CHAPTER 1

INTRODUCTION

1.1 Nonlinear Optical Materials

1.1.1 Background

The discovery of optical second harmonic generation in crystals by Franken et. al \(^1\), back in 1961, shortly after the demonstration of the laser spurred rapid progress in nonlinear optics (NLO). The success of their experiment saw early work in nonlinear optics concentrating on second harmonic generation. Since a laser can provide sufficiently strong, coherent radiation that can be efficiently converted in frequency by nonlinear optical effects, the rapid development was mainly due to the progress in laser technology. Scientists realised that any practical application of NLO would depend on the development of new materials. The primary application of nonlinear optical materials is the generation of new frequencies not available with existing laser sources. Tunable laser sources, ranging from the ultraviolet to the infrared (IR) may be achieved by frequency conversion of fixed-wavelength or tunable lasers by using nonlinear crystals.

With considerations of wave propagation effects and polarization, the nonlinear optical interaction in materials can be understood as the interaction between the phase of the propagating wave and generated polarization. In NLO, this is referred to as phasematching which will be discussed in the next
chapter. For second harmonic generation, phasematching implies that the phase velocity of the fundamental and second harmonic waves are equal in the nonlinear material. Optical materials are dispersive and it is not possible to achieve equal phase velocities in isotropic materials. However, Kleinman, Giordmaine, Maker et. al, and Akhmanov et. al demonstrated that phase velocity matching could be achieved in anisotropic, birefringent crystals using the birefringence to offset dispersion.

Second harmonic generation (SHG) and sum frequency mixing (SFM) have been widely used for frequency up-conversion, that is, the shifting of an optical beam to shorter wavelengths. Optical parametric oscillation (OPO), on the other hand, provides a means for frequency down-conversion with tunable coherent output. OPOs are powerful solid-state sources of coherent radiation with potentially large continuous tuning ranges. The possibility of optical parametric gain was first considered theoretically by Kingston, Kroll, Akhmanov and Khokhlov and Armstrong et. al. Adequate parametric gain with coherent oscillation was demonstrated by Giordmaine and Miller using lithium niobate, LiNbO₃. OPO in nonlinear crystals is perhaps the most unique aspect of nonlinear interactions as it generates coherent continuously tunable laser-like radiation. With suitable nonlinear optical crystals and pump sources, virtually any wavelength ranging from the ultraviolet (UV) to mid-infrared (IR) can be reached by OPOs.
1.1.2 Experimental Developments and Applications

As mentioned in the last section, early work in nonlinear effects were on SHG of laser radiation. Among all lasers, the neodymium lasers are most popular and are regarded typical representatives of the solid-state laser family. Examples are neodymium doped yttrium aluminium garnet (YAG), yttrium lithium fluoride (YLiF₄) and yttrium ortho-vandate(YVO₄). Most often the Nd:YAG laser is used, emitting at wavelength 1.06μm.

In a nanosecond regime, with a rather low pulse repetition rate of frequency, cesium dihydrogen arenate (CDA) and its deuterated isomorph, D-CDA are mainly used. Owing to their phase matching angle of 90°, hence a great angular bandwidth of SHG and a birefringence angle that equals to zero. This permits focusing of the fundamental radiation into the crystal as demonstrated by Kato. Power conversion efficiencies in the CDA and D-CDA were 57% and 45%, respectively. A 50MW Nd:YAG laser was used as the pump source with 12ns pulse duration. Because of a large angular bandwidth at θ=90°, CDA and D-CDA crystals are especially suitable for frequency doubling of multimode pulsed Nd:YAG radiation. Lithium niobate (LiNbO₃) are also used, but they have such drawbacks as photorefractive effect, that is, a change in refractive indices under the effect of laser radiation. Mg:LiNbO₃ and potassium titanyl phosphate (KTP) have large nonlinearities with relatively small beam walkoff, properties which are important in continuous wave (cw) operation. Potassium dihydrogen phosphate (KDP) offers excellent match of group velocities for 1064 to
532nm harmonic conversion which is needed when fundamental radiation is phase-modulated. The 0.03cm\(^{-1}\) absorption of KDP at 1.06\(\mu\)m might be a problem but can be reduced by deuteration in D-KDP to 0.005cm\(^{-1}\). This nonlinear material has been most commonly used during the past decade for frequency conversion of infrared solid-state laser radiation to generate intense laser radiation in the visible and ultraviolet.

1.2 Beta Barium Borate

The discovery of a new nonlinear crystal, beta barium borate (BBO) promised better conversion in harmonics generation of Nd:YAG lasers. The nonlinear optical properties of BBO have been demonstrated in the generation of second through fifth harmonics of 1.06\(\mu\)m neodymium laser radiation\(^{13}\). Besides frequency conversions, BBO has been a useful material in the development of reliable, widely tunable optical parametric oscillators\(^{15} - \)\(^{17}\). Compared to other nonlinear crystals, BBO has the advantages of being mechanically hard, chemically stable, and only slightly hygroscopic. Having a high damage threshold, it is transparent from 190nm to 3500nm and has a relatively large birefringence. It can, therefore be phase matched over a large spectral range. The effective coefficients of BBO is not large, but it is adequate for OPO applications with reasonable length of the crystal and pump power\(^{18}\).
1.3 Contents

The dissertation is divided into five main chapters which describe the experimental aspects of harmonic generation and parametric oscillators.

Chapter Two begins with the fundamentals of three-wave mixing and the propagation of light waves in uniaxial crystals. Optical parametric process is described which includes parametric oscillator tuning, gain, threshold and pump characteristics. Phase matching angles are calculated for BBO using simple mathematical formulas evaluated from Sellmeier dispersion relations.

Chapter Three describes the second harmonic generation of 1.06μm Nd:YAG laser radiation. Properties of nonlinear crystals, KD\*P and D-CDA as second harmonic generators are compared.

Chapter Four deals with 532nm pumped BBO OPO. Experiments with three different cavity configurations are detailed. This chapter begins with a section on straight cavity pumping and results that demonstrated its tunability and efficiency are presented and discussed. Two other section presents results from experiments using a pump reflector and a cavity design with intracavity pump steering mirror.

Observations and results of fourth harmonic generation are given in detail in Chapter Five. Problems faced in UV generation using both BBO and KD\*P are discussed. The 266nm radiation obtained by second harmonic generation of 532nm output of a Nd:YAG laser was used to pump a BBO OPO, described in Chapter Six. Experimental details of a 266nm pumped
BBO parametric oscillator are outlined and discussed in this next chapter. Osillation in the blue has been demonstrated using the configuration that incorporates a 266nm pump steering mirror within the OPO cavity.

Finally, in Chapter Seven, the progress and future research in harmonic generation and OPOs are considered, and conclusions drawn.
References to Chapter 1


