LIGHT GENERATION AND MANIPULATION FROM NONLINEAR RANDOMLY DISTRIBUTED DOMAINS IN SBN

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UNIVERSITAT POLITÈCNICA DE CATALUNYA

BARCELONA, 2014

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submitted this thesis in partial fulfillment

of the requirements for the degree of

DOCTOR

by the

UNIVERSITAT POLITÉCNICA DE CATALUNYA

BARCELONA, 2014

Dedicated to my loving family

Acknowledgments

First and foremost, I want to express my sincere gratitude to Prof. Dr. Jordi Martorell, my supervisor of this PhD thesis, for providing me the opportunity to work on the project “Generation of light in random structures”, guiding and motivating me all through the work of this thesis. With his immense experience and knowledge, meticulous and rigorous scholarship, he has always been giving me valuable advice, and inspiring me to overcome all the challenges we ever met.

Deepest gratitude is also due to Dr. Francisco Rodríguez, without whose knowledge and assistance this study would not have been successful. Dr. Francisco has accompanied me all through this project, and I have learnt a lot from him, both in knowledge and his strict attitude in science. I am greatly indebted to him.

I also give my sincere gratitude to Prof. Dr. Jorge Bravo-Abad for his valuable help in developing the theoretical models used in the thesis.

I would also like to take this opportunity to thank all the work members in ICFO-the institute of photonic sciences, whose valuable dedication to the administrative and instrumental services has provided me an excellent environment to focus on my work. Besides, they have made my time here more pleasant and memorable. I thank ICFO for giving me so many opportunities to learn new stuffs beyond science, and I will benefit from the invaluable knowledge and skills in language, business, and communication in a very long term.

Special thanks go to all my past and present group members, Roberto, Luis, Marc, Sergio, Alberto, Marina, Luat, Xavi, Rafael, Pablo, Camila, Paola, David, and Francesco, and other ICFO colleagues, Johann, Tonglai Chen, Xiaojuan Shi, Na Lou, Xiaojie Yang, Chaitanya, Sotiris, Dhriti, Chirag, Giorgio, Armand, Marc, Ulrich, Naeimeh, Carmen, Luis Martinez, and many others, for the discussion in science, advice in experiment set-up, or sample processing, especially, their friendship and help in my life.

I also would like to thank Prof. Yuren Wang, my supervisor of the master thesis in photonic crystal in Chinese Academy of Science, who guided me the first step into the field of photonics, and he has been giving me invaluable advice not only in academy but also in my personal life.

I would like to express my deepest love to my dearest family. I thank my dearest husband, Dr. Xingxing Huang. For his continuous encouragement and absolute support, I am able to do the best that I can. I thank my dearest parents and parents in law, Baoxiang Ma, Zhenshan Yao, Pengzhen Yang and Jinrui Huang, for their forever confidence in me. I also thank my dearest brother, Yan Yao, who has been taking good care of my family when I am studying abroad. Special thanks also go to my daughter Yueyao Huang, who has been a source of happiness and a huge motivation of fulfillment during my thesis writing.

Abstract

Disordered media with refractive index variations can be found in the atmosphere, the ocean, and in many materials or biological tissues. Several technologies that make use of such random media, as image formation, satellite communication, astronomy or microscopy, must deal with an unavoidable light scattering or diffusion. This is why for many years light propagation through random media has been a subject of intensive study. Interesting phenomena such as speckle, coherent backscattering or random lasing have been discovered and studied. More recently, researchers are beginning to investigate mechanisms to control light propagation through such media to enhance light transmission and sharpen the focus.

On the other hand, it has been known for several years that nonlinear random structures are able to generate light in an ultra-broad frequency range, without the need of angle or temperature tuning. Particularly interesting is the nonlinear light diffusion observed from materials with no change in the refractive index and which appear to be fully diffusion less to linear light propagation. However, a comprehensive understanding of the scattering when a nonlinear interaction takes place has not yet been given.

The core of the thesis focuses on the study of the nonlinear light generation and propagation from crystalline structures with disordered nonlinear domains but with a homogenous refractive index. A random distribution of non-linear domains is found naturally in the Strontium Barium Niobate (SBN) ferroelectric crystal. As opposed to other mono-domain nonlinear optical crystals commonly used for frequency up-conversion, such as Potassium Titanyl Phosphate (KTP) or Lithium Niobate ($LiNbO_3$), in SBN the nonlinear domain size is, typically, on the order of the coherence length or many times smaller than the size of the whole crystal. Such domains are usually several times longer in the c-axis direction relative to the plane perpendicular to that axis. Adjacent domains exhibit antiparallel polarization along such crystalline axis, with no change in refractive index.

In Chapter 1 we give a brief introduction to light generation and propagation in random media, describing the speckle, light manipulation and second harmonic generation (SHG).

In chapter 2, we study the nonlinear light generation and manipulation from a transparent SBN crystal. In its theoretical description we use a two-dimensional random structure consisting of a homogeneous background polarized in one direction with uniform rectangular boundaries, and a group of square reverse polarization domains with random sizes and located in random positions. The SHG from each domain is obtained using the Green´s function formalism. In the experiments, we alter the ferroelectric domain structure of the SBN crystal by electric field poling or thermal treatments at different temperatures. The SBN crystal structures after such different treatments are shown to be characterized by their SHG patterns.

In chapter 3, by measuring the spatial distribution of the second harmonic light in the c-plane, we demonstrate that the randomness in the nonlinear susceptibility results in a speckle pattern. We explain the observations as a result of the linear interference among the second harmonic waves generated in all directions by each of the nonlinear domains.

In chapter 4, we report on our experimental implementation of the wave-front phase modulation method to control and focus the SHG speckle from the random SBN crystal. This research creates a bridge between light phase modulation and nonlinear optics. Finally we perform a theoretical analysis to demonstrate enhanced efficiencies for nonlinear light focusing by the wave-front phase modulation method in different directions. Various types of nonlinear structures are considered, including the homogeneous rectangular crystal, the group of random domains, and the combination of both.

Resumen

Los medios desordenados con índices de refracción variables se pueden encontrar en la atmósfera, el océano, en muchos materiales o tejidos biológicos. Varias tecnologías que hacen uso de dichos medios como la formación de imágenes, la comunicación vía satélite, la astronomía o la microscopía, deben afrontar la dispersión o difusión de la luz. Por este motivo la propagación de la luz a través de medios aleatorios ha sido investigada exhaustivamente desde hace muchos años. Se han descubierto y estudiado fenómenos como el moteado, la retrodispersión coherente o el láser aleatorio. Recientemente, se están empezando a investigar varios mecanismos que permiten controlar la propagación de la luz a través de tales medios para aumentar la transmisión y mejorar el enfoque de la luz.

Por otra parte, se conoce desde hace tiempo que las estructuras aleatorias no lineales son capaces de generar luz en un rango de frecuencia ultra-ancha, sin la necesidad de sintonización de ángulo o temperatura. Es interesante la difusión no lineal de luz observada en materiales que no cambian su índice de refracción y que no presentan difusión a la propagación lineal de la luz. Sin embargo, aún no se ha

dado una explicación completa de la dispersión que se produce cuando tiene lugar una interacción no lineal.

El núcleo de la tesis se centra en el estudio de la generación y la propagación de luz no lineal en estructuras cristalinas con dominios no lineales desordenados pero con un índice de refracción homogéneo. Una distribución aleatoria de dominios no lineales se puede encontrar en el cristal ferroeléctrico de estroncio-bario-niobato (SBN). A diferencia de otros cristales no lineales monodominio, de uso común para la conversión ascendente de frecuencia, tales como el fosfato de potasio titanil (KTP) o el niobato de litio (LiNbO3), en el SBN el tamaño de dominio no lineal es típicamente del orden de la longitud de coherencia o más pequeño que el tamaño del cristal. Tales dominios son por lo general mucho más largos en la dirección del eje c en comparación con la dimensión en el plano perpendicular a ese eje. Dominios adyacentes exhiben polarización antiparalela a lo largo de dicho eje cristalino sin cambio en el índice de refracción.

En el capítulo 1 se proporciona una breve introducción a la generación y la propagación de la luz en medios aleatorios, describiendo los fenómenos de moteado, manipulación de la luz y generación de segundo armónico (SHG).

En el capítulo 2, se estudia la generación y la manipulación de luz no lineal a partir de un cristal transparente SBN. En su descripción teórica la SHG de cada dominio se obtiene usando el formalismo de la función de Green. En los experimentos, se altera la estructura del dominio ferroeléctrico del cristal SBN por polarización del campo eléctrico o tratamiento térmico a diferentes temperaturas. Las estructuras cristalinas SBN después de tales tratamientos se caracterizan por sus patrones de SHG.

En el capítulo 3, mediante la medida de la distribución espacial del segundo armónico en el plano c, se demuestra que la aleatoriedad de la susceptibilidad no lineal resulta en un patrón moteado. Se explican teóricamente las observaciones presentadas como resultado de la interferencia lineal entre las ondas de segundo armónico generadas en todas las direcciones por cada uno de los dominios no lineales.

En el capítulo 4, se describe el montaje experimental planteado para controlar y enfocar el moteado SHG del cristal aleatorio SBN por medio del método de modulación de fase del frente de onda (MFFO). Finalmente se realiza un análisis teórico para aumentar las eficiencias del enfoque de luz no lineal en diferentes direcciones por el método MFFO. Se consideraron diversos tipos de estructuras no lineales como un cristal homogéneo rectangular, un grupo de dominios aleatorios y la combinación de ambos.

Publications

1. C. Yao, F. J. Rodríguez, J. Bravo-Abad, and J. Martorell, "Wave-front phase-modulation control and focusing of second-harmonic light generated in transparent nonlinear random structures," Physical Review A, vol. 87, no. 063804, Jun. 2013.

2. C. Yao, F. J. Rodriguez, and J. Martorell, "Controlling the diffused nonlinear light generated in random materials," Optics letters, vol. 37, no. 10, pp. 1676–1678, May 2012.

3. F. J. Rodríguez, C. Yao, J. L. Domínguez-Juárez, J. Bravo-Abad, and J. Martorell, "Observation of speckle pattern formation in transparent nonlinear random media," Optics letters, vol. 36, no. 8, pp. 1347–1349, Apr. 2011.

Contents

Chapter 1

Introduction

Light propagation in random structures has attracted much attention due to coherent effects such as random lasing, coherent backscattering and light localization. Generally speaking, the term ¨random structure¨ in the linear optics context, refers to structures with inhomogeneous refractive index distribution, such as powders or suspensions. However, in nonlinear optics, there are structures with a homogeneous refractive index but a disordered distribution of the nonlinear susceptibility. For example, some ferroelectric crystals, such as Strontium-Barium Niobate (SBN), are transparent for linear light propagation, whereas the anti-parallel polarization of its ferroelectric domains may be distributed randomly in the crystal. Thus, its nonlinear light generation presents effects from a disordered structure.

The goal of this thesis is to investigate the coherent effects in the nonlinear light generation in random structures. Additionally, inspired by recent works in the control of linear light scattering through random media, we implement a novel method to re-concentrate the scattered light generation in random nonlinear materials.

This chapter contains three sections. The first section provides a general introduction to linear light propagation and generation in random media. This includes single and multiple scattering. Some examples of physical effects that appear in such kind of media are mentioned: coherent backscattering, random lasing, and speckle patterns. The concept of focusing light through random media is also briefly explained. More details about that can be found in chapter 3. The second section introduces the physics of nonlinear light generation and, more specifically, second harmonic generation. A special emphasis is given to phase-matching effects since they will be important to understand the following section and most part of this thesis. In the third section, we introduce the effects of a random structure on the nonlinear light generation.

1.1 General introduction to light propagation and generation in random media

When a beam of light propagates in a non-absorbing medium with a disordered inhomogeneous refractive index, the incident energy may be spread in all directions while the frequency of the incident beam remains unchanged. This phenomenon is called elastic scattering, and, the scattered field can be computed by solving Maxwell wave equations for the macroscopic electromagnetic field subject to the appropriate boundary conditions [1].

Single scattering and multiple scattering

Depending on the complexity of the random system, there are two kinds of light scattering limiting regimes, single scattering and multiple scattering. In random media with a small number of scatterers separated with sufficiently large distances, light scattering is best described with the single light scattering limit where the light scattered from each scatterer is not affected by the presence of the rest of scatterers. In that limit, the total light scattering from the random media at the far field can be described by the coherent sum of the fields scattered from each scatterer.

On the other hand, a multiple scattering theory is needed for random media with a large number of scatterers, occupying a large portion of the total volume. In this case, the light intensity received by each scatterer from the rest of the scatterers may be larger than the light intensity received from the incident field itself. The description of scattering effects from such media may become complicated but, many interesting optical phenomena originate from such type of scattering.

For example, coherent backscattering is an enhanced scattering in the backward direction due to the interference among multiple time reverse light scattering paths [2-9]. See Figure 1.1a, two input beams A_{in} and B_{in} are scattered back in the directions A_{out} and B_{out}, and coherent interference occurs, while one can be obtained from the other by time inversion, when the angle between A_{in} and B_{out}, or B_{in} and A_{out} is zero. Light localization may happen in a random structure with high density of scatters, where some photons are trapped in a single loop path [10]. In that event the scatterers would play the role of an optical cavity. When the scatters are embedded in a gain medium, random lasing can occur [11-19]. In a random laser, light is confined not by conventional mirrors but by random multiple scattering (See Figure1.1b).

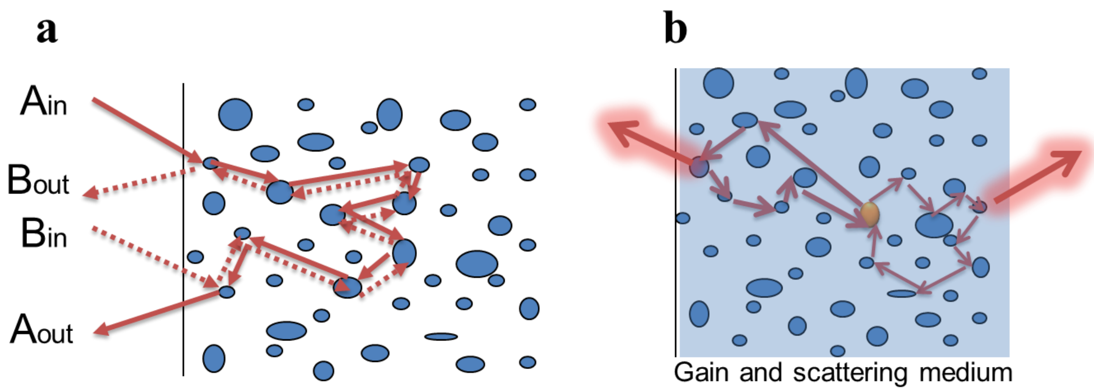

Figure 1.1: Schematic explanation of (a) coherent backscattering. (b) random lasing.

Speckle

When monochromatic and coherent light, such as laser light interacts with disordered media, the light is scattered into many different light paths with different propagation directions. The scattered light suffers constructive and destructive interferences forming an intensity pattern with bright and dark spots randomly distributed in space. This pattern is called the speckle. Some applications of the speckle effect have appeared in different fields since the invention of the laser [20-26]. Since it carries the material surface´s deformation information, it has been exploited, for instance, in holographic interferometry [27, 28]. On the other hand, speckle can be problematic in other applications such as, for example, in laser based display systems. In that event, reducing it becomes an important issue. Speckle reduction can be achieved by several means that produce many independent speckle patterns that are later averaged out on the detector [29, 30].

Control of linear light propagation in random medium

Interestingly, if the beam incident on the random medium is not a plane wave of light, but some structured beam with modulated phases on its cross-section, there is a special situation when the light will interfere constructively at one single point after the random media, while the intensity vanishes at other points. In this case, the speckle disappears and one obtains a sharply focused beam. The principle of light focusing after random media is attributed to the time reversal symmetry (See Figure 1.2), which can be realized by the phase conjugation method. The phase conjugation can be obtained with different techniques, such as phase conjugation mirrors based on nonlinear effects, transmission matrix measurement and subsequent wave-front shaping by a spatial light modulator, or wave-front shape optimization using a feedback algorithm, etc. [31-35]. Furthermore, without the need of any lens, researchers have achieved sharp focusing beyond the diffraction limit under these operations [35, 36].

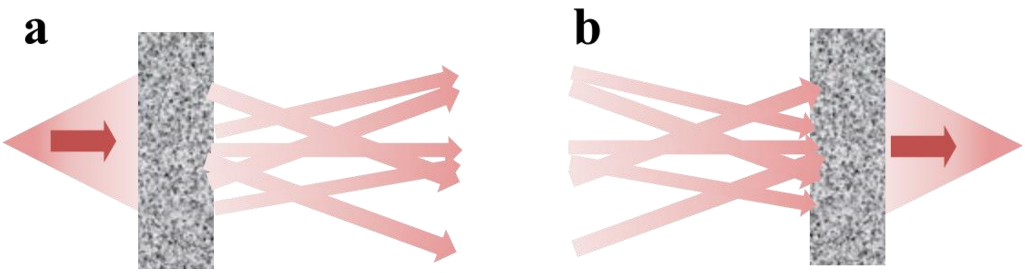

Figure 1.2: Schematic of (a) light scattering through a diffusive material and (b) light focused after one diffusive material under a wave-front shaped incident beam.

1.2 Nonlinear light propagation and second harmonic generation

Nonlinear light generation description

Light matter interaction can be described by using the relation between the electric field of light ***E(t)*** and the polarization ***P(t)*** of the material. In linear optics, the polarization induced in the material depends linearly on the electric field of light, and ***E(t)*** and ***P(t)*** have the linear relation:

$$\boldsymbol{P}(\boldsymbol{t}) = \varepsilon_0 \chi^{(1)} \boldsymbol{E}(\boldsymbol{t}) \tag{1.1}$$

where ε_0 is the electric permittivity of free space, and the constant $\chi^{(1)}$ is the linear susceptibility of the material.

In nonlinear optics, the optical response can be described by expanding ***P(t)*** into a power series of ***E(t)*** [37]:

$$\boldsymbol{P}(\boldsymbol{t}) = \varepsilon_0 \left(\chi^{(1)} \boldsymbol{E}(\boldsymbol{t}) + \chi^{(2)} \boldsymbol{E}(\boldsymbol{t})^2 + \chi^{(3)} \boldsymbol{E}(\boldsymbol{t})^3 + \cdots \right) \equiv \boldsymbol{P}^{(\mathbf{1})}(\boldsymbol{t}) + \boldsymbol{P}^{(\mathbf{2})}(\boldsymbol{t}) + \boldsymbol{P}^{(\mathbf{3})}(\boldsymbol{t}) + \cdots \tag{1.2}$$

$\chi^{(2)}$ and $\chi^{(3)}$ are known as the second and the third order nonlinear susceptibilities of the material. $\chi^{(2)}$ is a third-rank tensor and $\chi^{(3)}$ is a fourth-rank tensor. Due to the fact that in centro-symmetric materials, the nonlinear susceptibilities remain unchanged under symmetry operations, all the even-order susceptibilities such as $\chi^{(2)}$ and $\chi^{(4)}$ etc., are 0 for all such materials. Many of the ferroelectric crystals are non-centro-symmetric at room temperature, so they may produce effective even-order nonlinear effects.

Second harmonic generation (SHG)

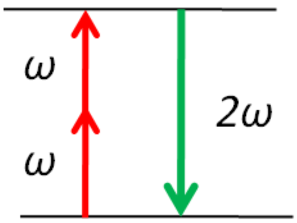

Figure 1.3: Energy diagram of second harmonic generation.

SHG is one of the most studied and used for applications second-order nonlinear optical process, where the power of the incident field at frequency ω is partially transferred into an oscillation at double frequency 2ω (See Figure 1.3) [38]. The polarization oscillating at 2ω acts as a source of electromagnetic dipole radiation. Due to the dispersion in the refractive index, the fundamental wave travels at different phase velocity relative to the SH wave. Thus, the generated second harmonic waves at different locations in the nonlinear material can interfere constructively or destructively. Their sum increases due to their constructive interference up to the coherence length L_c, at which distance the polarization and the generated second harmonic waves come out of phase, and the total SHG begins to decrease due to the destructive interference (See Figure 1.4 a).

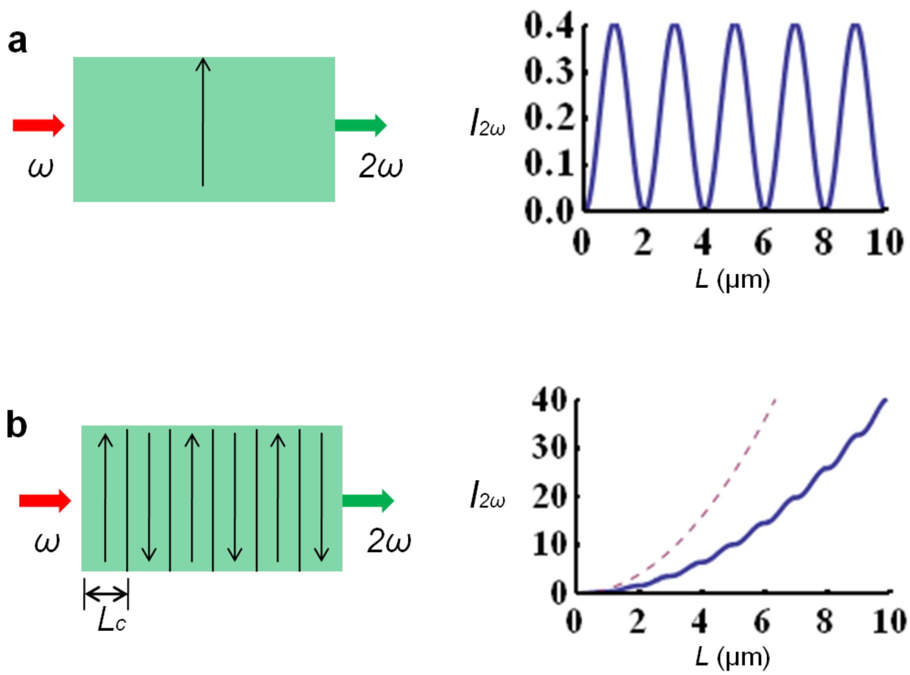

Figure 1.4: Spatial evolution of the second harmonic intensity from (a) a non-phase-matched crystal and (b) a quasi-phase-matched crystal. The dashed line in (b) represents the perfect phase-matching situation.

In order to obtain constructive interference over longer distances, some kind of phase-matching is required to maintain a fixed phase relation between the generated waves and the nonlinear polarization. This allows for the incident energy to be continuously transferred from the fundamental to the SH waves. With the quasi-phase-matching method, the nonlinear crystal axis is flipped at regular intervals, for example, by periodically poling a ferroelectric crystal. This allows the polarization response of the crystal to be shifted back in phase with the pump by reversing the nonlinear susceptibility. In this way, one can get a SHG signal that grows quadratically with the number of nonlinear domains (See Figure 1.4 b).

1.3 Phase-matching in random media

There are, essentially, two ways to meet the phase-matching conditions in different nonlinear structures to obtain efficient SHG. One way is by using the birefringence of the material [37]. The alternative is quasi-phase-matching by periodic pattern of the nonlinear susceptibility, as it was explained in the previous section [39-41]. However, these techniques are limited by the modest range of linear and nonlinear properties of natural materials, or require the complication of producing artificial structures or materials. Some ferroelectric crystals present naturally disordered structures that can provide relatively efficient light generation in an ultra-broad frequency range. The efficiency is lower than that in perfectly phase-matched structures but can be much higher than that in non-phase-matched materials.

According to the phase-matching condition which is required for momentum conservation during the SHG process, the grating vectors for the domain distribution of the nonlinear material $\boldsymbol{k_g}$ should satisfy the vector equation $\boldsymbol{k_2}=2\boldsymbol{k_1}+\boldsymbol{k_g}$, where $\boldsymbol{k_2}$ is the wave vector of the SHG light, and $\boldsymbol{k_1}$ is the wave vector of the fundamental beam [42].

$$|\boldsymbol{k_2}| = \frac{4\pi}{\lambda_1} n(2\omega_1),\ \ |\boldsymbol{k_1}| = \frac{2\pi}{\lambda_1} n(\omega_1),\ \ |\boldsymbol{k_g}| = \frac{2\pi}{2D} \tag{1.3}$$

Here $2D$ is the domain periodicity, λ_1 and ω_1 are the wavelength and frequency of the fundamental beam, and $n(\omega_1)$ and $n(2\omega_1)$ are the refractive indices of fundamental and SH light.

In the case of nonlinear light generation from random materials, the wavelengths of the fundamental beam and SHG signal are fixed, which means fixed absolute values of $\boldsymbol{k_1}$ and $\boldsymbol{k_2}$. A scheme of the quasi phase-matching condition is shown in Figure 1.5. We may see that, the larger the angle between the fundamental beam and the SH one, the larger $\boldsymbol{k_g}$ is needed for phase-matching while $\boldsymbol{k_g}$ should take values between [43]:

$$|\boldsymbol{k_2}| - 2|\boldsymbol{k_1}| \le |\boldsymbol{k_g}| \le |\boldsymbol{k_2}| + 2|\boldsymbol{k_1}| \tag{1.4}$$

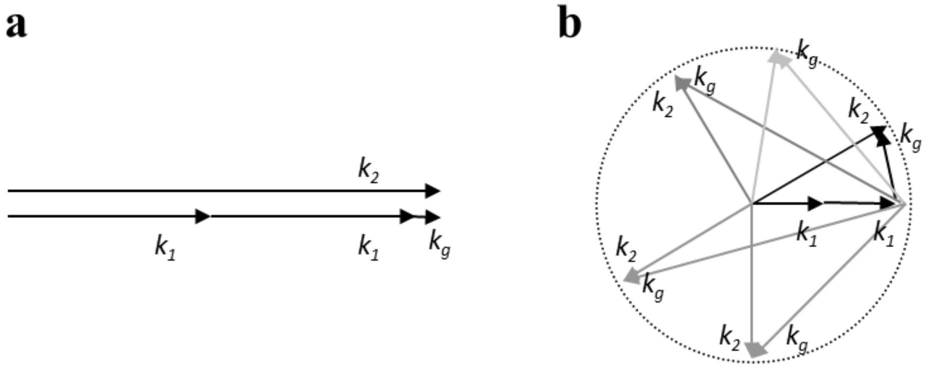

Figure 1.5: Schematic of the phase-matching relation between $\boldsymbol{k_1}$, $\boldsymbol{k_2}$, and $\boldsymbol{k_g}$ in the case of SHG in disordered media. (a) Collinear SHG. (b) Non-collinear SHG.

The SHG from disordered media grows linearly with the material length along the fundamental beam path (See Figure 1.6) [44].

To obtain a higher SH intensity from a random structure, besides increasing the material length, a better understanding of the relation between the SHG and the structure of the nonlinear random media is needed, so that the SHG efficiency can be optimized by modifying the random structure [45]. Moreover, with the wave-front shaping method, it is possible to concentrate the SHG after the disordered nonlinear structure, providing a new way to improve the SHG efficiency without modifying the random structure [46].

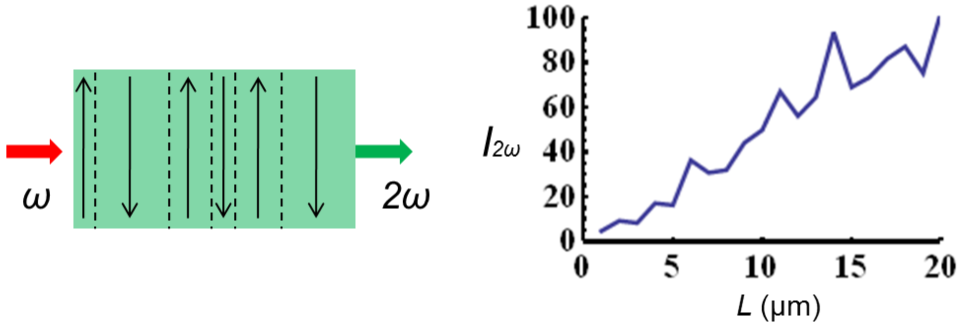

Figure 1.6: Spatial evolution of the second harmonic intensity from a disordered crystal.

1.4 Thesis summary

As indicated, light generation and propagation in random media is unavoidable in nature and frequently encountered. The coherence in the propagation and generation of light in such random media has been thoroughly studied leading to new applications such as coherent backscattering microscopy, random lasing and opaque lenses. When we consider the non-linear interaction in such random media, the coherent character or not of the phase-matching between interacting waves may play a determining role in the characteristics of the generated light. The aim of this thesis is to elucidate the intricate nature of such interaction and to propose new paths to counteract the effect of randomness in disordered non-linear media.

The remaining chapters explore the nonlinear coherent effect in a random media with homogeneous refractive index but a random distribution of nonlinear susceptibilities. Chapter 2 describes a theoretical model of the SHG from the whole random structure based on the interference of the single scattering from each domain of the structure, and this model successfully illustrates the relationship between the nonlinear light generation and the random structure. Chapter 3 reports a unique nonlinear light speckle pattern generated from a transparent nonlinear random structure. Chapter 4 first presents the experimental result of focusing the second harmonic speckle reported in chapter 3 by the wave-front shaping method, and then, with the theoretical model derived in chapter 2, demonstrates the different control behavior of the second harmonic light at different output angles.