CHAPTER 6

SUMMARY AND SUGGESTIONS FOR FUTURE WORK

6.1 SUMMARY

Solid-state lasers emitting in the deep UV offer a safe and effective alternative for the bulky excimer lasers. CBO is an important nonlinear optical crystal for third-harmonic generation of the Nd:YAG laser and for high-power UV generation because of its relatively high tolerance to laser damage threshold, large nonlinear optical coefficients and an adequate transparency in the UV region. CBO had an UV absorption edge shifts near 170 nm and so it can be used for deep UV light generation. CBO crystals can be grown from self-flux solutions using TSSG technique.

Large size crystals were grown from 74 mol% B$_2$O$_3$ solutions at 830°C using a-axis seeds. A rotation rate of 60 rpm and cooling rate of 0.1 °C/day were used for the growth of CBO crystals. After the growth, the crystals were cooled to room temperature in 48 h. The main problems with CBO crystals are evaporation of the cesium component during growth leading to spontaneous nucleations and high viscosity of the CBO solution and the presence of scattering centers. The spontaneous nucleations occur because of the high supersaturation induced by heavy evaporation at the edges of the crucible. The seed dissolution was attributed to harmful effect of the evaporated vapours. By placing a cover over the crucible the evaporation of cesium from the edges of the crucible was controlled and the spontaneous nucleations were suppressed. During the growth, the seed dissolution was
reduced by slightly dipping the seed into the solution and covering the exposed part of the seed with a platinum foil. By these changes in growth, a bulk crystal of size 50 x 45 x 45 mm$^3$ ($a \times b \times c$) and weighing 159 g has been grown.

Scattering centers in CBO crystals are responsible for the optical loss and reduction in the third harmonic efficiency. The crystal had scattering centers when they were illuminated with a He–Ne laser of 4 mW. From our investigations it was found that the presence of scattering centers did not depend on the purity of the raw material and on the growth rate of the crystals. It was found to depend on the temperature from which the crystals were grown. The crystals grown from 70 mol% B$_2$O$_3$ solutions at 790°C contained no visible scattering centers. But the growth of large and device quality crystals from 70 mol% B$_2$O$_3$ solutions was very difficult because of heavy evaporation. As the crystals grown from both 74 and 70 mol% B$_2$O$_3$ solutions differed in the temperatures at which the crystals were grown, we considered the effect of temperature to be a likely reason for the presence of scattering centers.

In order to check the effect of growth temperature on the presence of scattering centers, the weight loss of 20 mg powdered crystals was measured at different temperatures in air using thermo-gravimetry. The weight loss experiments showed that low growth temperature and the presence of a highly-evaporative solution are essential to obtain scatter-free crystals. During the slow cooling of the crystals, additional phase precipitation occurring near the critical 650°C region could be the reason for the scattering in CBO crystals. Although the scattering centers have been reduced by post-growth quenching of the crystals, it produced a green luminescence when illuminated with a deep UV laser. The green luminescence is caused by the cesium vacancies that are created by the
dissolution of scatters in the solid solution at high temperatures. To remove the scattering and green luminescence, the off-stoichiometric as-grown crystals were brought to the stoichiometric composition by the VTE process in the presence of cesium-rich solution and the scattering centers were reduced. By our hypothesis of retrograde solid solution curve we are able to explain the mechanism of scattering centers and green luminescence formation in CBO crystals.

Though quenching and VTE experiments were successful in reducing the scattering centers, the LIDT of these crystals were low than as-grown crystals. So an alternative approach was adopted and after growth the crystals were fast-cooled from about 830 to 500°C. Different crystals were grown by changing the fast cooling in 1h, 3h and 6h (The cooling rate: 330°C/h, 110°C/h and 55°C/h respectively) to shorten the time exposed in the state of the high temperature. The strength of the scattering centers decreased whereas the bulk LIDT of the fast-cooled crystals increased as the cooling rate was increased. CBO has a deliquescence property and the surface of the crystals gets degraded when exposed to moisture content in the atmosphere. To reduce the surface degradation, the polished crystals were transferred to a stainless steel chamber purged with Ar gas. We optimised the conditions for maintaining the degradation free surface after irradiation with a laser for practical application. The crystals were irradiated with a third harmonic (355 nm) of the Nd:YAG laser. The frequency of the laser was 10 Hz and the pulse width was 7 ns. The processed a-axis crystals were irradiated with the third harmonic beam at 300 mJ. After this processing the surface of the CBO crystal has been maintained for laser applications and this process can be applied to other NLO crystals.

The second harmonic (SHG) and third harmonic generation (THG) studies of CBO crystals. The CBO crystals were cut for type II generation and
the SHG was generated from 3, 6 and 12 mm crystals. The CBO crystal was cut for type II THG and a high power of 100 W and 150 W was generated. Thus CBO crystals can be used for high power SHG and UV generation. The summary of the present investigations and the suggestions for future research activities in CBO crystal are also presented.

6.2 SUGGESTIONS FOR FUTURE WORK

The probability that a new borate structure lacks a centre of symmetry is more than twice as higher than for non-borate structures. So work can be dedicated towards the investigation of new NLO crystals with excellent optical properties for UV generation.

Attempts can be made in growing large size CBO crystals and towards the growth of stoichiometric CBO crystals. More studies can be done towards the understanding of the scattering phenomenon in CBO crystals. It will be helpful in eliminating the scattering centers in CBO.

Future works can be directed towards the fabrication of NLO devices for enhanced conversion efficiency. CBO crystal can be useful for deep UV generation by the 8th harmonic generation of the Nd:YAG laser. So experiments can be conducted in generating more powerful and deep UV laser light.

The surface degradation of CBO during laser experiments has to be fully resolved inorder for it to be useful for long term practical applications. Suitable dopants in CBO crystal, which may enhance the optical properties can be found.