this sub-section, we present a comprehensive and expansive introduction to the fundamental principles that govern the captivating and multifaceted physics of light. These principles play an essential role in establishing the foundational framework of nearly all nonlinear effects that can be observed across a vast and diverse spectrum of laser-matter interaction processes. By thoroughly considering the propagation of light under the influence of relatively weak electromagnetic fields, one can discern that the fully symmetrical characteristics inherent to the electromagnetic field manifest through a robust and well-defined mathematical model of interaction. This intricately developed model yields linear relationships between the respective intensities of the electromagnetic fields involved and the observable outcomes that arise as a direct and immediate consequence of these interactions. Such linear phenomena are adeptly and effectively modeled through a comprehensive set of equations, along with classical field theory, which specifically pertains to the continuous and unimpeded propagation of light throughout various media. Among the most notable and well-studied linear optical effects, we frequently encounter reflection, refraction, and diffraction of light, each of which can also be elucidated through both ray theory and wave theories of light waves. In addition, the mathematical equations and formulations involved imply that the complex process of refraction can indeed be simplified to the fundamental laws of reflection that take place at various interfaces, particularly when closely examining scenarios involving totally reflecting interfaces. To further illustrate a straightforward yet insightful scenario of uniform propagation in a vacuum, we should note that light rays refract at angles that depend entirely and distinctly on the intrinsic relationship between the characteristics of the various materials present in the given context. As is commonly taught in high school physics curricula, for a specific medium characterized by a well-defined and known index of refraction, denoted as n , the wavelength of light, represented as λ , and its frequency, ν , are intricately interconnected through the fundamental electromagnetic wave speed, c , in a vacuum. In this important context, the term wavelength specifically refers to the spatial length of a single wave period, while the oscillation rate of both the electric field and the magnetic field oscillates consistently at a singular frequency, ν , appearing nearly in linear phase. Within this carefully detailed framework, ω serves as the angular frequency, while k signifies the wave number associated with the light waves being rigorously examined. In instances where the medium effectively and dynamically modulates its dielectric constant in response to variations in the intensity of the light field, phase, and/or polarization, the previously

mentioned linear qualitative scenarios diverge significantly and distinctly. This essential divergence gives rise to a vast and intricate array of nonlinear optical effects, which become critically important for a deeper understanding of more advanced and complex optical phenomena. On the experimental front, fundamental interferometric setups provide direct and unobstructed access to the five distinct electromagnetic wave parameters we discussed in depth earlier. Furthermore, the phenomena relating to the interactions between light and matter that have been appropriately illustrated above are also suitably modeled by linear optics, provided that the amplitude of the electromagnetic field remains sufficiently low for the linear approximation to hold true during observations. In stark and marked contrast, when laser field intensities increase significantly and nonlinear effects become increasingly prevalent within various materials and processes, this opens the door to the generation of several additional primary and secondary emitted frequency light signals. Consequently, it becomes abundantly clear that linear optics serves as an essential bridge that connects the distinct and informative realms of nonlinear optics, experimental atomic interactions, and modern light phenomena, alongside the foundational principles of classical electrodynamics. This connection thus leaves significant and important pathways open for thorough investigation, in-depth exploration, and groundbreaking research endeavors in future scientific inquiries and studies ^[17, 18, 19, 20, 21, 22].

Chapter - 2

Nonlinear Optical Effects

Starting from the fundamental concept that light is a specific term used to denote the very small segment of the electromagnetic spectrum that the human eye is capable of perceiving with clarity, we often find ourselves making the assumption that when we project an optical beam upon a given material, the resulting interaction, along with the ensuing behavior that we witness, is typically smooth, predictable, and linear in its nature. In fact, in the vast majority of instances, this holds true, and this scenario is indeed the one that we consistently observe throughout our natural surroundings, providing a comforting degree of certainty in our understanding of the world around us. However, it is quite fascinating to acknowledge that the interaction of light with a considerable array of materials, each possessing its own unique attributes and distinctive characteristics, can surprisingly lead to behavior that deviates drastically from what we might expect if we delve deeper, adopting a more nuanced and intricate context for our analysis. Consider, for instance, the vivid image of a pristine, crisp lettuce leaf, boasting a vibrant green color and a delicate, intricate texture, which catches the eye in a way that is reminiscent of a beautiful natural artwork meticulously crafted by nature itself. What does it truly look like when we carefully slice it meticulously with a sharp and well-maintained knife? We typically anticipate that the knife will sever the leaf in a tidy, clean manner, inevitably resulting in precise and evenly sized pieces that we can admire and serve to others in an appealing and enjoyable manner, thus making the act of preparation feel rewarding and satisfactory. Indeed, this is typically the case; however, it has been reliably substantiated through repeated observation, across many instances, that as the knife glides over the central hard rib of the leaf, an unexpected and notable curve emerges in the cut surface. This curve represents a distinctly observable 'nonlinear effect' that arises quite strikingly as a direct consequence of the very act of cutting, revealing the inherent complexities residing even in something as seemingly simple as slicing a green vegetable. The optical realm, too, encounters an analogously complex phenomenon of intrigue, reflecting a similar unpredictability that can evoke both fascination and puzzlement for those who diligently study the intricate mechanics of light and its interactions

with a diverse array of materials. Initially, a wide range of theoretical outcomes was achieved, which sought to quantitatively describe these fascinating nonlinear effects across different materials and under varied light interactions that could yield rich, diverse, and sometimes astonishing results, each illuminating a new dimension of our understanding of optics. Following that impressive period of inquiry, the first direct observations of such nonlinear optical effects began to be documented extensively, and these compelling predictions were subsequently validated in practice, gaining acceptance and widespread acknowledgment from the scientific community at large, thus bolstering the foundation of our knowledge in this important and evolving domain. Among some of the most well-known optical effects that have been both theoretically predicted and experimentally corroborated by diligent scientists and researchers include a variety of captivating phenomena such as self-focusing, multi-photon absorption, frequency mixing, harmonic generation, and even the remarkable occurrence of very high-order harmonic generation. Each of these distinct effects contributes uniquely to our understanding of light interactions and provides invaluable insights into the complexities of wave phenomena that govern much of what we experience in the world around us. These particular effects are undeniably of significant interest to researchers in the field, as they present an extensive variety of nonlinear phenomena that can unfold as a result of their intricate interactions under differing conditions and heterogeneous environments. The photons involved in these complex interactions, particularly those with asymptotically long wavelengths, surprisingly possess the remarkable ability to instigate the formation of new high-order harmonics while also facilitating increased levels of ionization in various mediums. This magical characteristic substantially expands the scope of potential applications for these intricate phenomena and effects that have deep implications for technology and science. As an intriguing, practical, and commercially viable application of these captivating effects, multi-wavelength sources have already emerged prominently in the marketplace, proving to be essential for groundbreaking advancements not only in advanced communications technologies but also in biophotonic imaging technologies that hold tremendous promise for transformative future healthcare applications and innovative therapeutic strategies. Such advancements could potentially revolutionize the landscape of patient care and medical treatment in the near future, assuring us that the usefulness of these discoveries extends far beyond mere theoretical interest. The previously mentioned effects, which span from frequency mixing through harmonic generation and even beyond, do not merely encompass the analysis and thorough elucidation of each of these complex phenomena but also involve

detailed examinations of their polarization states, energy transitions, and the various orders of harmonic generation that are meticulously observed throughout different media and materials. Furthermore, they introduce the underlying physics that governs these remarkable optical effects, allowing scientists and researchers to develop a sophisticated understanding and predict their behavior in various contexts and situations. This enriched comprehension is crucial for advancing the field and pushing the boundaries of what is possible in the innovative realm of optoelectronic applications, paving the way for new technological breakthroughs that can significantly enhance our daily lives. Additionally, various materials capable of manifesting this unique nonlinear behavior include a wide variety of substances such as aerosols, the ambient air, biological cells, diamond structures, and excited atoms subjected to high-intensity laser fields. These diverse substances contribute significantly to and enrich the vibrant discourse surrounding nonlinear optics. Moreover, femtosecond laser plasmas, glass beads, ions present in surface water, toxic airborne particulates, and many other substances populating our environment can also profoundly contribute to these fascinating phenomena, further complicating and enriching the theoretical landscape of optical interactions with matter in ways that continue to surprise researchers tirelessly exploring this domain. The development, characterization, theoretical exploration, and investigative experimentation pertaining to each of these remarkable optical effects is far from linear; rather, it is a deeply intricate tapestry of inquiry and research, and each topic constitutes a distinct and specialized line of research within the expansive area of optical interactions with matter. This captivating field continues to evolve and grow with every new inquiry into this compelling subject of study that constantly pushes the boundaries of human knowledge in illuminating and often unexpected ways. The mathematical approaches required to tackle these nonlinear optical effects, including several innovative perspectives that have not been elaborated upon in detail here, demand sophisticated instruments and methodologies capable of handling complex calculations with high efficiency and accuracy. As light behavior exhibits numerous nuances intertwined within its interactions, precise analytical tools and advanced experimental techniques are necessary for their fruitful exploration. Continued research in these vital areas, alongside technological advancements in the fields of lasers and materials development, will be explored further in future contributions specifically aimed at addressing unique challenges and exciting opportunities in the rapidly evolving realms of science and engineering. Additionally, this ongoing work ensures that our understanding of these complex phenomena continues to deepen and expand into new realms of application and insight, yielding fruits that not only open

doors to technological advancements that have yet to be imagined and realized but also promise to enhance the overall well-being and quality of life for individuals across the globe in profoundly meaningful and impactful ways [23, 21, 24, 25, 26, 27, 28, 29, 30].

2.1 Second Harmonic Generation

One of the most fundamental and simplest processes in the captivating and expansive realm of nonlinear optics is the intriguing phenomenon known as second harmonic generation (SHG). This remarkable and fascinating process is fundamentally regarded as the frequency doubling of the primary frequency that is inherently associated with an input optical signal. Within the complex and intricate interaction of this phenomenon, two photons that share the same frequency efficiently combine their energies in a highly effective manner, leading to the generation of a single new photon that possesses an energy, which is precisely double that of each of the contributing input photons. This serves as a clear illustration of a second-order effect at work in practical applications. The occurrence of the SHG process tends to take place significantly when the interaction length of the medium becomes comparable to the effective chi nonlinearity, thereby marking a vital and critical threshold for the phenomenon to manifest prominently and distinctly in a measurable way. An essential requirement necessary for achieving highly efficient second harmonic generation is the fulfillment of a condition known as phase-matching. The term phase-matching fundamentally refers to the delicate and intricate synchronization of the wave vectors that pertain to both the input waves and the corresponding output waves. To put it in simpler and more accessible terms, the photons that are traversing through the medium at different frequencies and wavelengths must align in such a manner that they propagate through the medium at effectively the same speed throughout the entire interaction region. To successfully attain this crucial and vital phase-matching condition, numerous approaches can be employed and utilized, including but not limited to the angular condition, temperature tuning, birefringence phase matching, and quasi-phase matching. These are just a few notable and widely employed techniques that are often implemented in practical applications across various fields. To provide a clear and visual representation of what phase-matching entails, a detailed phase-matching diagram is typically employed to illustrate and clarify the specific phase-matching conditions that are required for successful SHG to take place effectively. Furthermore, certain types of materials are regarded as particularly suitable and advantageous for enabling efficient second harmonic generation based on several necessary and critical factors that must be fulfilled within the

context of their application: these factors include the presence of a substantial second-order nonlinearity, alongside a broad transmission bandwidth that accommodates both the input signal and the generated SHG rays. Additionally, significant permissible birefringence is also essential as it further facilitates and enhances the nonlinearity necessary for the effective generation of harmonics. Considering these essential characteristics and factors, birefringent type II crystals and uniaxial crystals emerge prominently as particularly ideal candidates, as they encompass all the requisite prerequisites for effective SHG processes to occur efficiently. Consequently, the SHG processes that utilize these specific types of crystals tend to be notably simpler and are commonly applied in practical applications where efficient frequency doubling is desired and sought after for various technological innovations. Regarding the critical and discerning selection of suitable materials for SHG applications, a diverse range of materials can be effectively employed for this specific purpose in the ever-evolving field of nonlinear optics. The relative efficiencies of these carefully selected materials are also delineated and summarized in the following table, which serves to offer a concise yet comprehensive comparison of their performance in the context of SHG processes. This thorough comparison facilitates a clearer understanding of their capabilities and efficiencies when generating second harmonics from fundamental frequencies, making the selection process significantly more informative and insightful. In addition, it not only aids in the selection process but also enhances the overall comprehension of the complex interplay between various materials and their unique nonlinear optical properties, thereby enabling advancements and improvements in the fascinating and dynamic field of nonlinear optics [31, 32, 33, 34, 35, 36, 37, 38].

Nonlinear materials that are well-suited for second harmonic generation (SHG) comprise a remarkably diverse and extensive array of types, each exhibiting a rich tapestry of inherently unique properties, which collectively contribute significantly to their specific applications in an impressive number of scientific, engineering, and industrial fields. Among these materials, several notable examples stand out with particular prominence, one of which is AgGaS₂. This fascinating material is widely recognized for its distinctive relative efficiency ranking within the expansive and continually evolving field of nonlinear optics, which has become crucial in pushing the boundaries of what is possible in the realm of optical technologies. AgGaS₂ has garnered significant attention due to its exceptional efficiency characteristics, capabilities, and unique quasi physical properties, making it a cornerstone of research and application in the domain of nonlinear optics. Its widespread application in various researchers' experimental setups solidifies its status,

firmly establishing it as a critical component in numerous technological advancements and groundbreaking projects that demand high-performance characteristics, specifically tailored to meet the rigorous standards of modern optics. Furthermore, other materials such as KTP are also remarkably noteworthy; KTP boasts an efficiency score of 0.15, which serves as a critical and prominent indicator of its overall functionality in SHG applications. This efficiency score demonstrates that KTP possesses practical utility and functional advantages, although it does so with a more modest performance when compared to the leading and most efficient materials that dominate this advanced and intricate field. On the other hand, BBO stands out distinctly within the competitive landscape; it possesses a substantial relative efficiency score of 6, positioning it as a highly preferred and popular choice across numerous experimental settings and endeavors. Its high performance in generating second harmonics makes BBO especially valued for a plethora of applications that require exceptional efficiency, precision, and reliability in their optical properties to achieve the precise desired outcomes and high-quality results. In stark contrast to these high-efficiency materials, there are other examples such as ASN, ASD, TB1, and NB9, which consistently showcase a relative efficiency of 1 each. This level of performance suggests a relatively more standard and average capability when assessed against their high-performing counterparts. While these materials may not lead the efficiency rankings, they still hold considerable value and relevance in specific niche applications spread across various experimental contexts and scenarios, offering unique advantages that may be particularly suitable for certain types of research projects and industry demands that are less conventional and more specialized. An effective and reliable method for successfully detecting the emergence of Second Harmonic Generation (SHG) within an experimental arrangement is articulated through a sophisticated and meticulously developed technique known as autocorrelation. This method has been established and widely utilized within the scientific community over many years, involving the meticulous and careful recording of both the fundamental wave and what is conventionally referred to as the doubled frequency generated during the intricate SHG process. This sophisticated and intricate procedure is accomplished through the deployment of a high-resolution camera, which captures the necessary experimental data with impressive precision and clarity. Such detailed and systematic data collection ensures that the information gathered is not only accurate but profoundly useful for subsequent analyses in a variety of applications within the field. This well-organized and comprehensive process plays a critical role in assisting researchers and scientists as they diligently observe, measure, and analyze the pertinent figures

and data required not only for the angular condition but also for the temperature tuning techniques that are frequently employed in their experimental setups. In the specific context of SHG studies, both a pulsed laser, recognized for emitting light in brief and intense bursts, and various types of continuous lasers, which generate a steady and uninterrupted beam of light, can be effectively employed to achieve the desired outcomes and results in a diverse range of experiments of multiple kinds. This dual capability permits thorough exploration, meticulous investigation, and careful analysis of the complex and fascinating phenomena associated with SHG. Such versatility in laser application not only facilitates an enhanced examination of the diverse effects, methods, and intricacies related to SHG but also significantly augments the comprehensive understanding of this intriguing and multifaceted optical process. This growing understanding branches out into its myriad applications across a wide variety of fields of research and technological innovation, seamlessly paving the way for future discoveries, advancements, and potential innovations in the dynamic and ever-evolving field of nonlinear optics and beyond. The continuous exploration of this captivating area showcases the endless possibilities that lie ahead for researchers, technologists, and scientists as they endeavor to unravel the mysteries of light and its intricate interactions, ultimately expanding our knowledge and enhancing practical applications in our everyday life and various industries [39, 40, 41, 42, 43, 44, 45, 46, 47].

Chapter - 3

Nonlinear Optical Materials

Nonlinear optical materials are uniquely distinguished as those exceptional substances that possess the remarkable ability to display significant non-linear polarization phenomena when they interact with high-intensity light sources. This stimulating catalog of freshly identified materials, each showcasing illustrious non-linear optical properties, is steadily growing and diversifying in a truly extraordinary manner that reflects the current advancements in the field. The captivating nonlinear optical behaviors inherent in these materials find extensive and essential applications that span a wide range of practical uses, especially within vital frameworks like laser sources, optical amplifiers, electronic switchers, and a comprehensive array of other sophisticated photonic devices which are increasingly prevalent in modern technology. The essential characteristics of these nonlinear optical materials can be intricately correlated with the existence of non-uniform charge distribution within their molecular frameworks. Moreover, it is particularly noteworthy that these polarization moments have the potential to be time-varying in nature. Consequently, this property empowers the material to function as a modifiable or adjustable value medium, thereby enabling the exploration of unprecedented and various applications across multifaceted domains of science and technology that were previously unattainable. Generally, the prevalent nonlinear optical materials can be conveniently classified into three major categories: organic materials, inorganic materials, and hybrid materials. Organic materials frequently consist of a variety of organic molecules, typically characterized by an intricate web of sp^2 hybridized carbon atoms, which are crucial in contributing to their distinctive optical properties. Conversely, inorganic substances generally comprise crystalline formations and tend to be transparent while exhibiting remarkable qualities relevant to their unique optical functionalities and applications in photonics. The fundamental criteria necessary for acquiring exceptional non-linear optical properties in these materials are heavily tied to the essentiality of transparency, and thus transparency emerges as a pivotal parameter for discerning viable candidates among nonlinear materials. Hence, irrespective of the precise application under consideration, the primary material selection can

predominantly be influenced by their unique characteristics and behaviors in relation to transparency. At wavelengths that fall short of the transparency threshold, these materials become susceptible to potential damage, which can adversely affect their performance and longevity in practical applications. To qualify as a competent nonlinear optical material, a minimum damage threshold must be defined and maintained; it is vital for the material to preserve its non-linear coefficient across a wide range of laser powers, ensuring durability and dependability in various operational scenarios and settings. The progress of material development, particularly concerning organic types, can be performed with relative ease, attributed to the presence of a plethora of molecular configurations and symmetries existing at the microscopic level that can be skillfully modified and tailored to enhance their performance dramatically. The outstanding nonlinear optical characteristics observed can primarily be credited to the prevalent dipolar contributions. Particularly, those derived from molecular alignment, which can effectively be achieved through the application of a strong electric field pulse, subsequently resulting in the organized arrangement of the molecules into a well-structured crystalline formation. Meanwhile, it is crucial to recognize that considerably fewer inquiries have been directed towards inorganic and hybrid materials when juxtaposed with their organic counterparts, leading to significant gaps in understanding their full potential and capabilities. Consequently, there has been a focused exploration into the considerable potential offered by inorganic and hybrid materials, particularly in comparison to the traditionally acknowledged organic nonlinear optical substances, which experienced extensive research and development. Recent breakthroughs have been documented concerning the fields of organic, inorganic, and hybrid nonlinear optical materials, emphasizing the importance of further investigations. It is significant to note that numerous optical properties have been diligently measured and assessed based on the structural identities of various compounds examined, reinforcing the relationship between structure and optical functionality. It has been noted that any variation in structural characteristics will invariably yield consequential alterations in the optical performance abilities of the material, thus necessitating deliberate design and engineering practices. Recent scholarly investigations have concentrated explicitly on clarifying the intricate connection between the structural configuration of materials and their respective optical performance methodologies. In the design and development of new types of optical materials, the meticulous engineering of these materials to accurately align with the intended application category is fundamentally significant for their success. This design approach predominantly relies on complementary

characterization techniques specifically employed to thoroughly investigate the crystalline architecture of these materials, an essential factor for optimizing their functionality and efficiency in actual applications. The breadth of applications that may emerge from this line of inquiry is primarily dependent on the detailed analysis of the third-order nonlinear qualities exhibited by the crystal lattice, which bears considerable implications for innovations in the realm of laser optics and numerous other advanced technologies. Moreover, advancements in various forms of molecular engineering alongside modifications applied at diverse structural dimensions have been convincingly articulated and showcased in the latest scientific literature, illustrating the dynamic evolution and immense potential of nonlinear optical materials in contemporary and future applications [48, 49, 50, 51, 22, 52, 53, 54, 55, 8].

3.1 Nonlinear Crystals

A considerable and significant number of nonlinear optical phenomena have been meticulously observed and studied extensively with great success, highlighting the extraordinary breadth of this fascinating area of research. This remarkable achievement is largely attributable to the impressive advancements in the development and improvement of sophisticated nonlinear crystal devices. These highly sophisticated devices enable the attainment of remarkably high-efficiency second-order nonlinearity through the employment of various innovative techniques, which can be systematically categorized as either spontaneous or quasi-static in nature. Specially designed crystal structures that exhibit broken inversion symmetry are particularly effective and advantageous for demonstrating relatively high levels of second-order nonlinearity, providing fertile ground for further exploration. A prime and highly regarded example of such innovative and specialized structures is periodically poled lithium niobate, which has not only gained considerable prominence and recognition in this specialized field but also has driven extensive further research and applications in related areas that depend on its unique properties. Furthermore, bulk nonlinear crystals characterized by point group symmetries that disrupt inversion symmetry serve as exceptionally suitable candidates for showcasing impressive and remarkable second-order nonlinearity. Among these various options, KTiOPO_4 distinctly stands out as the most common and widely recognized prototype of nonlinear crystals that fulfill these crucial criteria effectively and efficiently. The performance metrics and operational efficiencies of these nonlinear materials can be significantly enhanced through a careful and deliberate optimization of various essential parameters. These parameters include crystal orientation and

temperature, specifically designed to ensure they work effectively in accordance with their phase-matching or quasi-phase-matching configurations, which is critical for achieving desired outcomes in practical applications. This meticulous and thorough optimization opens up a wealth of opportunities for realizing a diverse array of functions that encompass fundamental processes such as second-harmonic generation, sum frequency generation, difference frequency generation, optical parametric oscillation, optical parametric amplification, and numerous ultrafast nonlinear optical effects, all of which are integral to advanced applications in the ever-evolving domain of optics. Given their inherently highly nonlinear characteristics and properties, there has been an extensive and diverse array of both commercially available and research-grade nonlinear crystals that have grown increasingly popular in the rapidly evolving fields of nonlinear optics and ultrafast laser technology. Consequently, a significant volume of scholarly work and dedicated research efforts has been fervently committed to the discovery and comprehensive development of novel, high-quality nonlinear single crystals aimed at facilitating various fundamental research endeavors and a broad spectrum of practical applications, which are crucial for advancing this field. Nevertheless, challenges persist within the domain; specifically, the relatively small dimensions and intricate poling period of these crystals severely limit their overall capacity for efficiently handling high energy and power levels. This significant limitation inhibits broader, more ambitious applications in high power and high-intensity laser systems, which is a concern that researchers continue to diligently address through innovative approaches. To effectively tackle these challenges, several innovative processes and methodologies have been comprehensively explored, ensuring that the nonlinear optical crystals produced possess the desired ordinary and/or extraordinary optical properties, as well as the critical mechanical and thermal parameters necessary for their intended applications. Furthermore, special attention has been devoted to the spatial and temporal distribution of linear and nonlinear optical properties within the resulting crystals, a critical factor that can significantly influence their overall performance and functionality in practical settings. Notably, the incorporation of advanced glass ceramics has emerged as an established and highly effective approach, facilitating substantial and meaningful alterations in the fundamental parameters of the crystals that compose them. As a result, the application of glass ceramics not only fosters the use of the resulting composite structures as volume-effective active elements in specialized lasers and tunable radiation sources, but also aids significantly in the development of innovative radiation sources and advanced acousto-optical devices, enriching the landscape of available

technologies. Alongside these promising advancements, various analytical methods have been developed to thoroughly describe, quantify, and characterize the nonlinear optical properties exhibited by these materials, further enhancing our understanding of their capabilities and limitations. This ongoing evolution enriches our comprehension of potential applications in a rapidly advancing technological landscape, paving the way for exciting new developments and breakthroughs in the field of nonlinear optics and beyond, transforming the way we harness light for innovative purposes [56, 57, 58, 59, 13, 60, 61, 62].

Chapter - 4

Introduction to Laser-Matter Interaction

Lasers, which stand for "light amplification by stimulated emission of radiation" or, in more elaborate terms, can also be referred to as "optical amplification by stimulated emission of radiation," represent, without a doubt, some of the most sophisticated and highly advanced devices currently available in the ever-evolving realm of contemporary technology. They are marked by their impressive and unmatched ability to produce highly collimated and remarkably coherent forms of electromagnetic radiation. Depending on their precise operational wavelengths, lasers can be systematically classified into an extensive and varied array of types, which encompass several significant categories that include, but are certainly not limited to, solid-state lasers, gas lasers, semiconductor lasers, and dye lasers, among numerous others. In addition to these commonly referenced categories, with which many individuals might be somewhat familiar, there exists a multitude of additional classifications that arise from the specific gain medium they utilize. For instance, categories such as crystal lasers, glass lasers, liquid lasers, and soft matter lasers significantly enrich the expansive range of applications and functionalities that are accessible and applicable across various scientific, industrial, and medical fields. These varied types of lasers demonstrate remarkable versatility when employed in numerous contexts, showcasing the profound impact lasers have in driving and facilitating technological progress in different spheres. Furthermore, by contemplating the intricate underlying principles of the various pumping mechanisms utilized in lasers, in harmonious combination with the excitation sources necessary for their effective operation, we can further categorize lasers into two primary types: continuous-wave lasers and pulsed lasers. Among these significant groups, pulsed lasers exhibit an exciting and diverse array of designs and configurations—including intricate pulsed structures characterized by pulse durations that can oscillate from nanoseconds to femtoseconds. These configurations have garnered considerable attention and heightened interest due to the substantial advancements recently achieved within the dynamic field of laser technology and its broad array of practical applications spanning a multitude of domains and practices. This rapidly evolving field of laser

technology, distinguished by increasingly higher power outputs alongside enhanced capabilities for producing ultra-short pulse durations, unveils extraordinary opportunities for researchers and scientists alike to explore and understand the fundamental and intricate interactions that transpire between the unique emissions of lasers and the myriad forms of matter that can be scrutinized and studied using these powerful technological tools. As we delve deeper into the complex dynamics of laser-matter interactions, it becomes abundantly clear that the behavior of light can manifest in various forms, such as scattering, absorption, reflection, or even transmission by any sampled material or substance. This rich and diverse behavior empowers researchers to gather a plethora of invaluable insights and detailed information concerning the specific materials and substances targeted in their meticulous investigations. Moreover, laser beams—operating at exceptionally high intensities within the near-infrared, visible, or even within short-wavelength spectral ranges—when suitable filters are expertly chosen for specific applications, are recognized as extraordinarily powerful instruments that are extensively utilized in a variety of industrial applications, crucial scientific research endeavors, and numerous technological advancements that significantly enhance our understanding of the universe and everything within it—revealing the endless potential and versatility of laser systems. By harnessing the robust capabilities of high-intensity light, paired with the innovative use of ultrashort pulse durations, fascinating phenomena categorized as "nonlinear" interactions gain prominence. This prominence arises primarily due to the rapid dynamics that intricately engage with the population ranges of electrons alongside the nonlinear excitation processes that markedly impact the unique and varied behavior of the targeted material. Within this intricate framework, the concept of "nonlinearity" can be thoroughly elaborated and solidly understood through its nuanced relationship with the intensity of the incoming light field, as well as its subsequently observable effects on the intrinsic material properties being examined with precision. Importantly, the emerging and complex processes associated with the vibrant field of nonlinear optics depend intricately on the intensity of the incident radiation while also being closely connected to the specific characteristics and particulars of the pulse duration currently applied in various experimental setups. The exponential growth in the domain of nonlinear optics, in conjunction with groundbreaking innovations in ultrafast imaging techniques, advanced materials processing, and complex visualization methodologies, has become increasingly feasible and attainable. Such remarkable progress is largely attributable to substantial advancements in the design and architectural refinements of pulsed laser technologies that

greatly facilitate and bolster these essential scientific endeavors. The fast and dynamic processes at play within this intricate realm of investigation are meticulously examined through the lens of excitation-induced alterations observable within the physical properties of a vast variety of materials. These investigations also delve deeply into the novel phenomena that either arise from or significantly involve complex interactions among atoms, molecules, nanostructures, and a diverse array of solid-state materials that are under close study and scrutiny within the scientific community. Consequently, the ongoing integration of innovative laser technology with cutting-edge advancements in material science continually paves the way for astonishing discoveries and transformative advancements across multiple scientific and technological domains. This showcases the immense potential and profound impact that this remarkable field of study can have on our understanding, manipulation, and overall comprehensive comprehension of the intricate world that surrounds us. It opens up new avenues for fresh innovation and exploration that were previously deemed unimaginable or far beyond our immediate reach, emphasizing the critical importance of ongoing research and development in this remarkable area of inquiry, which continues to immensely shape our interaction with the world at large, presenting novel challenges and unexplored opportunities in the exciting journey of scientific advancement [63, 64, 65, 66, 67, 68].

4.1 Absorption and Emission Processes

The absorption of a photon by an atom or a molecule signifies far more than merely a reaction; it encapsulates an immensely significant and intricately complex process that plays an essential role in the elaborate mechanisms involved in the vital transfer of energy from a lower excitation state to a considerably higher excitation state. This crucial energy transfer is not only fundamental but also arises as a primary prerequisite for a vast array of applications that exist within both the realms of physics and chemistry, as it constitutes the underlying basis for numerous fascinating phenomena observable in the natural world surrounding us. Moreover, this vital energy transfer is also pivotal in many technological innovations that intricately shape our daily experiences in multifaceted ways. Such innovations have a profoundly significant effect on everything ranging from the efficient lighting of our homes to providing the essential power needed for numerous devices that we often take for granted, seamlessly permeating our routines and influencing our interactions with various forms of technology. For this vital energy transfer process to occur with optimal efficiency, it becomes paramount that the energy of the incoming photon precisely matches the

measurable difference between the two distinct energy levels that are integrally involved in facilitating the process. This fundamental relationship can be mathematically articulated with the expression $h\nu = E_{1,2} - E_{0,2}$, where $h\nu$ signifies the energy of the photon, a concept deeply embedded in the profound principles of quantum mechanics. The terms $E_{i,n}$, where the indices i and n represent different energy levels taking on integral values such as 0, 1, 2, and beyond, denote the various excitation energies that are uniquely linked to the specific states of the atom or molecule engaged in this dynamic and intricate interaction of energy and matter. This captivating and multi-layered process can transpire in either a highly selective manner, where specific energy levels are purposefully targeted and chosen for transition, thus facilitating highly efficient excitation, or in a broader, more nonselective manner that allows for an extensive array of interactions to take place. This indicates that the fundamental nature of the light—its frequency, wavelength, and energy—coupled with the way it interacts or aligns with the atom or molecule, significantly influences the overall processes of excitation and emission. After the successful transition of an atom or molecule to the higher excitation state, it will, in due course, revert back to a lower excitation state through one or multiple nonradiative transitions, which are remarkably significant phenomena deserving notable attention and study. These nonradiative transitions occur over extraordinarily brief time spans, typically ranging from femtoseconds to picoseconds, presenting an intriguing glimpse into the nuanced behaviors of atomic and molecular systems while illuminating the complex interactions occurring at the quantum level. During these rapid and transient transitions, the excited atoms or molecules, which were existing in that highly energetic state, will emit photons as they gracefully transition back down to the lower energy states they originally occupied prior to reaching the state of excitation. In scenarios where the emission occurs subsequent to the absorption of a photon and involves transitions from state n back down to state 0, this specific category of emission is referred to as spontaneous emission. Spontaneous emission is uniquely characterized by its inherent nature of not requiring any additional light to activate or initiate the release of a photon into the surrounding environment. Conversely, if the atom happens to be elevated in a highly excited state n at the precise moment when the next photon absorption occurs, it creates significant opportunities for the emission of photons to be influenced or stimulated by any arriving light shortly after, providing a clear pathway for amplified reactions to take place, which can lead to a cascade of photonic activity. Both the various absorption processes that lead to the initial excitation and the corresponding spontaneous or stimulated emission processes play

crucial roles in significantly elevating the concentration of atoms or molecules that ultimately reside in the final lower state of excitation. From this build-up of atoms in the lowest energy state, pure light generation can occur, particularly through the stimulated emission process. This sophisticated process stands as a central mechanism for the efficient generation of coherent light, which is paramount in numerous scientific and industrial applications. The intricate equilibrium and complex interactions among these three foundational mechanisms—absorption, emission, and stimulated emission—establish a fundamental backbone for the sophisticated phenomena behind laser or amplified light generation, showcasing the remarkable elegance and precision inherent in nature and the longstanding laws that govern it. Furthermore, the behaviors and distinct characteristics of the resulting laser or amplified light are profoundly shaped and influenced by various parameters such as temperature, injection techniques, and pumping conditions, all of which can experience considerable fluctuations and variations that alter their states. This complex interplay of intricate variables makes an in-depth and comprehensive understanding of these processes absolutely essential for the broader study of laser technology and optics, which are indispensable fields in the modern landscape of scientific inquiry and technological advancement. Their impacts span a diverse range of applications, reaching from telecommunications to intricate medical devices, and extending further into countless other domains and innovations that underpin much of contemporary life. These factors emphasize their significance in modern society, reinforcing their relevance in a continually evolving technological landscape that continually necessitates innovative breakthroughs and advancements in multiple fields of study and industry ^[69, 70, 71, 72, 73, 74, 75, 76].

Chapter - 5

Theoretical Models in Nonlinear Optics

Nonlinear optics is an exceptionally fascinating and dynamic field that is fundamentally concerned with the intricate and multifaceted interactions that occur between intense light fields and various forms of matter. The primary purpose of nonlinear optics is not only to describe these phenomena but also to conduct a thorough and comprehensive analysis of the significant impact that the nonlinear characteristics of different media exert on the inherent properties and behaviors of the light wave itself. A common and widely adopted approach utilized within the expansive and continually evolving realm of nonlinear optics involves the use of a carefully constructed theoretical model specifically tailored for the light wave. This model is fundamentally based on a comprehensive collection of nonlinear wave equations, which have been meticulously derived and developed through rigorous and intensive theoretical work. Consequently, the nonlinear wave equations arise from the foundational principles that underlie energy conservation and momentum conservation equations. This leads to the realization that the complex optical field can indeed be elegantly formulated and derived from Maxwell's equations as well. A particularly notable equation that is frequently employed in the study and exploration of nonlinear media is the nonlinear Schrödinger equation, commonly referred to simply as the NLSE. The NLSE is extensively utilized to effectively describe the propagation of wave packets as they traverse through strongly nonlinear media. This particular aspect is especially relevant in specific scenarios where the influence of absorption can be deemed negligible or minimal for practical considerations. Furthermore, in situations where the impact of dispersion can also be considered insignificant or minor, the standard formulation of the NLSE may be thoughtfully simplified into what is known as the envelope form. This envelope form represents the most common representation of the NLSE, which is widely employed in a variety of weakly and moderately nonlinear regimes, ultimately facilitating a clearer understanding of the intricate and complex dynamics involved in the propagation process. An immensely significant outcome arising from the theoretical framework of nonlinear optics is that, under suitable conditions and due to the intrinsic nonlinearities present within the medium, the input wave

packet is capable of undergoing a fascinating and elaborate rearrangement. This intricate and complex rearrangement process allows for the generation of a monochromatic stationary wave specifically within the context of the replica-symmetric (RS) phase, highlighting the intricate behaviors and phenomena that can arise in such systems. While this theoretical treatment provides us with a broad and comprehensive perspective on the anticipated behavior of a given system, the actual experimental verification of these insightful predictions within the expansive and multifaceted field of nonlinear optics can, at times, present considerable and formidable challenges. These challenges frequently arise when researchers attempt to create physical realizations of the described intricate structures, where both the transverse and longitudinal properties of the light field have been meticulously studied and analyzed in considerable detail. Recently, in a series of innovative theoretical studies, researchers have suggested that considering the transverse and longitudinal nonlocal influences could potentially introduce an additional scattering effect into the complex phase diagram under consideration. This effect may indeed compete with the behaviors associated with the established replica-symmetric (RS) phase point stable behavior, thus providing exciting avenues for deeper insights and understanding into the dynamics of the system. The theoretical framework in nonlinear optics serves as an invaluable and powerful tool for predicting the specific types of fluctuations anticipated, as well as over what extensive range of system parameters these fluctuations will arise. In many instances, this honored and respected framework can offer statistical predictions regarding the probability that a particular system will manifest in a specific state of interest. Currently, analytical tools in this sophisticated and advanced domain are being developed further for enhanced precision, accuracy, and reliability. For more intricate and complex systems, it is often much more pragmatic and efficient to engage in numerical methods, which can more effectively manage and address the complexities involved in such fascinating nonlinear phenomena and intricate interactions, paving the way for groundbreaking discoveries and new advancements in the field [77, 78, 79, 25, 80, 81, 82, 83, 18].

5.1 Nonlinear Schrödinger Equation

The nonlinear Schrödinger equation (NLS) is widely recognized and esteemed as one of the most fundamental and critical examples of an equation that supports solitary wave solutions, which are of immense interest and hold critical importance in the expansive realms of both mathematics and physics. These solitary wave solutions embody a distinctive, coherent type of wave that can traverse expansive areas of space without undergoing any significant

alterations to their shape or structure as time progresses. This remarkable characteristic allows these waves to preserve their form entirely until they eventually meet a certain boundary or obstruction in their path, thereby affecting their continuity and stability in a notably significant fashion. Due to this fascinating and captivating property, the derivation, as well as the analysis and subsequent in-depth study of these equations, have blossomed into pivotal focal points within significant areas of research that pertain to nonlinear waves and their extensive and diverse applications. The phenomena surrounding solitary waves, along with the comprehensive and intricate descriptions of nonlinear propagation unfolding in directions perpendicular to the principal movement of the waves, hold paramount importance for the thriving and rapidly expanding field of nonlinear optics—a discipline that has relatively quickly developed into one of the most critical and vibrant sectors of research within the broader field of optical sciences. This encompasses an array of intriguing phenomena and complex interactions that arise when solitary waves interact with various mediums. Nonlinear optics primarily addresses the intriguing and intricate interactions that transpire between very intense laser beams and various forms of matter, which are often described and modeled with precision and care by employing a well-defined wave equation. In this critical context, the conventional or standard case is represented most effectively by the linear Helmholtz equation, which is well-known for its straightforward and relatively uncomplicated nature that simplifies the analysis of wave behavior in diverse situations. However, when it comes to the highly intense laser beams so commonly encountered in modern applications, the influence that matter has on the wave cannot be accurately or sufficiently described merely by a term that is directly proportional to the intensity of the wave itself, especially since many complexities and nuances arise in these particular situations that challenge conventional understanding. This essential realization motivates and necessitates further exploration into the detailed and intricate study of the nonlinear wave equation, as researchers delve deeper into the complexities and fascinating phenomena that arise from such challenging conditions and specialized environments. Within this captivating realm of study, substantial emphasis is laid on the focused and thorough investigation of solitons that meet the stringent criteria established by the wave equation framework in a variety of contexts and applications. This deeper and more comprehensive exploration has significantly enriched our understanding across numerous areas, especially in advanced applications such as airborne communication, including satellite-based systems and wireless technologies, as well as non-dispersive signal transmission that has been made reliably effective through the innovative incorporation of optical

glass fibers. These fibers have, in fact, become absolutely critical in the development of contemporary communication systems, enabling rapid and reliable signal transmission over great distances. In the subsequent section, we will present a detailed and systematic derivation of the nonlinear Schrödinger equation, outlining the essential methodologies and stages involved in this significant and essential process of derivation. Before we engage in a more expansive and comprehensive explanation of the detailed steps involved in this derivation, it is crucial to mention that we will establish the boundary conditions of the wavefunction as being set to zero. This approach is in strict alignment with a standard procedure employed in mathematical physics, which is designed to circumvent any potential difficulties and complexities that could arise during the intricate integration process necessary to derive the equation accurately and comprehensively. It is also vital to acknowledge at this juncture that the experimental realization of these boundaries is virtually impossible in the framework of a finite system that is positioned within an infinite domain, as boundary effects become critically significant and influential in determining the behavior observed in real-world scenarios. Nevertheless, the behavior manifested in the regions where boundary influences can be perceived as negligible provides invaluable insights into the overall character and properties of the system being studied, enabling deeper explorations into the wave dynamics involved. Achieving full soliton propagation within a sufficiently vast domain is actively pursued by approaching the limit where L tends towards infinity ($L \rightarrow \infty$). This consideration not only aids in the thorough and meticulous analysis of the system but also effectively establishes the limits of the system size that can be methodically and thoroughly analyzed, as τ emerges as the maximal duration of the solution time applicable to the given conditions. It is at this critical point, at this specifically defined juncture, where the finiteness of the system begins to exert its influence and impact the behavior of the wavefunction and the spectral properties that it exhibits. This, in turn, leads us to a deeper understanding of the intricate interplay between solitons and the conditions governing their propagation. This understanding strongly underscores the compelling need for continued research in this vital realm of study within the context of modern physics and its advanced applications, which beckons future inquiries and in-depth investigations into the rich phenomena that lie within the nonlinear dynamics domain, inviting a host of possibilities for discovery and innovation in the years to come [84, 85, 86, 87, 88, 89, 90, 91, 92, 93].

Wave packet propagation can be thoroughly and effectively described and understood through the use of a robust, sophisticated, and comprehensive solution to the highly significant nonlinear Schrödinger equation. This pivotal

equation serves as an essential mathematical construct that governs the intricate, complex, and multifaceted dynamics of wave functions across a remarkably wide array of physical contexts, phenomena, and applications. It stands as one of the most crucial and highly regarded equations in modern physics, encapsulating an incredibly diverse variety of intriguing regimes and extraordinary phenomena that captivate the interest of researchers, scholars, and practitioners across various scientific disciplines alike. Among these phenomena is the captivating and remarkable occurrence of soliton propagation—that is, the stable, localized wave packets that astonishingly maintain their shape while traveling at constant speeds through a given medium without experiencing any form of dissipation, deformation, or significant change in their form. To accurately and efficiently determine the solutions to this complex equation, researchers, scientists, and mathematicians commonly rely on a range of advanced, sophisticated, and carefully designed numerical simulations. These simulations play a crucial and indispensable role in thoroughly analyzing, understanding, and ultimately unraveling the complexities of the diverse behaviors and intricate dynamics of wave propagation over time, further advancing the field of wave physics. With a multitude of methods and strategies available for solving the equation numerically with precision, one particularly notable and widely utilized technique is the split-step Fourier method. This powerful and effective method has been proven to be remarkably effective and efficient across a broad spectrum of applications, thus making it an essential tool in the evolving and rapidly advancing field of wave dynamics and propagation. The split-step Fourier method represents a highly versatile approach that is especially beneficial for a variety of applications in the crucial field of optical communications, wherein precise and accurate manipulation of wave packets is essential for the efficient, reliable, and effective transmission of ranging amounts of information over long distances, particularly in today's technologically driven society. In contrast to the finite difference time domain method, which demands a detailed, often cumbersome, and computationally expensive resolution of the changes in the dispersion profile over time, the split-step Fourier method greatly simplifies the essential computational processes involved. By adeptly transforming the nonlinear Schrödinger equation into a more manageable ordinary differential equation, this approach becomes significantly easier to resolve both analytically and numerically across various specific cases. As a direct consequence of these transformations, it does not require excessive computational resources or substantial processing power, thus enabling researchers to perform their analyses and computations more swiftly, effectively, and efficiently than

would otherwise be possible in many complex scenarios. Additionally, this method provides a simpler and less computationally intense approach to model wave packet propagation across a diverse array of different scenarios, frameworks, and mediums. Such admirable characteristics make it a preferred and popular choice among researchers, engineers, and professionals working diligently in the fascinating and evolving field of wave dynamics and propagation. By effectively leveraging this innovative and powerful method, one can gain much greater depth and clarity of insight into the underlying principles affecting a wide range of wave phenomena, accurately analyze sophisticated and intricate wave behaviors, and reliably predict the responses and interactions of diverse wave packets in various demanding contexts. This significant contribution to our overall understanding leads to groundbreaking advancements and breakthroughs in crucial fields such as photonics, quantum mechanics, and telecommunications, where the precise understanding and manipulation of subtle wave behaviors are central to the ongoing innovation and technological development that characterizes the modern era. Through the judicious application of these powerful numerical techniques, the possibilities for practical, real-world applications expand dramatically in unprecedented ways, thereby impacting numerous technological frontiers and enhancing our ability to interact meaningfully with the wave characteristics that surround and influence us in our daily lives. The integration of such methodologies into advanced research continues to elucidate the underlying principles of complex wave phenomena, thereby fostering continued exploration and discovery in the ever-evolving world of scientific inquiry, which remains as vital and dynamic as ever in the information age [94, 95, 96, 97, 98, 99, 100, 101, 102, 103].

Chapter - 6

Experimental Techniques in Nonlinear Optics

In numerous prominent research laboratories that are widely scattered throughout the globe, there exists an ongoing, intense, and immensely active pursuit of experimental quantitative determination of nonlinear optical responses that span across a diverse and wide-ranging array of materials and experimental settings, showcasing the profound depth and richness of this intricate scientific field, which captivates and challenges researchers alike. The landscape of this endeavor is characterized by a remarkable diversity in the materials being studied and an extensive variety of experimental conditions, all aimed at unraveling the complex behaviors exhibited by nonlinear optical phenomena. It is absolutely vital and crucial to possess complete, thorough, and detailed information regarding the various advanced experimental techniques that are employed in the careful and meticulous investigation of a multitude of complex physical phenomena that are seamlessly interwoven within the expansive and crucial field of material science. Having access to such information not only enhances the effectiveness of research efforts but also ensures that researchers are well-equipped to navigate the multifaceted challenges presented by these phenomena. This is especially true for those intricate phenomena that are intricately related to nonlinear optical behaviors, as well as the numerous innovative laser experiments that often accompany such investigations, which can lead to fascinating discoveries, innovative applications, and groundbreaking advancements in a multitude of sectors, profoundly influencing both fundamental science and applied technology. This chapter is specifically intended to provide a thorough, comprehensive, and well-structured description of the various sophisticated experimental setups and diverse methodologies that are commonly utilized for the extensive and in-depth study of nonlinear optical phenomena, which bear immense relevance to the landscape of modern scientific research and critical innovation. Amongst these diverse techniques are the prominent time-resolved pump-probe techniques that allow for unprecedented temporal resolution, providing invaluable insights into the dynamics of light-matter interactions; the highly valuable and efficient third harmonic generation that significantly enhances

signal detection and clarity in experimental observations; and the exquisite and finely-tuned second harmonic generation, all of which are just a few notable and widely-adopted examples in the field that contribute substantially to our detailed understanding of these complex phenomena and their implications. The setups and alignment procedures needed for each of these sophisticated techniques will be described in meticulous detail, paying careful attention to the specific experimental requirements, nuances, and idiosyncratic characteristics of the equipment used, as well as the fundamental importance of following proper protocols and established guidelines to ensure accurate, reproducible results that can stand up to rigorous scientific scrutiny. Furthermore, in direct relation to the experimental results that are obtained through these advanced methods, potential applications within various rapidly evolving technological domains—including communications, photonics, optical computing, and medical diagnostics—will also be highlighted thoroughly, explained comprehensively, and pointed out for the reader's thoughtful consideration and deeper understanding of their profound significance and utility in practical real-world scenarios. Moreover, the section that is dedicated to elaborating on the critical aspects of data processing, analysis, and interpretation constitutes a substantial and significant part of this work, where the treatment will strongly focus on effective modelization techniques, thorough statistical analysis, and the theoretical predictions that can be drawn from the results achieved through these rigorous experimental efforts to enhance understanding and drive further innovation. For many of the experimental techniques that necessitate special and specific alignment procedures, detailed and explicit alignment steps, along with pertinent safety warnings and precautions, are indicated clearly and unequivocally within the individual sections of the current chapter to ensure the safety and efficacy of the experimental investigations undertaken. For the various techniques that do not entail these strict alignment requirements, standard recommendations, best practices, and fundamental principles should always be meticulously adhered to and taken into account to ensure the maximum precision and reliability of the results, thus avoiding common pitfalls and mistakes that could compromise the integrity and validity of the research findings. It is abundantly clear that there are numerous difficulties, complexities, and inherent limitations that are fundamentally associated with both the underlying physical principles and the intricate material characteristics involved in these multifaceted experimental approaches that must be reckoned with. The primary aim of this part is to equip the experimentalist with a concise yet highly informative and useful overview of the various established methods that are routinely employed, while also indicating the practical challenges,

obstacles, and considerations that one must effectively cope with during the course of real-world experimental investigations in this dynamic and rapidly advancing domain, addressing both the theoretical and experimental facets of contemporary research, leading to enriched understanding and capability. Nowadays, many new and advanced facilities, along with cutting-edge technologies, have become increasingly available for researchers in this essential area of inquiry, presenting exciting opportunities for breakthroughs and advancements. These technological advancements include time-resolved THG in innovative commercial spectrometers that allow researchers to explore new territories, and groundbreaking transient absorption techniques specifically designed to operate efficiently within the UV realm on the ultrafast femtosecond timescale, reflecting the rapid pace of progress and innovation that continues to shape this vital and ever-developing field. As we look forward to the exciting and promising future of this fascinating field, it will undoubtedly prove to be both interesting, invaluable, and essential to remain well-informed and keep abreast of all the latest developments, discoveries, advancements, and innovations in techniques that are readily available for ongoing research in this vibrant and dynamic area of scientific study that continuously evolves and expands. The continual evolution of experimental methods, innovative tools, and novel insights will undoubtedly lead to new directions, groundbreaking insights, and transformative breakthroughs, reshaping our understanding of nonlinear optical phenomena and their wide-ranging implications across various scientific disciplines, thereby opening up entirely new avenues and possibilities for exploration and applications that could fundamentally redefine current methodologies, enhance the technological capabilities of the future, and significantly contribute to the advancement of science and technology as we know it [104, 105, 22, 21, 106, 107, 108, 42].

6.1 Z-scan Technique

The Z-scan technique prominently stands out as an extensively utilized and remarkably effective method for the thorough and comprehensive characterization of nonlinear optical materials, establishing itself as an invaluable tool within the expansive and ever-evolving field of photonics. This vibrant domain has witnessed tremendous growth and evolution over recent years, profoundly transforming the landscape of optical research and engineering in profound ways. The fundamental principle that solidly underpins this innovative approach involves meticulously measuring the variation of transmittance of a highly focused light beam as the thickness of a sample, which is positioned with exceptional precision within that focus, is

carefully traversed back and forth across a specific range of distances. Alternatively, this intricate process can also be understood as the systematic and organized scanning of the focus itself across the sample in an incremental manner, ensuring a comprehensive analysis of the material's nuanced response to various optical stimuli. In this complex and multidimensional context, transmittance is explicitly defined as the ratio of the total optical power transmitted through the sample to the incident optical power directed squarely at the nonlinear sample. It is critically important to emphasize that the sample must be aligned with unparalleled precision during these experiments, as the nonlinear effects that typically occur tend to be quite weak, subtle, and sensitive to any variabilities present in the experimental setup. This high precision in alignment is not merely a technical requirement; it becomes essential, ensuring that the experimental signals—which need to be accurately registered—are significantly amplified, enhancing their visibility, clarity, and overall significance. Such amplification serves not only to magnify these signals but also to bolster the accuracy and reliability of all subsequent measurements and analyses, significantly enriching the comprehensive understanding of the material's optical properties and interactive behaviors. Furthermore, an experimental Z-scan curve can be influenced by a plethora of complexities and issues, as well as by factors that are deeply intrinsic to the specific experimental setup being utilized. To begin with, it is imperative to meticulously determine the experimental transmittance curve with the utmost accuracy and precision. In practical terms, to carry out the nuanced and often complex measurements of the nonlinear parameters of focus using the Z-scan technique effectively, the position of the focal plane along the z-axis needs to be deliberately controlled and skillfully adjusted to gather high-resolution data that can reveal intricate details of material interactions and processes. This method, which focuses specifically on analyzing transmittance as a function of aperture size, operates efficiently within the geometrical approximation framework, ensuring that the results derived from the experimentation process are both reliable and reproducible—requirements that are crucial for achieving meaningful advancements within our optical knowledge bases. The resulting curve obtained from these precise measurements can be phenomenally valuable for a multitude of reasons, with its detailed analysis unlocking deeper insights into the underlying physics at play within the tested material. It provides crucial insights concerning transmittance at the focus and thus allows for meticulous calculations of the f-number, the coefficient of defocus, and detailed quantifications of several degrees of defocus or other relevant metrics that pertain to the optical characteristics of various materials. These quantifications can significantly enhance our comprehension of optical

behaviors and interactions, which are critically important in many applications throughout the vast domain of photonics. Additionally, we could gather even further insightful information regarding the sample itself and its intricate interaction with the focused light, encapsulating important parameters such as the conicity factor, which can yield fruitful and meaningful data about the material's intrinsic properties and its dynamic behaviors in response to different experimental stimuli or varying conditions. The analysis of this meticulously collected data will be diligently geared towards achieving the best possible fit of the experimental results to theoretical expectations, while simultaneously aiming to compare these results with an established calibration method that can effectively serve as a credible benchmark for evaluating the accuracy and consistency of all significant findings reported. Consequently, assuring the integrity of all reported data becomes paramount. This calibration process could potentially encompass a variety of fitting approaches, each uniquely tailored to the specific characteristics of the experiment and the nature of the materials that are being thoroughly investigated. These fitting methods may include a linear fit, comprehensive Boltzmann functions, advanced sigmoid models, and even the more complex Beuers 4-dependent functions, among other sophisticated methodologies. Notably, this methodical and computational decision-making process plays an indispensable role in helping to validate a Gaussian approximation concerning the transmittance at the focus, which is vital for accurate data interpretation and subsequent applications that span various technological landscapes. Furthermore, issues and challenges encountered in practical applications of the Z-scan technique are discussed in meticulous detail. This encompassing discussion covers crucial and essential steps such as system calibration, adept noise reduction setups, careful configuration of camera drivers, and thorough sample preparation—each step contributing significantly to the integrity and reliability of the entire experimentation process. The Z-scan-based technique is broadly recognized as a robust and versatile tool for the nonlinear characterization of a wide array of materials. It extends its applications far beyond mere measurements. Its versatile applications are not limited to this; rather, they encompass measuring the complete spectrum of third-order nonlinear optical phenomena, which play an essential role in the modern optics field, encompassing a variety of scenarios and technologies that extend to telecommunications as well as advanced imaging techniques. Moreover, the Z-scan technique is particularly well-suited for assessing advanced materials that demonstrate noteworthy self-focusing properties. These properties have critical implications in various applications involving phase-conjugating mirrors and saturable absorption materials, such as the intriguing and

sophisticated InGaAs quantum wells, which stand at the very forefront of technological innovation in the expansive field of photonics. Through this method, researchers can significantly enhance and deepen their understanding of these materials, thereby stimulating the development of innovative optical materials and devices. This effectively helps push the boundaries of current technology in this exciting and rapidly evolving field of science and engineering, while simultaneously offering pathways toward new discoveries and innovative applications. As researchers delve deeper into these advanced materials, they can uncover novel applications and phenomena, opening up dynamic avenues for further exploration and remarkable technological advancements that ultimately benefit society as a whole. Thus, the Z-scan technique remains a cornerstone in the relentless quest for knowledge within the ever-evolving realm of nonlinear optics, continuously paving the way for future breakthroughs and discoveries that will indelibly shape the next generation of photonic technologies ^[109, 110, 111, 112, 113, 114, 115, 116].

Chapter - 7

Nonlinear Optics in Photonic Devices

Nonlinear optics is currently playing an increasingly critical and pivotal role in enabling the emergence of entirely new generations of highly advanced photonic devices that are making significant waves across a multitude of diverse fields and industries. By effectively avoiding the fundamental trade-off that is typically associated with speed versus energy consumption found in traditional electronic circuits, it becomes possible to break the conventional cost-complexity trade-off often observed in conventional systems, thus opening new avenues for innovation. This groundbreaking breakthrough is paving the way for the extensive and rapid development of photonic devices that are suitable for a broad and diverse range of applications, which could significantly transform industries and technologies alike into new paradigms of efficiency and efficacy. Already, there have been numerous exciting and promising applications of nonlinear optics manifesting in both theoretical concepts and in experimental demonstrations of laser-based devices that are specifically engineered for signal processing, advanced sensing applications, and high-resolution imaging techniques. These fascinating nonlinear optical phenomena being explored hold immense potential to provide a solid foundation upon which to devise cutting-edge optical switches or even enable the amplification of optical signals directly through the intelligent and strategic use of powerful laser beams. Moreover, the tunability in wavelength that is offered by advanced laser sources can serve as an exceptionally valuable and highly effective tool for facilitating wavelength conversion with precision, while also significantly enhancing the sensitivity of various detection techniques that are commonly employed in practical, real-world scenarios and applications. This extensive spectrum of diverse applications magnificently showcases the intricate and often complex interplay between optical fields and the devices that rely on them for their operation. It demands a comprehensive and in-depth understanding of the principles of modern nanophotonics, alongside the innovative materials that are involved in the intricate processes at play. Recent research trends indicate that the concept of nonlinearity is not just an abstract idea; it is intrinsically and directly coupled to the physical dimensions and specific configurations of the devices

themselves that are being developed. The advances achieved in nanophotonic technologies will, in many ways, be profoundly shaped by the fundamental characteristics and unique properties of nonlinearity itself. The availability of nonlinearity within a given material system is a material-dependent quantity that can vary significantly across different contexts and applications, which adds to its inherent complexity. However, several effective and innovative methods and strategies can be employed through which the amount of nonlinearity can be judiciously optimized specifically for targeted applications, thereby enabling improvements in performance and functionality in innovative ways. The increasing maturity in advanced fabrication and characterization techniques for dielectrics and two-dimensional (2D) materials is opening a unique and unprecedented window of opportunity. This expansive window allows researchers and engineers to creatively bring together an exceptionally diverse class of materials in highly innovative and imaginative ways that have not been seen before. For instance, materials such as silicon, often referred to as a fast material due to its remarkable rapid response characteristics, when effectively combined with phase-change materials such as Germanium-Antimony-Telluride (GeSbTe), represent fundamental and transformative options for the fabrication of next-generation all-optical switching devices. These devices hold the promise of transformative performance enhancements that could redefine the landscape of photonic technologies and their diverse applications. In the realm of all-optical saturable absorbers, transition metal dichalcogenides, including the notable examples of MoS₂ or MoSe₂, can be effectively exploited to take full advantage of, and harness their unique and differentiating nonlinear optical properties. However, the challenge in this exciting and evolving research area lies in the complex task of matching nonlinearity among these disparate and distinct materials within composite systems. The primary design challenge is to not only optimize the nonlinearity itself but also to ensure that suitable phase-matching conditions are meticulously and effectively met to facilitate the intended functionality of the devices. Additionally, volume nonlinearity manifests itself in waveguides constructed from chalcogenides and even in clever, innovative polymers that can act as outstanding electro-optic modulators for advanced applications across various markets and fields. Furthermore, the sensitivity of devices can be meticulously refined and optimized within photonic bandgap crystals, where the fundamental and major effects of these materials include a slow group delay, combined with a small group velocity that ultimately enhances overall performance significantly. Nonetheless, it is essential to identify, address, and resolve several key practical challenges and pressing issues that must be thoroughly addressed

before these innovative and groundbreaking technologies can be deemed commercially feasible for widespread use in everyday applications. The capability of devices may be notably hindered by various losses occurring within the device itself or at critical device interfaces, which might adversely impact the overall performance and functionality. Moreover, many of these advanced devices may require elaborate and sometimes cumbersome experimental setups that can complicate the process of deployment, as they often necessitate a substantial degree of detailed signal processing to achieve the desired performance metrics in actual real-world deployment scenarios. This complexity indicates the vital importance of continued research and development efforts aimed at refining these technologies for practical use, ensuring that they not only meet but exceed the expectations of end users in various industrial applications [17, 7, 117, 118, 119, 13].

7.1 Nonlinear Waveguides

Waveguiding systems inevitably maintain an incredibly essential and prominent position within the captivating, intricate, and consistently developing field of photonics. These advanced systems currently function as critical components that are absolutely fundamental to the overall operational functionality of integrated photonic devices, which are increasingly utilized across a diverse, extensive array of research areas and practical applications. These highly innovative devices act effectively and efficiently as the optical counterparts of traditional electrical circuits, providing groundbreaking solutions and significantly augmenting capabilities across numerous applications in diverse fields spanning telecommunications, sensing technologies, biomedical applications, and beyond. Furthermore, the various very practical applications of waveguides, which have been thoroughly discussed and elaborated upon throughout this section, appear to remain predominantly confined to the well-established linear operational regime that is widely understood, accepted, and recognized within the scientific community. However, as the dimensions of their claddings and core materials begin to scale excessively large, the significant and profound influence of nonlinear effects begins to predominate and assert itself assertively, ultimately transforming the way in which these waveguides behave under a wide variety of different conditions. At the nanoscale level, these nonlinear effects typically manifest in fascinating forms like self-focusing, where the beam is intrinsically inclined to focus itself naturally due to the nonlinear interactions that are actively at play within the medium. Additionally, phenomena such as self-phase modulation occur, resulting in an evolution and intricate change over time in the beam's characteristics, including significant alterations in

frequency and phase in complex and multifaceted ways that reflect the intricate nature of light-matter interactions. These two critical and fundamental phenomena are absolutely paramount in determining how light propagates within waveguides, while their modulation is exclusively dictated by the intrinsic and inherent properties of the materials employed in these innovative waveguiding structures. In addition to the purely dielectric devices that have been the central focus of much extensive research and inquiry in this evolving field, there exist highly fascinating alternative devices through which light interacts intriguingly with materials exhibiting pronounced second-order susceptibility. This particular characteristic ultimately amplifies the manifestation of various nonlinear effects, rendering them even more pronounced and inherently significant within the unique contexts and operational parameters of those very devices. Within these distinctive types of devices, the demanding power requirements impose significant restrictions, so the length of the interaction between photons and matter is primarily determined by the careful selection of materials that possess the lowest potential losses, which is absolutely critical for optimal performance. These optimized materials often include advanced configurations such as etalon cavities, which are specifically engineered for optimal performance to maximize overall effectiveness and system reliability in delivering superior results. The pioneering theoretical framework that describes the complex dynamics of confined beam propagation was introduced in the highly notable year of 1975, marking a significant milestone within the rapidly advancing and intricate field of photonics research and applications. This theoretical projection suggested that any notable amplification in the local beam intensity would subsequently elevate the nonlinear refractive index right at the very center of the beam itself, thereby creating fascinating, dynamic changes in its overall behavior that merit further exploration. The notable and significant elevation in this refractive index is a direct result of a reduction in the carrier density within the beam, which leads to the intriguing, captivating phenomenon of self-focusing that plays a critical role in the overall optical waveguiding process. This phenomenon causes the beam's energy to become concentrated in an increasingly smaller area, generating a series of consequential events that ultimately produce a subsequent decrease in the effective beam radius. This notable reduction is subsequently followed by a corresponding increase in power density, leading to intricate and complex interactions happening within the waveguide medium that need careful consideration and review from researchers. In the context of modern and highly innovative waveguides, this fundamental shift results in a notable reduction of the core refractive index, effectively creating a waveguide

characterized by a power-dependent nonlinearity that significantly impacts its behavior and performance profile. This paradigm shift leads to profound implications for waveguide design, optimization, and overall operational performance across various applications, such as enhancing data transmission capabilities and improving the efficiency of nonlinear optical devices. Due to the inherent spatial confinement that exists within these sophisticated waveguides, the complex interactions of self-focusing and self-phase modulation can culminate in a local collapse of the beam inside the core, which poses unique challenges to overcome while simultaneously providing exciting and promising opportunities for further research and development in the constantly evolving and innovative realm of cutting-edge photonics technology. The ongoing exploration of these intricate and fascinating nonlinear phenomena could very well lead to groundbreaking advancements in technologies encompassing, but not limited to, optical switching, signal processing, and innovative laser design strategies that push the boundaries of what is currently feasible. As a result, these rapid developments could herald a new era in the capabilities and practical applications of photonic devices within various industries, offering transformative potentials that expand their utility. Such advancements will significantly enhance their functionality and performance across various fields, propelling the industry forward into enticing new realms of possibility, exploration, and practical application for the future of photonics and its transformative power in modern technology, which can redefine how information is processed and transmitted globally [120, 121, 122, 123, 124, 125, 126, 127, 128, 129].

Chapter - 8

Applications of Nonlinear Optics

The field of nonlinear optics has unmistakably witnessed an extraordinary and remarkable growth trajectory, accompanied by substantial advancements over the course of the past four decades. This transformation has profoundly altered its status, establishing it as one of the most exciting and rapidly evolving domains within the vast expanse of modern science. The immense and extensive scope of this periodical, which is dedicated entirely to the exploration and elucidation of this vital area of study, is exceedingly broad and encompasses a wide array of phenomena that are continuously evolving, growing, and expanding our understanding of the complex and nuanced interactions between light and matter in innovative and diverse ways. The extensive range of topics that can be observed in this captivating special issue beautifully highlights and underscores this very fact: applications of the most cutting-edge theories related to frequency manipulation, polarization adjustments, or the intricate process of phase mixing are rapidly emerging and making significant contributions to considerable progress within the field; the intricate and complex propagation of laser beams as they traverse through diverse inhomogeneous or anisotropic media presents an astonishing array of challenges that require innovative solutions, critical thinking, and groundbreaking methodologies; and the sophisticated and dynamic nature of laser-matter interactions observed across various regimes—such as multiphoton ionization, electron avalanche processes, or dielectric breakdown phenomena—are particularly noteworthy and represent significant areas of ongoing research that hold tremendous promise for potential future discoveries that could revolutionize our technological capabilities and deepen our understanding of the physical principles that are involved. Furthermore, the exploration into the intricate realms of atomic and molecular physics remains not only pivotal but also central to the ongoing advancement of our understanding in this all-encompassing and comprehensive field; developments in nonlinear optical spectroscopy and state-of-the-art imaging techniques are advancing at an exhilarating pace, continually pushing the boundaries of what is possible, attainable, and conceivable in the manipulation and capturing of light properties for a wide variety of innovative applications.

An intriguing array of diverse applications related to fiber optics are continually emerging, evolving, and adapting to meet the ever-growing and dynamically changing demands of modern technology and contemporary society; alongside this trend, there is also a cutting-edge focus on the generation and characterization of coherent radiation occurring across various spectral regions, which is gaining increasing attention. This includes not only ultraviolet (UV) light and visible light but also significantly extends into the infrared (IR) spectrum, where a wealth of intriguing possibilities, innovative solutions, and practical applications are becoming more apparent with each passing day as researchers diligently explore new methods for harnessing such emissions. In this broader context, we aspire to showcase some of the very latest and most captivating applications that are currently resonating within the expansive field of nonlinear optics. This endeavor ultimately aims to demonstrate that the theoretical advancements made in concert with the ongoing research efforts—efforts that are required to effectively address increasingly complex challenges in this area—are all excitingly intertwined within this vibrant and dynamic discipline, continuously inspiring future innovation, exploration, and devoted scientific inquiry. This periodical also hopes to elucidate, illuminate, and outline some of the promising directions that current research initiatives are undertaking, as well as where they may very well be headed on the near horizon in their relentless and dedicated pursuit of knowledge. This pursuit leads to a deeper and more comprehensive understanding of various phenomena that profoundly impact our daily lives, influence industrial applications, and shape the continuous development of emerging technologies. Additionally, nonlinear effects are being actively harnessed in a vast and varied array of practical applications, with telecommunications standing out as one of the most notable examples, where they demonstrate exceptional potential for facilitating essential processes such as wavelength conversion, signal regeneration, pulse reshaping, and a wide array of other crucial signal processing functionalities that are integral to the sophisticated and intricate infrastructure of modern communication systems. This ultimately enhances the overall efficiency and effectiveness of these systems, which serve as the lifelines of our interconnected world, enabling seamless global communication and dynamic information exchange. Furthermore, the fascinating realm of medical imaging and diagnostics is on the verge of experiencing significant enhancements through the innovative and strategic utilization of nonlinear phenomena. This paves the way for improved and more effective procedures such as angiography, advanced blood flow imaging, mammography, and various other crucial diagnostic techniques that are vital and indispensable for effective and efficient healthcare practices

in today's society. These remarkable advancements are helping to save lives and promote better health outcomes for individuals everywhere, representing a crucial intersection of technology and healthcare that is actively transforming medical practices for the better. Moreover, the ongoing development of new low-cost devices that are specifically tailored for high-performance spectroscopic analysis is actively paving new pathways for valuable and effective chemical analysis applications within both security and environmental monitoring contexts. This showcases the remarkable versatility, practicality, and profound relevance of nonlinear optics in our increasingly interconnected world, where scientific advancements play an essential role in addressing the contemporary challenges we face. In addition to this, the manufacturing sector is witnessing the innovative techniques of laser micromachining and nanoprocessing being actively pursued, particularly aimed at the precise and efficient production of advanced photonic devices as well as highly sensitive sensors that measure a broad array of various physical and chemical properties with unparalleled accuracy and reliability. This further illustrates the wide-ranging and diverse impact of nonlinear optics across various technological fronts and sectors in our rapidly advancing and constantly evolving world. The quest for progress, innovation, and the exploration of new frontiers in both science and technology is continually defining our reality, transforming possibilities into tangible realities that enhance the quality of life and push the boundaries of what humanity can achieve, while highlighting the critical and fascinating role that this essential field plays in driving forward the frontiers of knowledge and practical application across diverse disciplines and industries [130, 131, 132, 133, 134, 119, 135, 136, 137].

8.1 Biomedical Imaging

In recent years, the field of biomedical imaging has rapidly emerged as one of the most vital and indispensable tools utilized for diagnosis, drug development, and a myriad of other scientific studies that are critical to advancing healthcare. With this marked evolution in the field, there has been a significant increase in interest surrounding the captivating concept of molecular imaging. In response to this heightened interest, many innovative and novel techniques focused on multimodal imaging are currently being employed across various research and clinical settings, reflecting the forward momentum of scientific inquiry. Today, it is now more feasible than ever to visualize *in situ* biological processes occurring in living tissues or organs using increasingly sophisticated and advanced methods that allow for deeper penetration into tissues compared to traditional imaging modalities. Among

these innovative methods, nonlinear optical microscopy stands out as a widely favored technique specifically designed for biomedical imaging, particularly in scenarios where high-resolution imaging is of utmost importance to researchers and clinicians alike. This particular class of imaging technique adeptly combines multiple different approaches and methodologies, including two-photon microscopy, multi-photon microscopy, and second harmonic imaging, creating a synergistic effect that significantly enhances imaging capabilities beyond prior limits. The primary advantage of nonlinear optical microscopy lies in its remarkable ability to facilitate and achieve high-resolution imaging while utilizing autofluorescent proteins or harnessing the intrinsic contrast of regular biological tissue, which helps in drawing out critical contrasts that are vital for analysis. For such types of microscopy to be effective, the excitation light intensity must be sufficiently high, ideally achieving levels adequate to fully saturate or nearly saturate the nonlinear optical effects that are so essential to the imaging process. Generally, the predominant mechanism responsible for generating tissue contrast is closely related to the precise distribution of molecular chromophores or the presence of light-scattering structures that are carefully contrasted within the focal volume of the imaging device employed. This advanced capability allows researchers not only to observe a vast array of various cellular processes in real-time but also to delve deeply into the intricate study of disease mechanisms or potential disease progression, thereby enhancing our understanding of complex medical conditions. Indeed, two-photon imaging has showcased a broad range of valuable applications across multiple disciplines, making a significant impact in fields such as neuroscience, ophthalmology, and cancer research, where precise imaging is crucial for timely and accurate diagnoses. Additionally, the technique holds remarkable potential in real-time diagnostics, particularly when applied in an endoscopic configuration or employed as an invaluable tool during intraoperative diagnostics. This capability is especially beneficial in the realms of neurosurgery and other medical fields where rapid assessment and targeted interventions are critical to successful patient outcomes. However, despite the vast applications and widespread utility of nonlinear optical microscopy, there exist certain limitations in the breadth and scope of this technique. For instance, the imaging speed can be constrained by both the inherent nonlinear mechanisms that govern the imaging process as well as the scattering properties of the tissues being examined, which remain a topic of active investigation. In recent years, numerous publications have shed light on the notion that nonlinear optics can serve as a powerful instrument meticulously tailored for medical professionals' purposes, offering transformative insights

into disease pathology. Notably, case studies illustrate the potential of second harmonic imaging when utilized to investigate deeper tissues, showcasing the efficacy of the second harmonic to autofluorescence ratio of NADH as a means to vividly visualize the invasive front in cancerous tissues, thus enhancing diagnostic accuracy. Furthermore, when combined with complementary imaging techniques, nonlinear optical microscopy can significantly enhance or improve the real-time visualizations obtained during complex neurosurgical procedures. In their critical research, the group has elucidated the impressive capabilities of two-photon autofluorescence specifically aimed at the *in situ* diagnosis of skin cancer. The primary findings of this research illustrate the diverse types of cellular structures present in the epidermis, emphasizing subtle differences in the morphology and shape of cellular features among various samples analyzed. Notably, the phenomenon of nuclear pleomorphism, characterized by varying shapes and sizes of the nucleus within cells, can also be meticulously observed and recorded using these advanced imaging techniques, contributing to diagnostic differentiation and disease characterization. The aforementioned examples of applying nonlinear optics in imaging at the cellular level vividly underscore the extensive possibilities and groundbreaking potential of this advanced methodology in contemporary biomedical research. Beyond the inherent capabilities of nonlinear optics in facilitating detailed imaging, a significant role is also played by the ongoing stimulation and deliberate development of sophisticated image processing techniques that complement imaging. Analyses incorporating artificial intelligence facilitate the execution of quicker and more precise evaluations of cellular images, marking a monumental step forward in the field of medical imaging. This fusion of technology in imaging and analytical processes represents an inseparable element of real progress, especially in the context of contemporary technological advancements, and aligns seamlessly with the ideals of personalized medicine as the field continues to evolve and grow, paving the way for more effective and individualized therapeutic strategies [138, 139, 140, 141, 142, 143, 144, 145].

Chapter - 9

Future Directions in Nonlinear Optics

Although an extensive and nearly comprehensive array of nonlinear optical phenomena has been meticulously observed, examined, and documented throughout countless experiments and rigorous studies conducted over numerous years, there undeniably remains substantial space and considerable room for a vast multitude of new insights and groundbreaking advancements that exist within this vibrant and ever-evolving field of nonlinear optics. This ongoing continuation of exploration and research in the realm of nonlinear optics showcases its truly expansive, innovative, and highly exciting potential in the contemporary world of scientific research and experimentation, capturing the attention and fascination of both seasoned researchers and enthusiastic newcomers to the field alike. Indeed, nonlinear optics is currently at the forefront of scientific inquiry, driving numerous pioneering technologies that span across multiple scientific disciplines, intricately involving physics, engineering, materials science, and a plethora of additional fields that contribute to its richly diverse landscape and robust development. There is an increasingly keen and ever-growing interest in emerging technologies, which is anticipated to continue propelling the growth and ongoing evolution of this remarkable and promising field for many years to come, creating ample opportunity for new discoveries and applications that can advance not only technology but also our understanding of fundamental physical phenomena. A particularly compelling and noteworthy example of this sustained and profound growth can be prominently and clearly witnessed in the rapidly advancing field of supercontinuum light sources. These exceptional light sources are uniquely and remarkably capable of producing an extraordinarily broad and continuous spectrum of light, and they are increasingly proving to be essential tools in both rigorous scientific research as well as various practical applications that span across a broad range of different domains of science and technology. These extraordinary light sources are highlighted as a prime focus on nonlinear interactions taking place in innovative and newly developed materials, which are currently undergoing extensive research and in-depth development. This ongoing exploration is revealing their intriguing properties and fascinating capabilities, thus pushing the boundaries of what is currently feasible and achievable in the realm of

cutting-edge optical technology, opening new avenues for exploration and further innovation on multiple fronts. Alongside these exciting developments, the area of topological insulators is experiencing a significant surge in attention and recognition among researchers and scientists alike, especially when the unique characteristics of these materials are evaluated and assessed in conjunction with both metasurfaces and metamaterials. These advanced materials exhibit remarkably enhanced nonlinear responses that possess great potential to transform a wide array of applications in the practical world, reaching from telecommunications to advanced imaging systems and even beyond into previously uncharted territories of technological development. The recent surge of keen interest in the topological properties of light, which notably includes a thorough and detailed investigation of how these extraordinary properties can be effectively harnessed and utilized for practical applications, could pave the way for novel and innovative solutions. These emerging solutions would involve intricate interactions between nonlinear and quantum optical phenomena in forthcoming experiments and studies that endeavor to push the envelope even further and drive the field forward with new insights and breakthroughs. The merging of these two exciting and groundbreaking fields is anticipated to discover immensely promising applications in cutting-edge quantum technologies, as well as within sophisticated quantum communications and advanced quantum information processors. These processors are highly sought after and regarded as the future cornerstone of innovation in today's fast-evolving and diverse technology landscape, reflecting the ongoing evolution and continuous development in the fields of science and technology as a whole, and embodying the potential for groundbreaking advancements that could distinctly change the technological landscape. A significant and transformative paradigm shift is currently taking place in the way nonlinear technologies and processes are ingeniously engineered and meticulously developed to align with contemporary needs and objectives. Current trends are moving towards a more integrated and holistic approach that effectively considers a comprehensive range of multiple interrelated factors and elements at play in the design and overall functionality of these advanced technologies. Furthermore, specialized machine-learning techniques, including sophisticated artificial intelligence and potent deep learning algorithms, are now being employed in unprecedented and innovative ways to intelligently optimize the nonlinear processes that lie at the core of these forward-thinking technologies. This showcases the profoundly interdisciplinary nature of contemporary research as it amalgamates various domains of expertise and knowledge. This modern and contemporary approach is fundamentally characterized as being

convergent and open-ended, thereby allowing for exploration, innovation, and iterative refinement of technological designs and concepts. It moves beyond merely relying on a direct modification of a known or already understood device design or architecture, and instead wholeheartedly embraces and encourages the birth of novel ideas and groundbreaking inventions that have the potential to significantly change the landscape of various scientific fields and applications. While these groundbreaking technologies represent an intriguing and captivating direction in the expansive realm of nonlinear optics and its multitude of applications, there are certain challenges that undeniably arise—such as the scalability and economic costs associated with AI-assisted design methodologies and their effective implementation in practice across various sectors of the economy. These challenges need to be thoroughly examined, critically discussed, and resolved in future research initiatives in order to ensure the long-term viability and success of the technologies being developed for real-world applications. In addition, it is increasingly understood that real, meaningful progress in the rich and vibrant interdisciplinary fields of biology, chemistry, physics, and materials science—rather than relying exclusively on a singular approach that is narrowly focused on one discipline—will yield a wealth of new and exciting outcomes. These promising outcomes can significantly benefit a variety of fields and applications, thus shaping the trajectory of future scientific endeavors in an increasingly complex and interconnected landscape that demands interdisciplinary collaboration. Emerging trends demonstrating the miniaturization of integrated optical components, alongside their fascinating functionalities, are starting to manifest across a variety of applications, including those within the biomedical, quantum, and photonic sectors. In these cutting-edge areas, meaningful advancements and groundbreaking innovation are crucial for future discoveries and technological evolution, acting as a catalyst for further research and exploration. This ongoing exploration and persistent innovation are ensuring that the knowledge and technologies developed from these valuable interdisciplinary efforts will continue to enhance our understanding and capabilities in the years to come. By inspiring further research and development across various scientific fronts, the profound impact of interdisciplinary collaboration on scientific progress is not only magnified but also solidified, fostering a robust environment for innovation and discovery that promises to push boundaries and redefine possibilities in the future, bridging the gap between theoretical discoveries and practical applications across numerous fields [146, 147, 148, 149, 150, 151, 152, 153].

9.1 Quantum Nonlinear Optics

Until now, we have elucidated the intricacies of the expansive field of

geometrical optics, a discipline that continues to enrich our understanding of light propagation and interaction. This field, coupled with a detailed description that is fundamentally rooted in the principles of a Gaussian light-field propagating through a consistent and uniform linear medium, serves as an effective framework for comprehending the rapid and sometimes highly complex nature of ultrafast optical pulses. The phenomenon known as filamentation, which arises dynamically within these pulses, is essentially a consequence of their profound interaction with a diverse variety of gas media that encircle them and influence their behavior. When we consider this interaction on a macroscopic scale, the behavior of ordinary or standard light as it interacts with matter can be effectively modeled and described through the application of Maxwell's electromagnetic equations in conjunction with the eikonal equation, a formulation that relates to the rays of light that we commonly observe. However, as we endeavor to extend this traditional description into the nanoscale realm or to delve into the coherent regime that is frequently referred to as the so-called quantum limit, the reliability of our previous understanding becomes increasingly tenuous and questionable. It is in this challenging and intricate context that a more comprehensive and suitable analytical tool emerges in the sophisticated form of quantum mechanics, which contrasts significantly with the conventional Maxwell's theory of electromagnetism, thereby creating a rich ground for exploration and discovery. We now feel compelled to leverage quantum electrodynamics, which serves as a seamless and thorough amalgamation of quantum mechanics and electromagnetic theory, in order to uncover and unravel the intricate complexities associated with these interactions and the phenomena that arise from them. While the detailed narrative surrounding the principles of quantum mechanics will be systematically unveiled in the subsequent subsections of our discourse, we take this present opportunity to provide a foundational introduction or motivation by underscoring the pivotal point that quantum states and fields cannot be accurately localized simultaneously at the same instant. Instead, it is firmly established that these fields can only reveal localized quanta at any given moment in time, which are widely recognized as photons—the fundamental units that embody light. The behavior manifested by these photons is inherently wave-like in its essence, highlighted by their remarkable capacity to superimpose upon one another, in addition to delivering energy that is constituted of localized packets, efficiently contributing to the rich tapestry of our understanding of light. This wave-like nature of photons can be effectively illustrated through the use of laser beams as quintessential representatives of these principles. We have firmly established that a medium can undergo a second-order non-linear

susceptibility effect, routinely described in the scientific literature as chi-squared, which significantly contributes to the dynamic intricacies of these interactions. Ultimately, we find ourselves deeply immersed in the exciting realm of research known as quantum nonlinear optics—a vibrant, exhilarating field where the quantum states of light engage in complex and intricate interactions with nonlinear media, fostering an environment of discovery and innovation. The quantization process of electromagnetic fields, or the emergence of the quanta of modes, gives rise to peculiar effects and phenomena that are conspicuously absent in the semi-classical theory of nonlinear optics, marking a significant shift in our theoretical framework. This area of study constitutes the very core of our current educational curriculum, representing a lively and stimulating area of ongoing research inquiry, driven by curiosity and the desire to uncover the secrets of light. Herein lies a compelling and intriguing challenge: the paradigm shifts drastically as our comprehension of the interaction between light and matter evolves from a purely particle-based view to one that genuinely appreciates and celebrates the wave-like nature of light itself, enriching our understanding of both. Throughout the course of our studies, this exploration will extend into the intricate and complex field that encompasses the quantum many-body nature of light, revealing a tapestry of interaction unseen in traditional optics. Very recently, we have witnessed the innovative and powerful emergence of distinct quantum states of laser beams, particularly emphasizing the transformative production of entangled photons. Illustrative examples of these remarkable advancements include notable instances such as ladder states and self-Q-plasma states, each representing a fascinating evolution in our understanding of quantum mechanics. All of these remarkable nonlinear effects give rise to a stunning spectrum of quantum phenomena, paving the way for exhilarating advancements and breakthroughs in the expansive realm of quantum information engineering. Furthermore, it is of paramount importance to highlight that the generation of these non-classical states of light is not merely central to the evolutionary trajectory of groundbreaking quantum technologies but also encompasses the significant challenge of accurately characterizing and comprehending these newly emerging quantum technologies. In essence, all of the rapid advancements taking place within the domain of quantum mechanics are poised to elevate the entire field of nonlinear optics into a multifaceted quantum domain defined by intricate many-body interactions, ultimately reshaping our understanding of the universe around us. Currently, a multitude of laboratories across the United States and Europe are distinctly devoted to exploring the profound and complex quantum dimensions of nonlinear optics. We are witnessing the birth of a global collaboration

involving numerous experimental groups, each utilizing a diverse array of techniques and methodologies to delve into this interdisciplinary phenomenon, thereby enriching and enhancing our comprehensive understanding of quantum phenomena and their implications for future technologies [154, 155, 156, 157, 158, 159, 160, 161, 162].

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Conclusion

In conclusion, this paper discussed the nonlinear optical phenomena under the carbon laser ablation experimental and simulation studies showing the important role played by the K-M mechanism. The established theoretical principles are universal and may be used both for the fundamental research of interaction of intensive light and matter as well as for the mechanism developing for the ominous application: high-intensity pulsed laser energy delivery into the material. Different suggestions were provided for the potential endeavors in the latest developmental cognizance and exploiting the hitherto uninflected domain in physics, materials science, biological science, optics, and engineering. These intriguing phenomena are of interest not only to physicists working in the field of laser damage and high field science, but also to a multi-disciplinary audience working across a diverse array of research disciplines. It is worthwhile to note the contribution here to medicine, cancer therapy, ophthalmology, hard and soft tissue processing, spectroscopy, extreme ultraviolet and x-ray generation, application of high-power lasers to the generation of terahertz radiation, and many others fields in the materials science and high-energy physics research.

True to the collaborative spirit of all progressing science, the future of understanding and exploiting the nonlinear matter phenomena optically will respond sensitively to the beneficence of individuals from many backgrounds. Clearly, it is a fiendishly difficult no answer to the question of which of these proposals are the best avenue to pursue; and while the configuration of experimental groups in universities has a large impact on the choice, it is rare for major agencies to adopt such a stratagem. The advantages of developing new sources at the powerful apex of the NHPE have been overshadowed by both the complexity and costliness of the driver-lasers. The surface conversion techniques would scale poorly into the gigaJoule regime needed for fission, as the reduction in target diameter would have to necessitate extreme focusing, ineffective at incident intensities in excess of 10^{22} . New ablative pressures using increased-velocity, heavy-ion beams were proposed to confine fissional cores, achieving an order of magnitude increase in implosion density. Some of the key challenges facing nonlinear optics are linked to the limited range of

natural materials with the necessary nonlinear spectroscopic processes. Large bandwidths and high dispersive requirements can make it hard to achieve resonant enhancement of a linear coefficient in the same material, in particular in the solid state. Approaches based on optical frequency matter as well as the periodic poling in the solid state and chalcogenide glasses and organic materials and integrated photonics could be used to bypass these limitations.

10.1 Summary of Key Points

In this review we present some fundamental concepts, characteristics and results of nonlinear optics. We discuss different nonlinear optical effects and describe them in the context of relevant potential materials, often novel artificially created substances. We also discuss theoretical approaches to nonlinear optical phenomena, based on numerical calculations or analytic models. The presented theoretical analysis is validated on experimental demonstrations, including pulsed laser beams passing through specific materials. Such materials are chosen because their specific properties highlight and enhance particular aspects of nonlinear optical effects. Nonlinear optics has a broad range of applications, in various fields and domains of science and technology. For example, in telecommunications, soliton propagation through fibers is an application of the balance between dispersion and nonlinearity, while biomedical imaging and optical arts are enabled by highly specific processes involving interaction of ultrafast light pulses with biological tissues and materials. Optical sensors leveraging nonlinear optical effects for measuring light chromatic dispersion in specific materials are also used in astronomy to search for exoplanets. Several keywords, such as second-harmonic generation, revealed a broad interest and ongoing research activity in these directions. During the last decade, particular interest has focused on nonlinear optical phenomena in novel materials. Today, the research community is increasingly involved in applying nonlinear optics in quantum and other highly advanced and rapidly developing areas, such as quantum information technologies, quantum communication and quantum computing. Ongoing, current and future challenges are to develop a range of innovative materials, to establish a thorough understanding of their properties, and design systems, devices and methods for nonlinear optics based on the results of mathematical and theoretical modelling. These directions are expected to lead to a step further towards more extensive practical applications in industry. As a consequence, researchers working on nonlinear optics and laser-matter interaction face a rapidly expanding field with an increasing need for new results and innovative solutions.

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