S.P.41. – MOOSAD, K.P.B. – Studies on some Nonlinea Optical Phenomena - Optical Phase Conjugation and Continuous wave second harmonic Generation – 1989 – Dr. V.P.N. Nampoori.

Nonlinear optice has emerged as a new area of physics following the development of various types of lasers. A number of advancements, both theoretical and experimental, have been made in the past two decades, by scientists all over the world. However, only few scientists have attempted to study the experimental aspects of nonlinear optical phenomena in Indian laboratories. This thesis is the report of an attempt made in this direction.

The thesis contains the details of the several investigations which the author has carried out in the past few years. On optical phase conjugation (OPC) and continuous wave (CW) second harmonic generation (SHG). OPC is a new branch of nonlinear optics, developed only in the past decade. The author has done a few experiments on low power OPC in dye molecules held in solid matrices, by making use of a Degenerate Four Wave Mixing (DFWM) scheme. These samples have been characterised by studies on their absorption-spectra, fluorescence spectra, triplet lifetimes and saturation intensities. Phase conjugation efficiencies with respect to the various parameters have been investigated. DFWM scheme was also employed in achieving phase conjugation of a broadband laser (Nd: Glass) using a dye solution as the nonlinear medium.

SHG is one of the most important of the several second order nonlinear optical effects, in view of its apllications. Although SHG is common with pulsed lasers, it is not so in the case of CW lasers. The author has developed an indigenous system for SHG in a ring dye laser, making use of this Lithium lodate crystals. The tunable coherent ultraviolet beam thus obtained was successfully applied for spectroscopic investigations.

The thesis has been set in six chapters and two appendices. The FIRST CHAPTER is a general introduction to nonlinear optics. Various nonlinear optical effects due to the second and third order nonlinear susceptibilities are discussed. The derivation of an expression for nonlinear optical susceptibility is outlined.

CHAPTER TWO is an account of DFWM and OPC. The throries of DFWM in transparent and absorbing media are examined.

CHAPTER THREE contains the details of the experimental investigations made on a few low power saturable absorbers as candidates for low power OPC. The specific cases studied are the dyes eosin, erythrosin B and Rose Bengal, taken in thin films of gelatin, polyvinyl alcohol (PVA) and boric acid glass. The properties of these systems in relation to their suitability for OPCare experimentally investigated. which include the absorption spectra, flourescence spectra, triplet-life times, and saturation intensities. The absorption spectra indicated that the boric acid glass samples do not absorb strongly at 514.5 nm wavelength while in the other two films it does, indicating that the boric acid glass samples cannot be used for working at the wavelength 514.5 nm of the argon ion laser. The time evolution of the fluorescence emission is shown to be a quick check for saturation in absorption in these films. However, the actual determination of the saturation intensities were done by plotting the transmission vs. incident intensity curves. It was found that the transmission increased indefinitely, suggesting that irreversible damage is occuring to the dye molecules. Saturation intensities were therefore measured by repeatedly plotting the transmission vs. incident intensity curves and making sure in each case that no irreversible changes had occured. These meaurements were supported by measurements of the triplet lifetimes also. A few studies of the fluorescence of these dyes in the above matrices, which, though interesting, are not directly related to the OPC studies, are described in the Appendix I.

SECTION I OF THE CHAPTER FOUR is the details of the studies of OPC in the above samples. Phase conjugation efficiencies versus the input powers, absorption lengths and wavelengths are studied and compared with the theoretical models discussed in the previous chapters. Of three dyes used, erythrosin B showed the highest efficiency, both in gelatin and PVA films. Efficiencies were always greater in PVA films than in gelatin films. The best efficiency achieved was about 10¹⁻⁵, for erythrosin B in PVA film. Efficiency varied with the input power - peaked around the saturation intensity and then decreased. This was not only due the inherent behaviour of the interaction but also due to the laser-induced damage of the dye molecules. This damage is found to occur very fast. Efficiency varies with the absorption legnth also. It was found that there exists on optimum for this. Another aspect affecting the efficiency is the "wash-out" effects due to vibration pick-ups. This also was found to be very severe.

In the SECTION II OF THE CHAPTER FOUR, OPC experiments using a Nd: Glass laser are described. An absorbing dye Kodak 14015, dissolved in 1,2, dichloroethane is used as the nonlinear medium, in a simple DFWM scheme, here are only few reports of OPC via DFWM of broadband radiations. Actually, there are no reports at all, where OPC of Nd:Glass laser has been achieved by a DFWM technique. The present investigations showed that it is possible to achieve this.

CHAPTER FIVE presents the details of the attempts made on developing an indigenous system for CW SHG, using Lithium lodate Crystals. Two types of crystals were used - "perpendicular-cut" and "Brewster-cut". The best output power obtained was about 1.85 milliwatts. Tunability of a few nanometers was possible. This beam was used in an experiment for detection of OH radicals in air contained in a multi-pass cell. The details of this experiment are also reported briefly in this chapter.

CHAPTER SIX is a summary of the main features of the work.

APPENDIX I presents the details of the studies on fluorescence spectra of the samples used for low power OPC. APPENDIX II is a pedagogical aspect of OPC. It is suggested that this method of obtaining an OPC beam is ideal for introducing OPC as laboratory topics in post-graduate courses. Details are discussed, alongwith suitable experimental evidences.

Most of the results reported in the thesis have been published elsewhere or submitted for publication.