Development of nonlinear crystals and blue SHG laser

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HMRL, Hitachi Metals, Ltd., Kumagaya, Saitama 360 Japan

Compact solid-state blue laser have great potential for use in optical data storage, laser beam printing, particle countering, reprographics, holography, and fluorescent bioanalysis. We report recent progresses in qualities of LiB_3O_5 and $K_3Li_2(Ta_xNb_{1-x})_5O_{15}$ nonlinear crystals which are essential in manufacturing bulk-type blue SHG devices. We also review newly developed violet-blue laser, 20 mW output, using intracavity frequency doubling of a diode laser pumped $Cr:LiSrAlF_6$ laser with low loss LiB_3O_5 crystal as a frequency doubler.

1. Introduction

All solid state lasers continue to make significant progress in a variety of optelectronic systems. Especially compact solid-state blue laser has great potential as a alternative of air-forced Ar or He-Cd lasers for the use in optical data storage, laser beam printing, particle countering, reprographics, holography, and fluorescent bioanalysis since these compact devices offer potential advantages in size, efficiency and operating lifetime.

We have been working on developing compact, near 400 nm blue laser light, laser-diode-based second harmonic generation (SHG) devices for replacing the existing Ar or He-Cd gas laser market by investigating both intracavity bulk type and quasi-phase matching (QPM) waveguide type structures. We also have developed nonlinear crystals such as LiB_3O_5, $K_3Li_2(Ta_xNb_{1-x})_5O_{15}$ $LiNbO_3$, $KNbO_3$, β–BaB_2O_4 for the use of our SHG laser configuration.

We have recently reported that 10 mW of violet-blue output at 427 nm was obtained with intracavity frequency doubling of a diode laser pumped $Cr:LiSAF_6$ laser by using LiB_3O_5 crystal as a frequency doubler. Selecting appropriate nonlinear crystal is one of the most important factor to make real products for manufacturing. The newly developed all solid state blue laser has advantages of long lifetime, small size, low power consumption, no forced-air cooling compared to the gas lasers.

In this paper we review the progresses in qualities of LiB_3O_5 and $K_3Li_2(Ta_xNb_{1-x})_5O_{15}$ nonlinear crystals and the development of blue SHG lasers.

2. Development of LiB_3O_5 single crystals

Lithium triborate (LiB_3O_5, LBO) crystals have been widely investigated for high power frequency conversions because of its critical and noncritical phase matching, wide transparency range and chemical stability. Although the nonlinear optical coefficients of LBO are relatively small when compared to $LiNbO_3$, $KNbO_3$, and $KTiOPO_4$, the phase matching characteristics , low losses and the high damage threshold make the material an appealing choice in SHG or OPO applications [1].

Crystal growth studies of LBO are in the beginning stages, and there are many interesting problems to solve. Flux growth studies from B_2O_3 excess melt is the most likely technique to yield good crystals. However, crystal growth from a glass-forming melt is often difficult, since the high viscosity limits the mixing and mass transport in the melt. We studied the top seeded solution growth of LBO from an excess B_2O_3 solution. The parameter investigated included the Li_2O/B_2O_3 ratio, the rotation, pulling and cooling rates [2]. Many of the problems encountered, such as hopper growth and inclusions, can be attributed to the high viscosity of the solution, and methods of increasing the forced convection was examined by the melt simulation with the tracer method as shown in Fig. 1. Clear crystals of approximately 42*48*30 mm have been grown as shown in Fig. 2.

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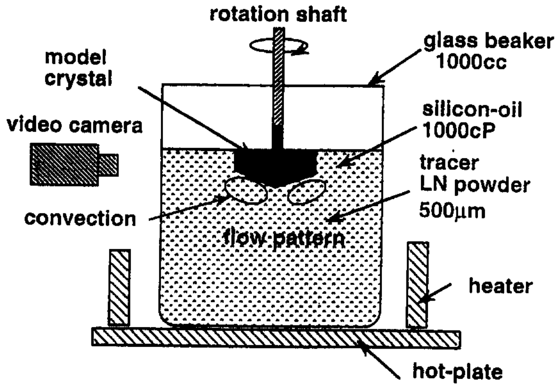

Fig. 1 Experimental set up of the melt simulation using the tracer method.

Fig. 2 LBO crystal grown by TSSG method.

3. Development of $K_3Li_2(Ta_xNb_{1-x})_5O_{15}$ single crystals

Single crystals of $K_3Li_2Nb_5O_{15}$ (KLN) crystals are expected to be the best materials for blue laser frequency conversions, because they have advantages in high damage resistance, low losses, wide noncritical phase matching characteristics compared with KN crystals. Recently Yoon et al. have succeeded the crystal growth of homogeneous KLN crystals by the micro pulling down method [3], and reported SHG properties. As grown KLN crystals are pale yellow color, however, this limits the usability of these crystals for high power blue SHG applications, since materials absorb generated blue laser light and give rise to the instability of crystal temperature.

On the other hand, Fukuda has been reported that substituting of Ta ions for Nb in KLN could decrease the coloration [4]. We have followed that report, and have investigated the crystal growth of $K_3Li_2(Ta_xNb_{1-x})_5O_{15}$ (KLTN) since the optical properties of KLTN such as transparency, SHG characters have not been reported yet.

Single crystals of KLTN were grown by the Kyropoulos technique and by the conventional czochralski technique as used in the preparation of KLN. Starting materials were 5N purity K_2CO_3, Li_2CO_3, Ta_2O_5, and Nb_2O_5 powders and were mixed in non-stoichiometric proportion, with excess K_2CO_3 and Li_2CO_3. Single crystals were grown over the melt composition range from x= 0 to 1 at a constant ratio K/Li=3/2. Fig. 3 shows the $K_3Li_2(Ta_xNb_{1-x})_5O_{15}$ (x=0.8) single crystals with 10 mm in diameter and 45 mm in length. Figure 4 shows the optical transmission spectra of both KLN and KLT crystals. The transmission spectra were not calibrated for reflection loss. Comparing the transparency of KLN with that of KLT, KLN crystals are pale yellow in color and show broad decrease of transparency from 400 to 500 nm near the absorption edge and have large absorption coefficient, $\alpha = 2.1\ cm^{-1}$ at 400 nm. On the other hand, substitution of Ta ion improves the transparency in blue light region and changed crystals to colorless, clear, and transparent. KLT crystal, end crystal in KLTN system, exhibit a shift of absorption edge toward shorter wavelength and its calculated absorption coefficient a at 400 nm of KLT is about $0.3\ cm^{-1}$.

Fig. 3 $K_3Li_2(Ta_xNb_{1-x})_5O_{15}$ (x=0.8) single crystals grown by do·ble crucible czochlalski method.

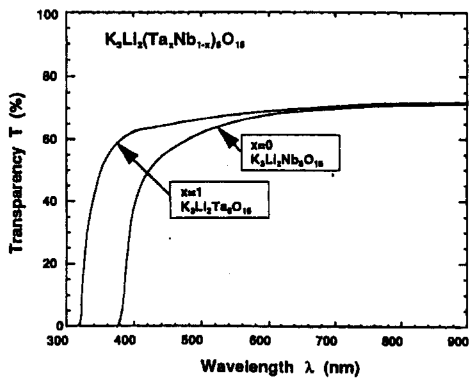

Fig. 4 Optical transmission spectra.

4. All solid-state blue SHG laser

Frequency doubling of a near i.r. laser is one way to generate blue light. We report on the generation of 20 mW of cw blue (430 nm) output power by intracavity frequency doubling of a diode-pumped $Cr:LiSrAlF_6$ laser. $Cr:LiSrAlF_6$ is an interesting new laser material because it can be laser diode pumped and oscillates in the near i.r. region, from 780 to 1000 nm [5]. In addition, because the absorption bands of the $Cr:LiSrAlF_6$ crystal are approximately 100-nm-wide, it is reasonably insensitive to the fluctuation of the diode laser frequency. We investigated the effect of changing the bandwidth of the fundamental laser radiation generated by the $Cr:LiSrAlF_6$ laser [6]. Using this bandwidth-controlled $Cr:LiSrAlF_6$ laser, we characterized several nonlinear crystals and cavity configurations for the $Cr:LiSrAlF_6$-SHG laser.

FIG. 5 shows the configuration of all solid-state blue laser. Birefringent filters (BF) were used as band selectors for the $Cr:LiSrAlF_6$ laser. Nonlinear crystals $KNbO_3$,β-BaB_2O_4, and LiB_3O_5 crystals were investigated for the $Cr:LiSrAlF_6$-SHG laser. Each nonlinear crystal was 5 mm in length, cut for 860 nm TYPE I phase matching at room temperature, and AR coated for 860 and 430 nm. The conversion efficiency was 0.54 %/W for KN and 0.003 %/W for LBO when the fundamental beam diameter was 60 μm. The spectral bandwidth of the nonlinear crystal is an important parameter for the LiSAF-SHG laser. Higher blue output power was obtained when the fundamental bandwidth was wider than the spectral bandwidth of the nonlinear crystal. 20 mW of blue output power at 430 nm was obtained with intracavity frequency doubling of a diode laser pumped $Cr:LiSrAlF_6$ from 400 mW incident power. Fig. 6 shows the all solid state violet-blue laser module, 50 *60*150 mm^3 in size, 60 W power consumption,<2%p-p power stability. Newly developed all solid state blue laser has advantages of long lifetime, small size, low power consumption, no forced-air cooling compared to gas lasers.

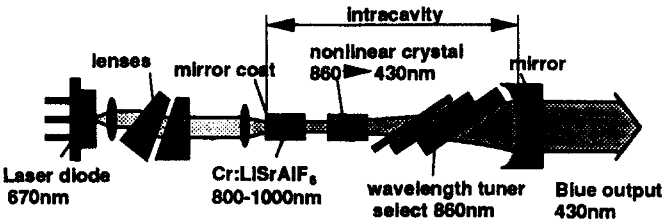

Fig. 5 Configuration of all solid-state violet-blue laser.

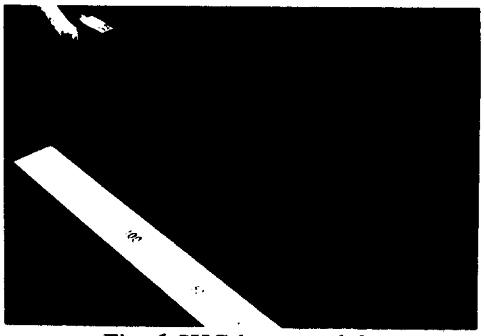

Fig. 6 SHG laser module.

5. Summary

We have reviewed recent progresses in qualities of LBO and KLTN crystals and blue lasers developed in Hitachi Metals. With careful control of the parameters, clear LBO crystals of approximately 42*48*30 mm have been successfully grown by the TSSG method. KLTN crystal has high potential as a nonlinear crystal for blue SHG applications. Ta substitution for Nb site in KLN crystal improves the transparency spectra especially in a visible wavelength region, decreases absorption coefficient , changes its color to clear and transparent from pale yellow, and shifts the absorption edge. We also review newly developed violet-blue laser, 20 mW output, using intracavity frequency doubling of a diode laser pumped $Cr:LiSrAlF_6$ laser with low loss LiB_3O_5 crystal as a frequency doubler.

6. References

1. C. T. Chen, et al., J. Opt.Soc.Am. B6 (1989) 616
2. S. A. Markgraf, et al., J. Cryst. Growth 140 (1994) 343
3. D. H. Yoon, et al., Jpn. J. Appl. Phys. 33 (1994) 3510
4. T. Fukuda, Jpn. J. Appl. Phys. 9 (1970) 599
5. S.A. Payne,et al., J. Appl. Phys. 66, 1051 (1989)
6. T. Miyai, et al., Appl. Phys. Lett. in press.

Development of nonlinear optical crystals and blue SHG laser

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National Institute for Research in Inorganic Materials

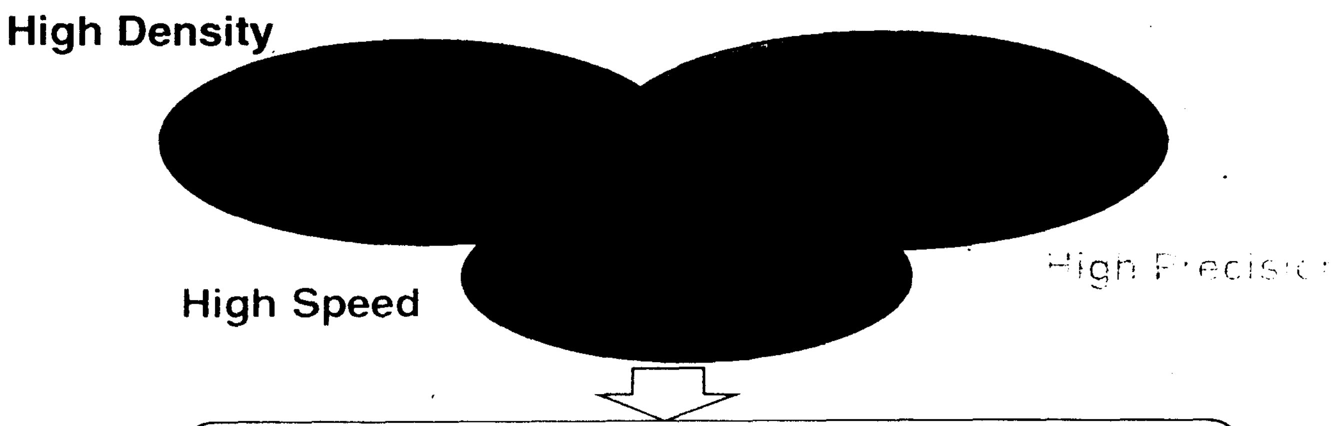
OUTLINE

1. Introduction

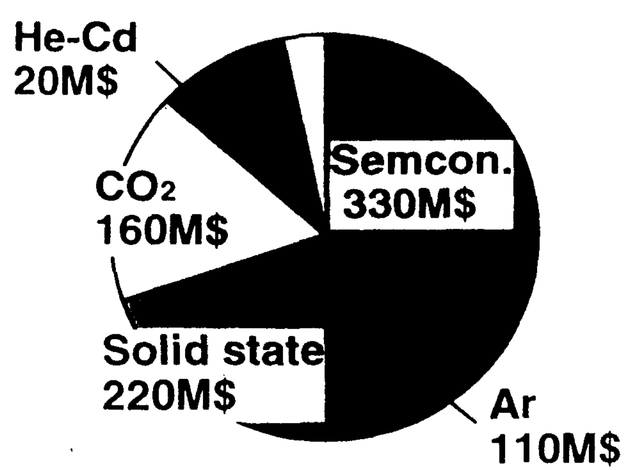
   Why blue laser?

2. Newly developed blue SHG laser
3. Development of nonlinear crystals
4. Summary

Why compact Blue Laser?



Requirements for shorter wavelength light source



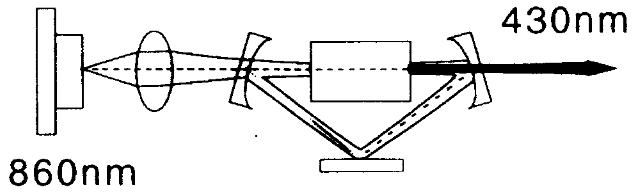
Laser Market 1088M$ (1995)

< Issues of gas lasers >

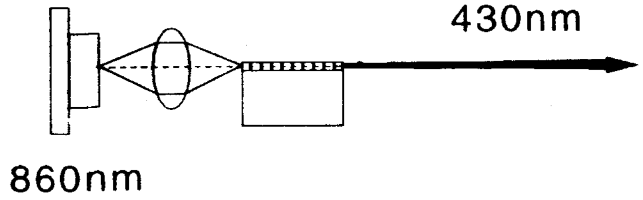
- short life time
- large size
- high power consumption
- forced air- or water cooling system
- high maintenance cost
- wavelength

Common Issue for Blue SHG Laser

External resonant diode doubling

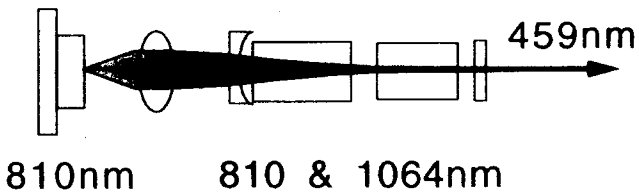


Direct diode SHG in periodically poled wavegides

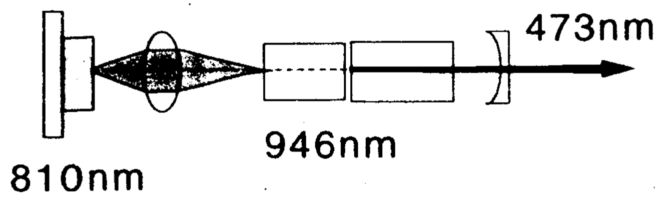


Critical for Laser Diode frequency

Signal-resonant sum frequency generation

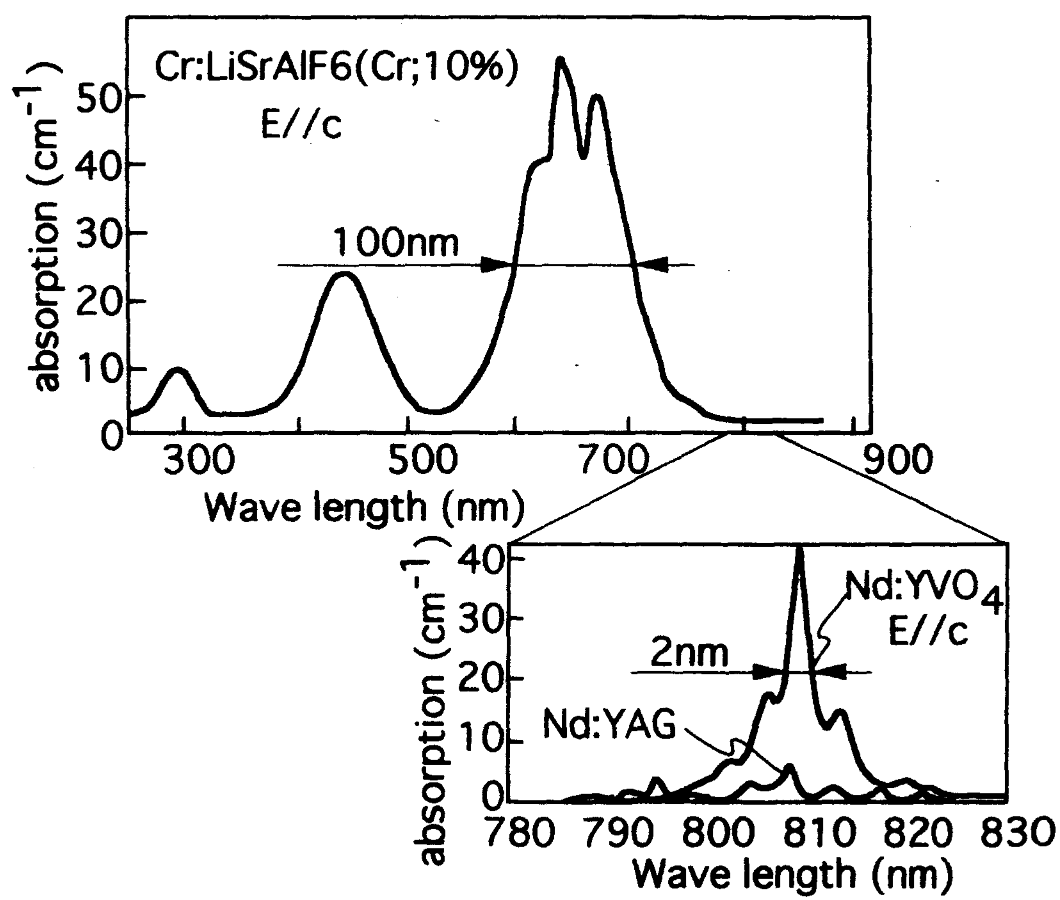


Intracavity SHG of YAG (946mm) laser



Hitachi Metals, LTD.

Laser crystals



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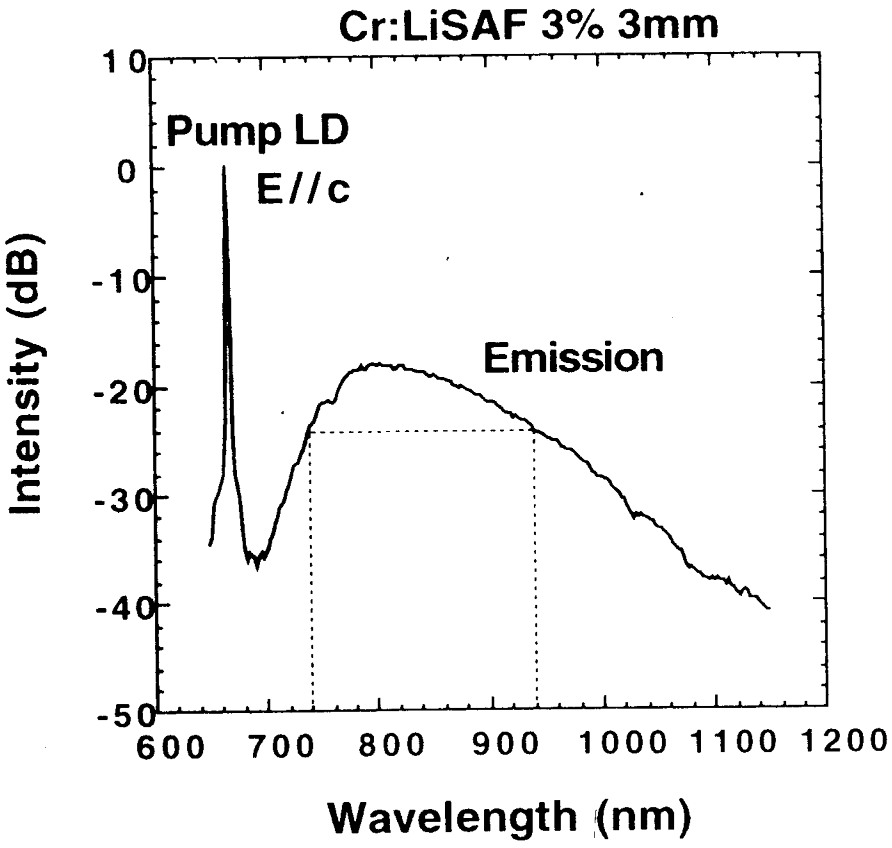
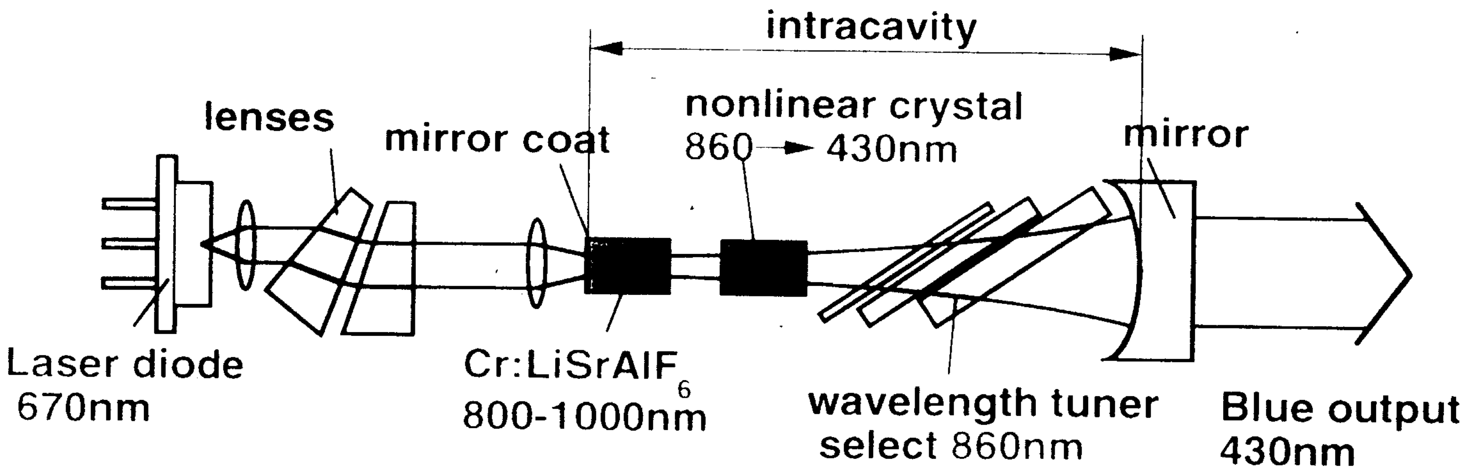
Characteristics of Cr:LiSAF crystal



- ◆tunning range
  750～990nm
- ◆cross section
  $4.8 \times 10^{-20} cm^2$
- ◆life time
  67μs

Hitachi Metals, LTD.

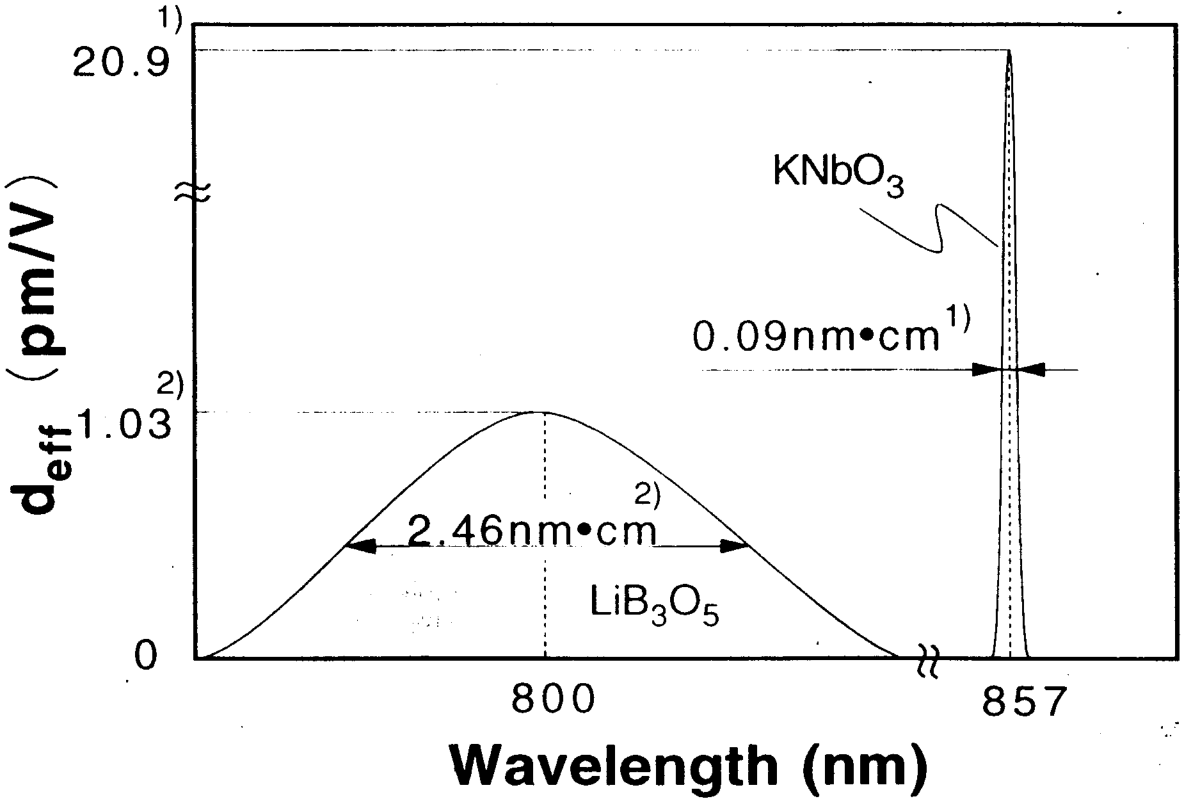
Configuration of our Blue SHG laser



< Features >

1. Noncritical in the frequency change of laser diode
2. Wide selectivity of blue output wavelength
3. Good reproducibility of laser module

Charactristic of Nonlinear Crystals



1) K. Asami, OPTRONICS, No.4-6 (1993)
2) A. Nebel, et al., Opt. Lett. 16, p1729 (1991)

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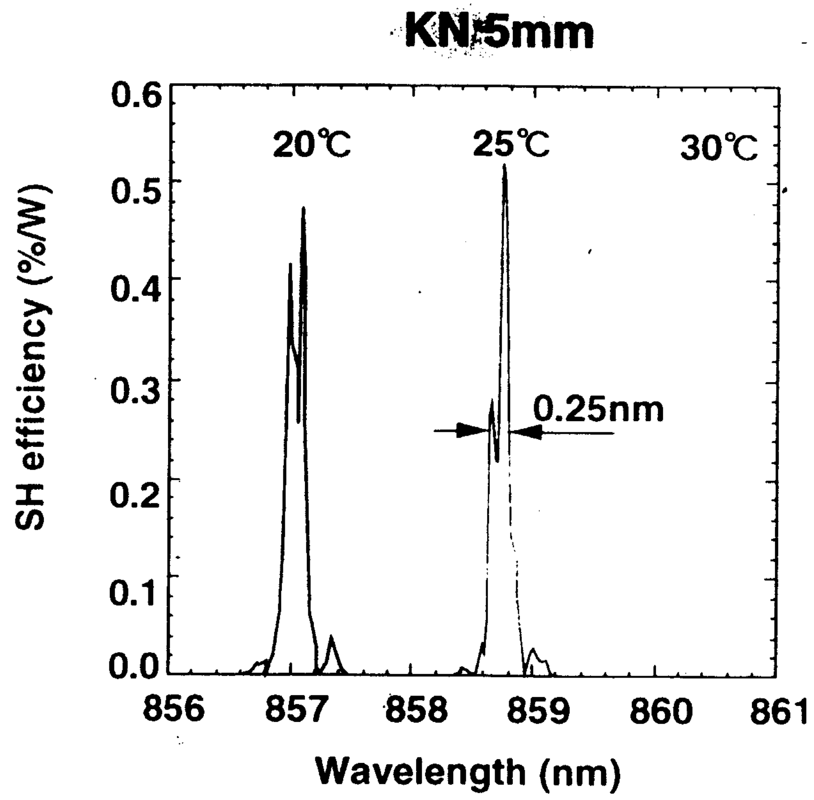
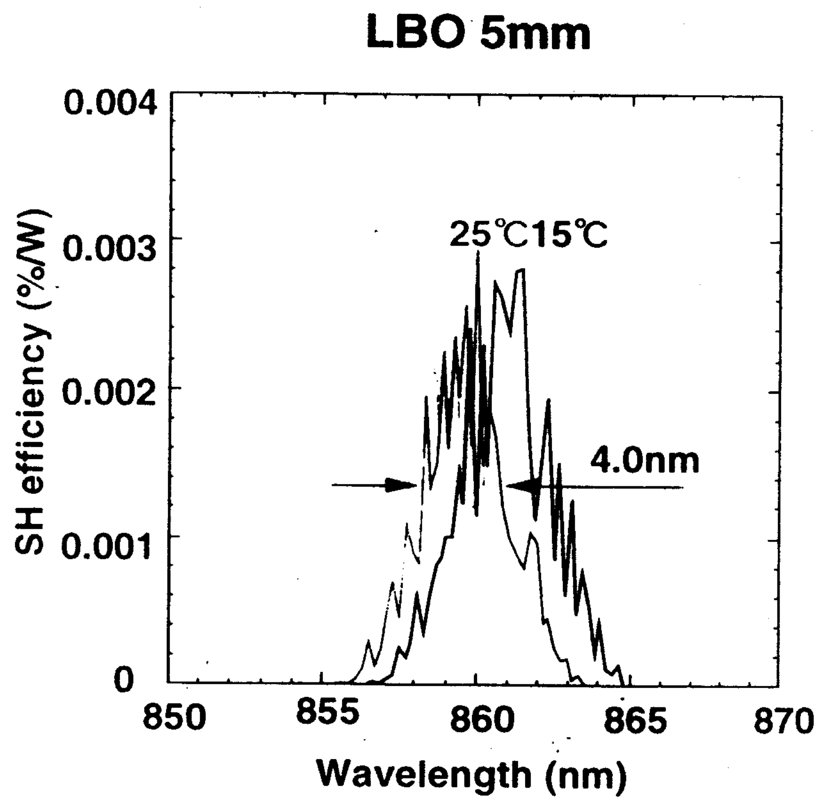
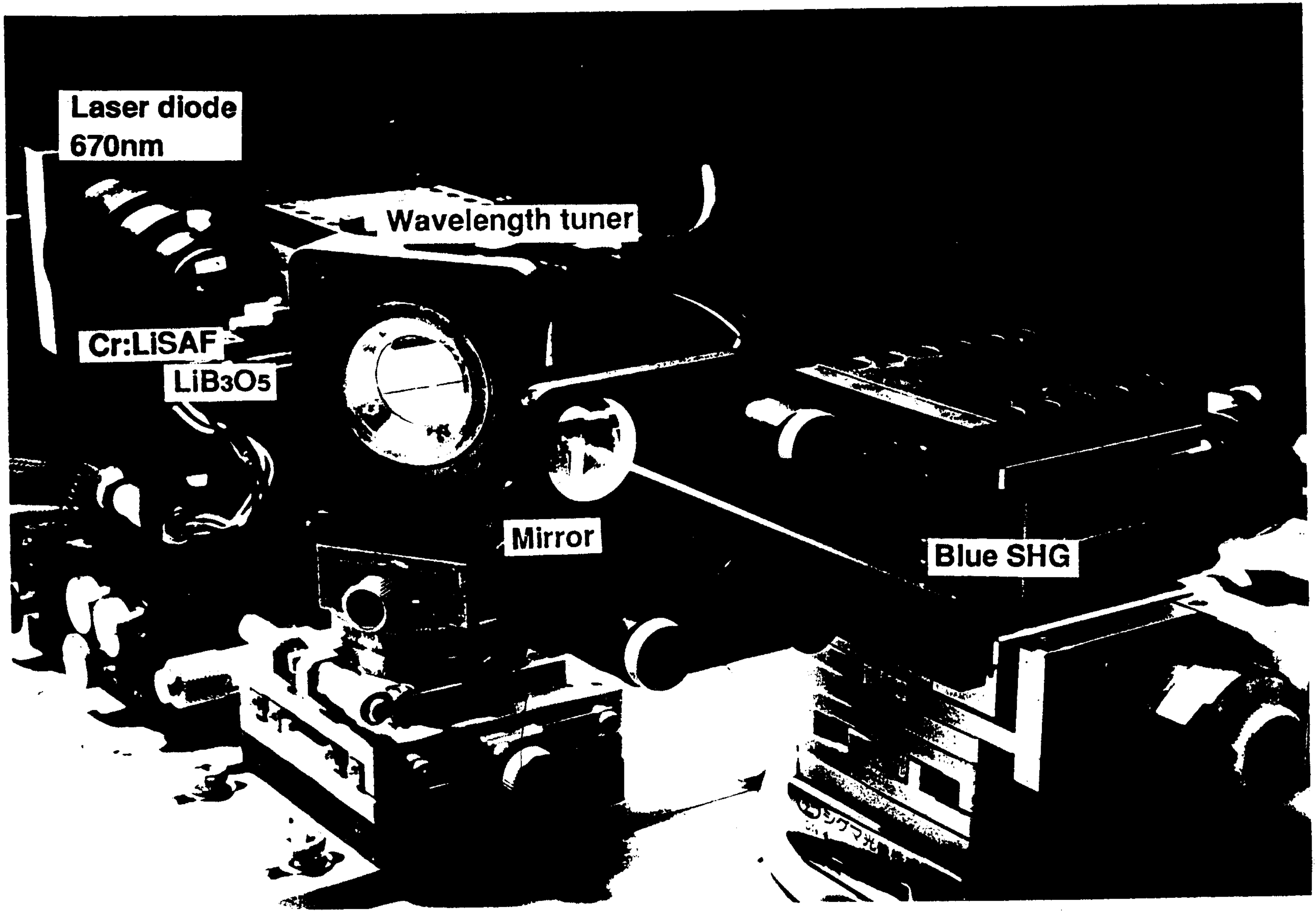
Temperature tolerance of SH phase-matching of KN and LBO



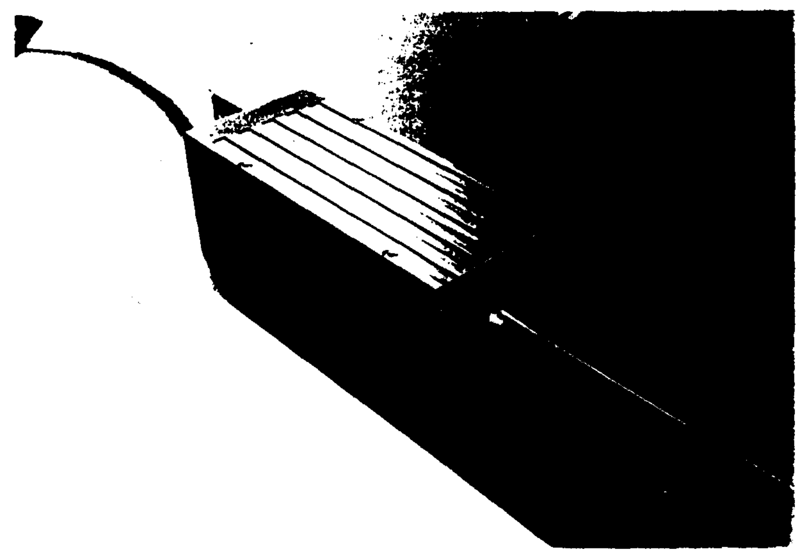


10mW of blue laser radiation by intracavity frequency doubling of a diode pumped Cr:LiSAF



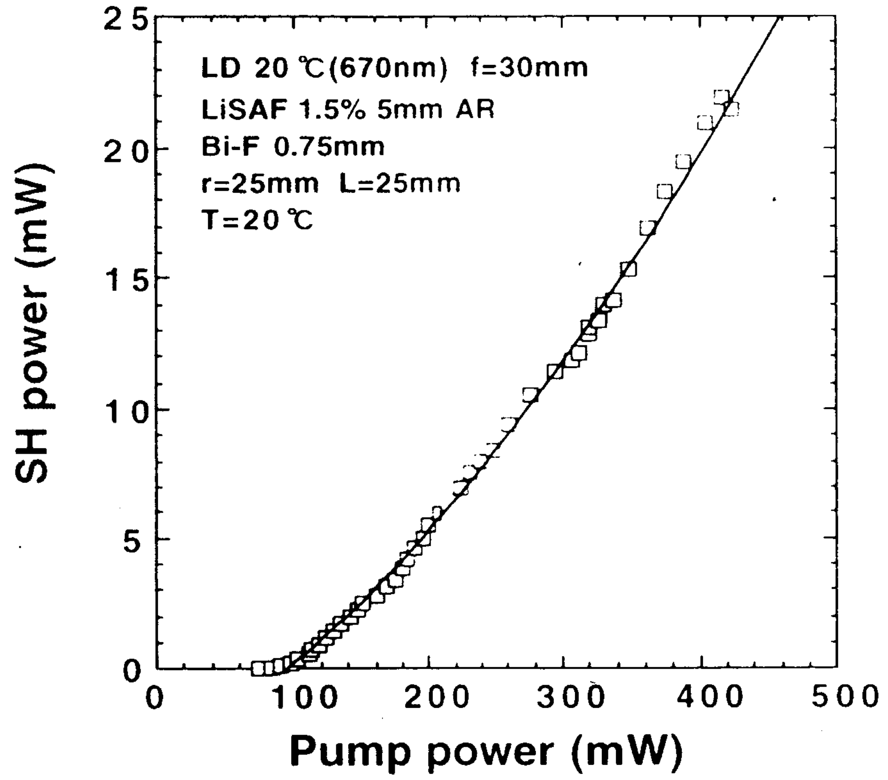
Characteristics of SHG Blue laser

- ◆ **Shorter wavelength**
  430nm
- ◆ **Exchange and adjustment free**
  long life time
- ◆ **Easy optical alignment**
  smaller size, 1/10 of gas laser
- ◆ **High reliability**
  no water-or forced air cooling
- ◆ **High beam quality**
  TEM_{00}

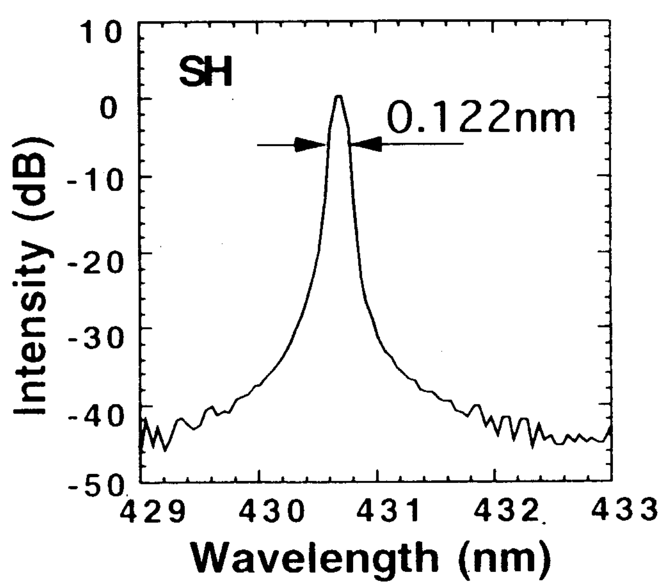


Hitachi Metals, LTD.

Blue output profile of SHG module



Spatial Profile : TEM00
Power Stability : <1%
Extinction Ratio : 650:1



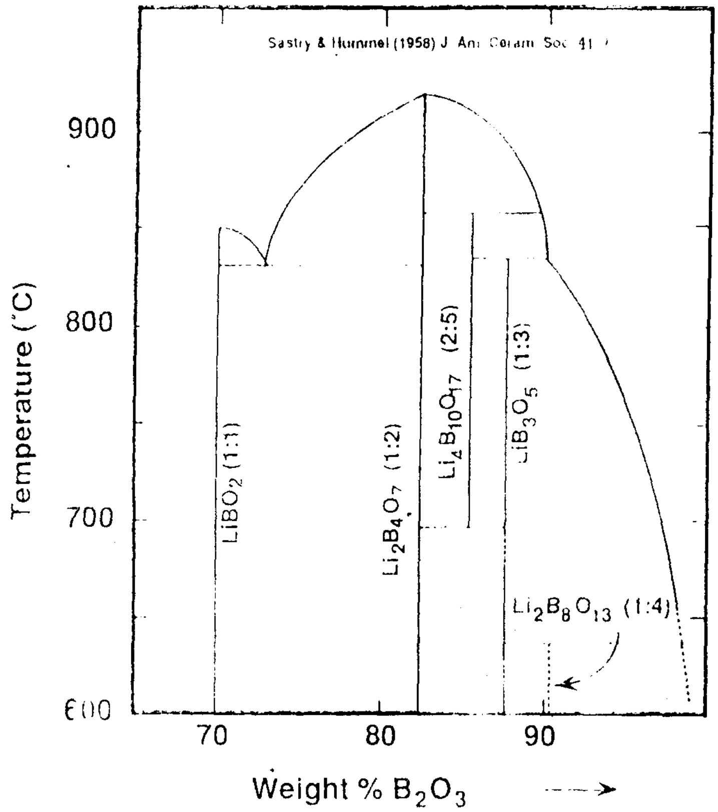
Hitachi Metals, LTD.

Nonlinear crystals for Blue SHG

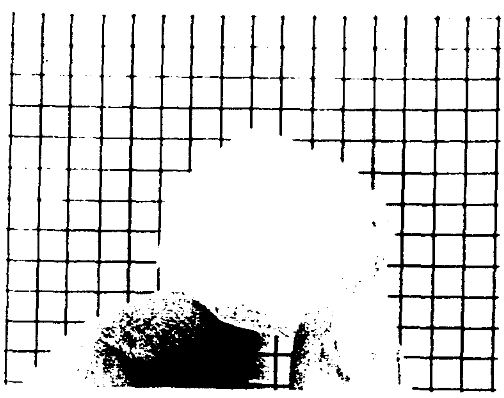
	$KNbO_3$	[redacted]	$LiTaO_3$ QPM	$K_3Li_2Nb_5O_{15}$
Nonlinar coeff. (pm/V)	21		25	14
Acceptance band width(nm ·cm)	0.09		0.15	0.3
Absorption Coeff. (%/cm)	1.5		0.5	0.5
Phase-matching wavelength (nm)	860		660-3000	800-3000

Hitachi Metals, LTD.

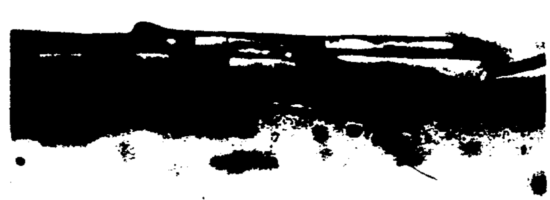
Issues in the LiB_3O_5 crystal growth



- LBO decompose at 830 °C
- high viscosity from B_2O_3 excess melts (1000 mPa sec)
- inclusion of flax, bubbles
- unstable growth (hopper-growth)
- cracks and spontaneous nucleation



hopper-growth



inclusions

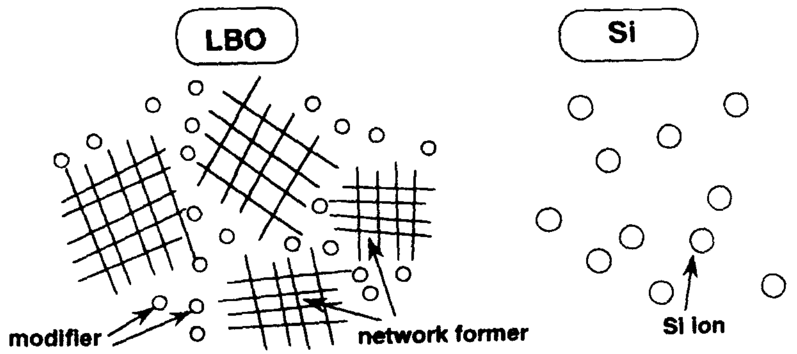
Crystal growth of LBO from high viscosity melt

< Key crystal growth technique >

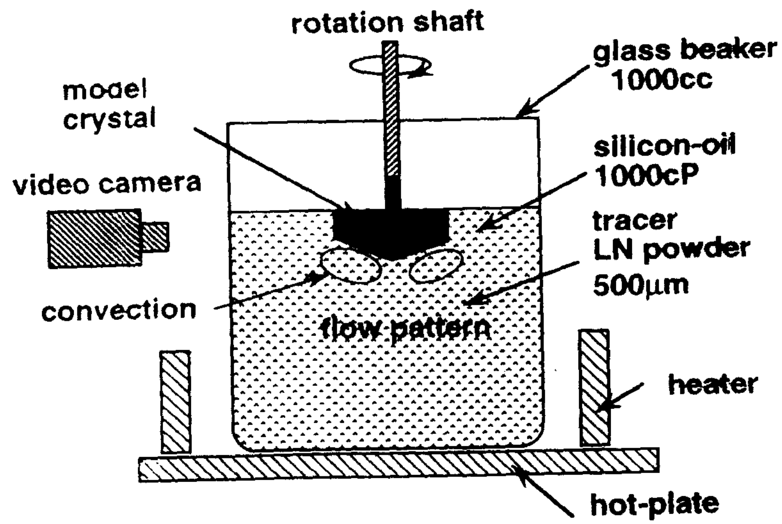
1. Melt temperature control
   · development of TSSG furnace

2. Melt convection control
   · melt simulation with tracer method

How is the melt structure of LBO?



very high viscosity : 1000 mPa sec	⇔	low viscosity : 0.7 mPa sec
high supersaturation low diffusion large boundary layer		low supersaturation high diffusion small boundary layer



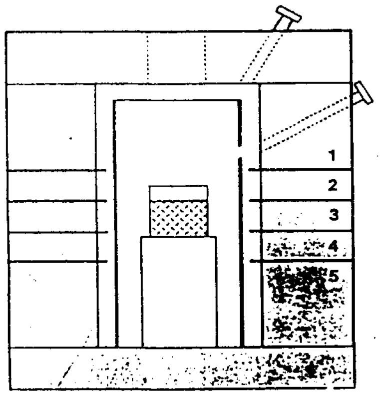
Features of developed TSSG furnace

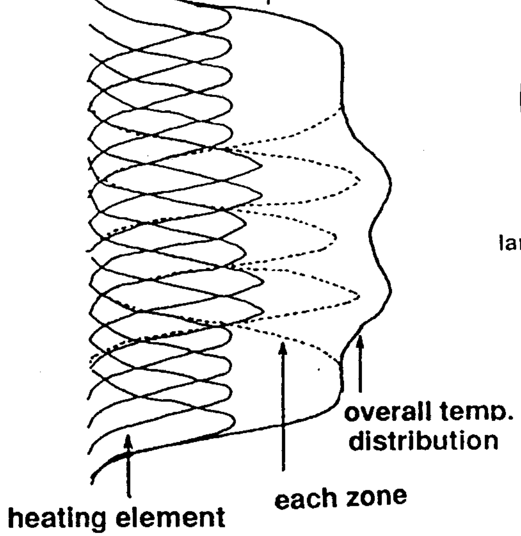
1. 5 zone independent heating
2. Automatic control for the temperature distribution of melt
3. Triple feedback loop for temperature control
   temperature stability <900 ± 0.05°C, setting 0.001°C

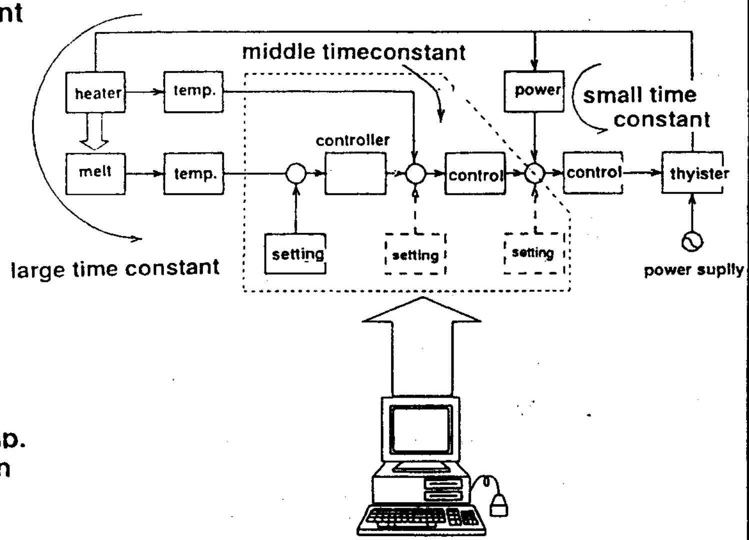
$$T(x) = \Sigma\, a_i\, H_i \exp\{-(x-x_i)^2/b_i\}$$

H_i: heater setting temp.
a_i: contibution
b_i: furnace constant

Kanthal Elements
Pt Crucible :
785 ml





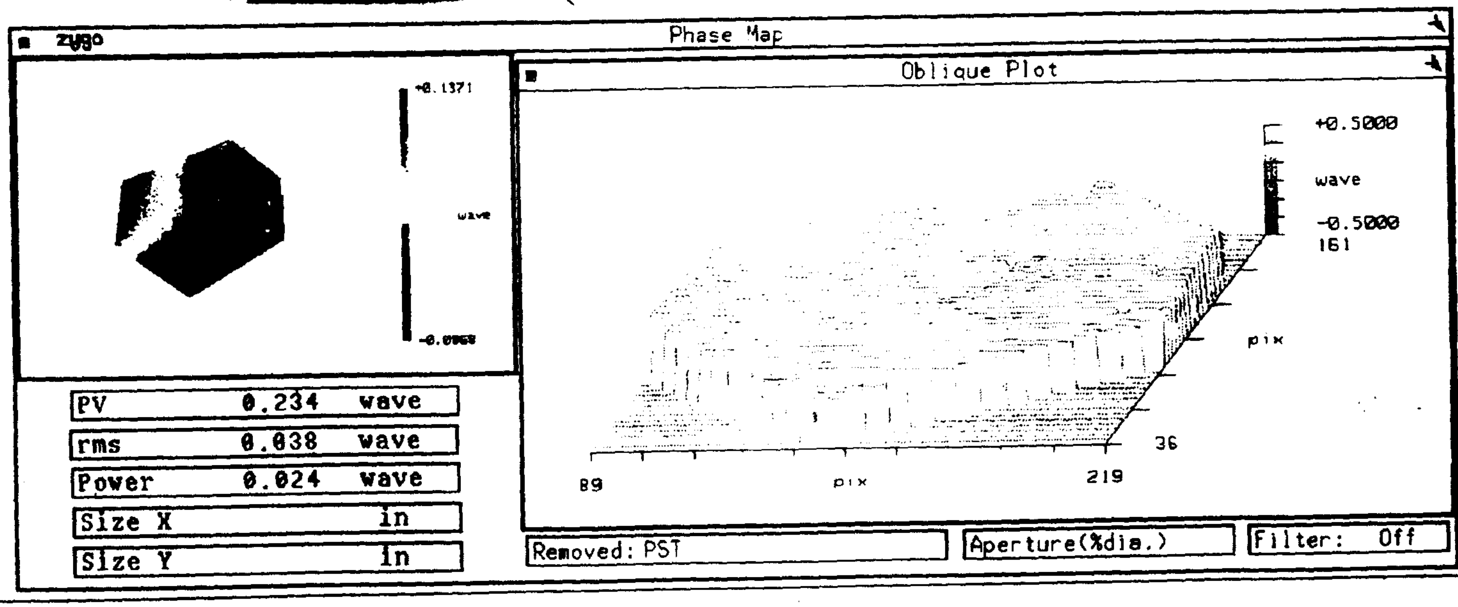


Low Loss LBO crystal grown by TSSG

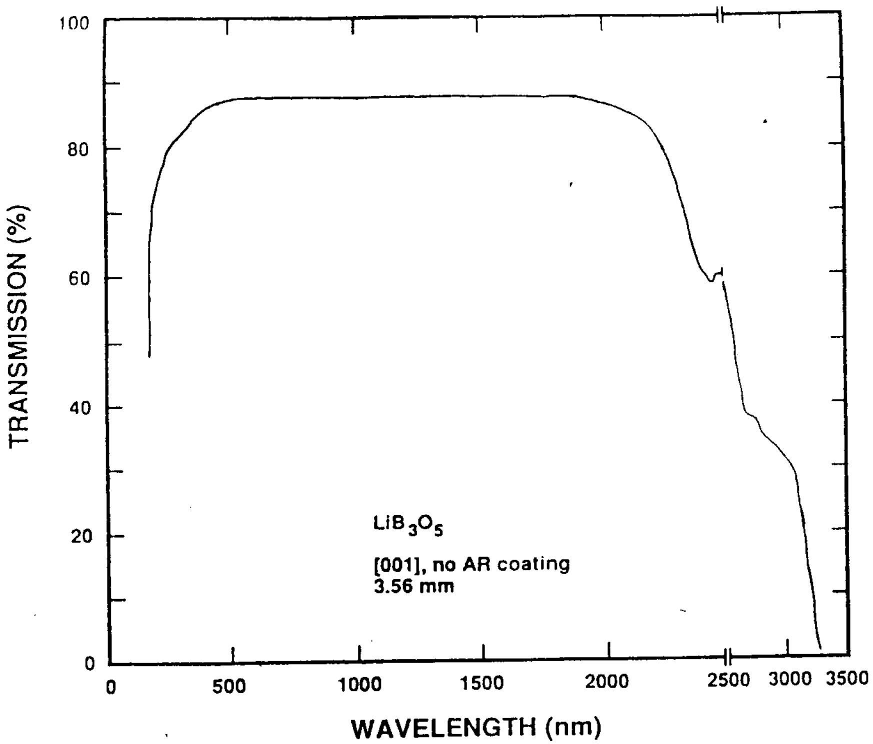
top view of <001> axis grown LBO   46 * 40 *34 mm^3

loss < 0.1%/cm

$\Delta n = 1x\ 10^{-6}$ / cm

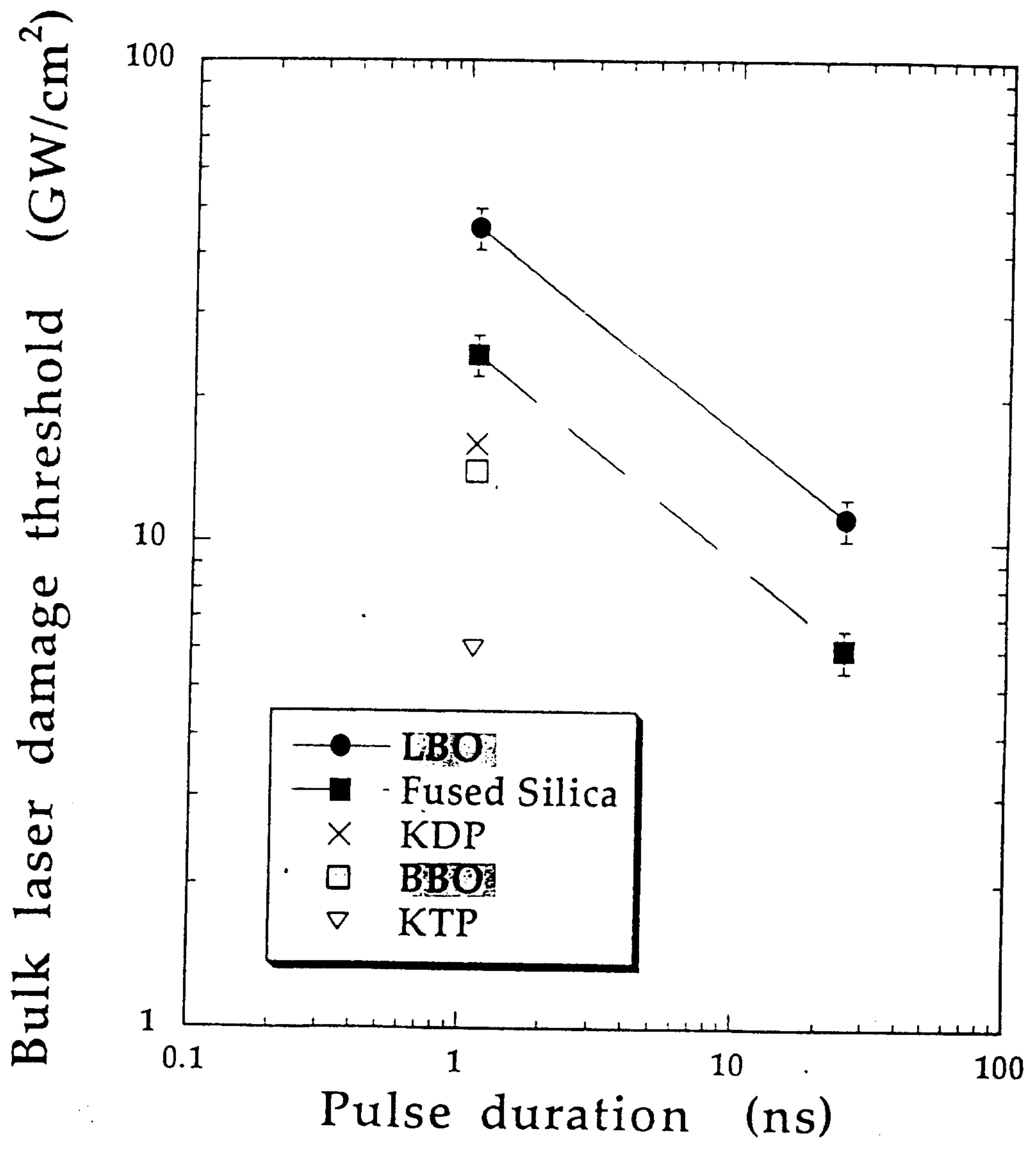


Transparency Spectra and Impurity of LBO



element	LBO	KTP
Na	< 1.0	8.3
Mg	< 0.3	3.0
Al	< 0.1	3.0
Si	< 1.0	25.0
Ca	< 1.0	1.0
Cr	< 0.1	6.9
Mn	< 0.05	< 0.4
Fe	< 0.1	< 6.9
Co	< 0.05	0.4
Ni	< 0.1	1.0
Cu	< 0.05	0.6
total	< 1.0	56.5

Bulk damage threshold of LBO at 1.064μm as a function of pulse dulation



Advantages and disadvantages of LBO for SHG

<Advantages>

- high transparency, low impurity
- high optical homogeneity
- high damage threshold

Bulk laser damage threshold of LBO with various laser irradiation directions, polarizations and positions in crystal.

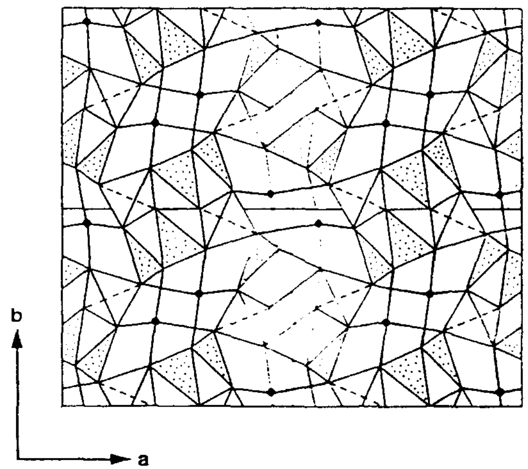
Laser input direction	Polarization direction	Damage threshold (GW/cm2)	Location
<001>	<010>	43.6	seed
<001>	<010>	42.7	center
<001>	<010>	44.7	bottom
<001>	<100>	42.3	seed
<001>	<110>	46.4	seed
<010>	<100>	45.0	seed
<010>	<001>	42.3	seed
<100>	<001>	47.7	center
<100>	<010>	43.0	center

<Disadvantages>

- no phasematching for YAG-FHG
- difficulty to change refractive index by doping other element

<Crystal Structure of LBO>

⊠ Space Group: $Pna2_1$, a = 6.50 Å, b = 7.38 Å, c = 5.15 Å, Z = 4



⊠ Lithium occupies tetrahedral sites inside the helices.

Structure is composed of BO_3 and BO_4 polyhedra (making B_3O_7 groups) in a spiral along [001].

reported by C. Chen, et al.
J. Opt. Soc. Am. B6,616(1989)

Nonlinear crystals for Blue SHG

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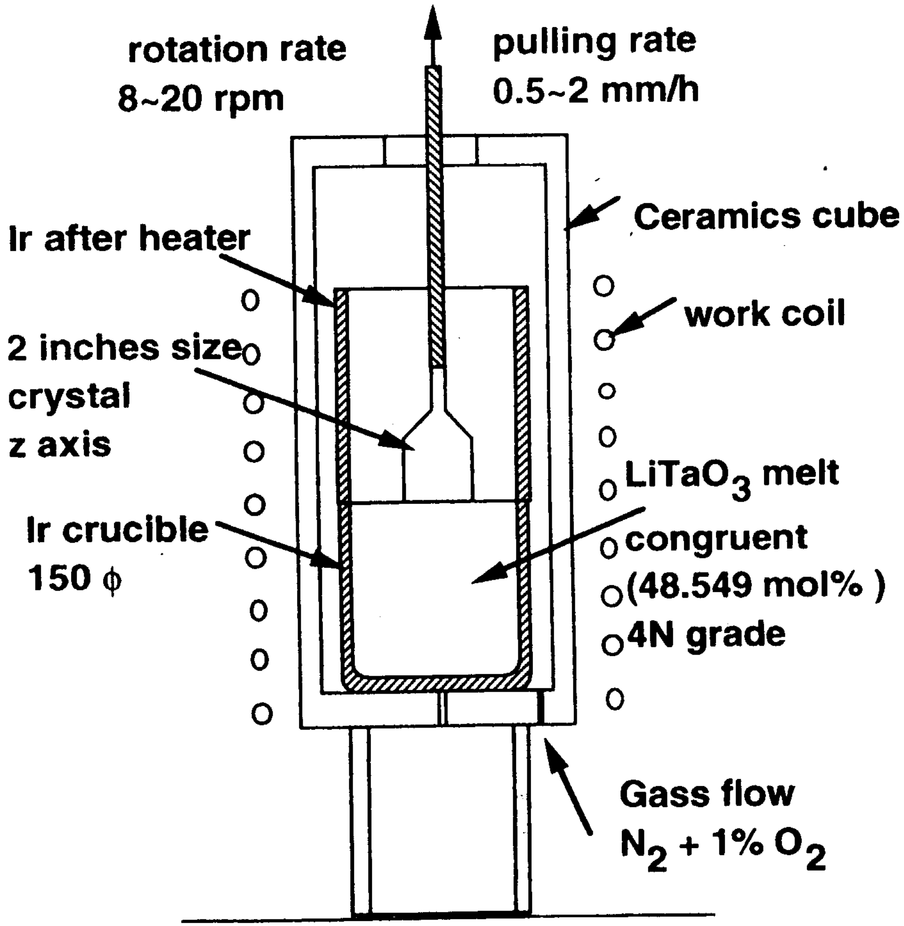
Nonlinar coeff. (pm/V)	21		25	14
Acceptance band width(nm ·cm)	0.09		0.15	0.3
Absorption Coeff. (%/cm)	1.5		0.5	0.5
Phase-matching wavelength (nm)	860		660-3000	800-3000

Hitachi Metals, LTD.

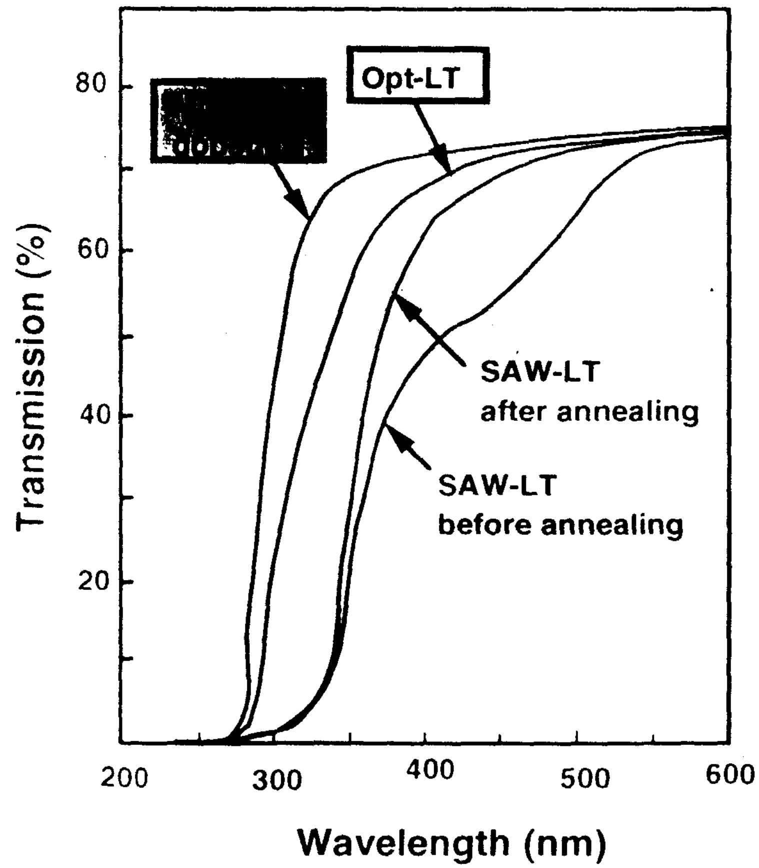
Crystal growth of optical grade $LiTaO_3$ -Z crystals

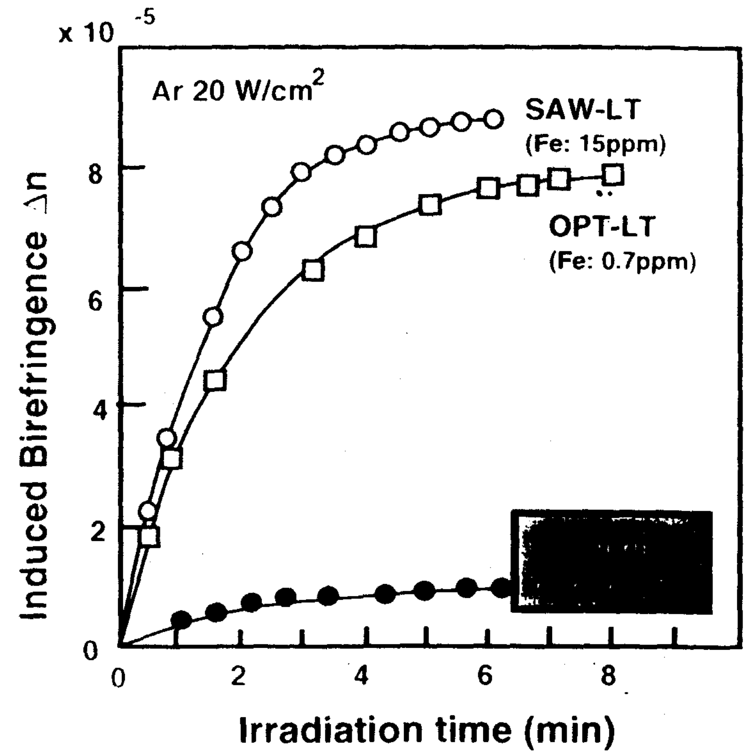


<Issue> <key point>
1.crack : growth interface
2.coloring : impurity,dopant
3.optical damage : anneal,dopant

2 "φ LT z-axis crystal

Improved transparency and optical damage resistance in LT

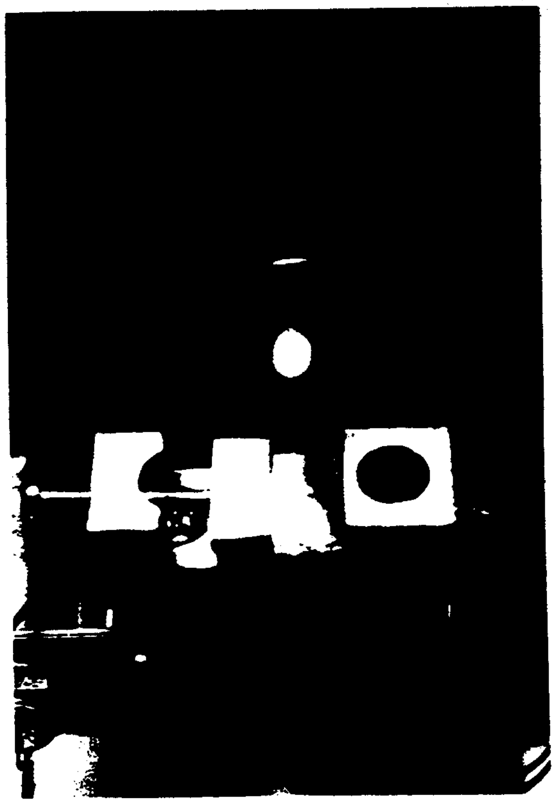




Blue quasi-phase matched harmonic generation by LT

<issues>
1. stability
2. reproducibility
3. thickness

Periodically inverted spikelike domains on LT-z
3.2μm period, 400~200μm depth



Direct blue SHG from LD

Nonlinear crystals for Blue SHG

	$KNbO_3$	[redacted]	$LiTaO_3$ QPM	$K_3Li_2Nb_5O_{15}$
Nonlinar coeff. (pm/V)	21	[redacted]	25	14
Acceptance band width(nm ·cm)	0.09	[redacted]	0.15	0.3
Absorption Coeff. (%/cm)	1.5	[redacted]	0.5	0.5
Phase-matching wavelength (nm)	860	[redacted]	660-3000	800-3000

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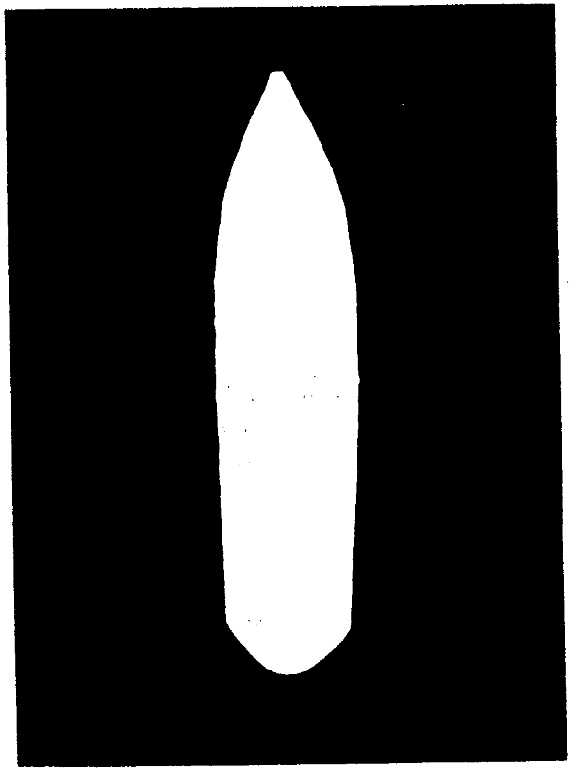
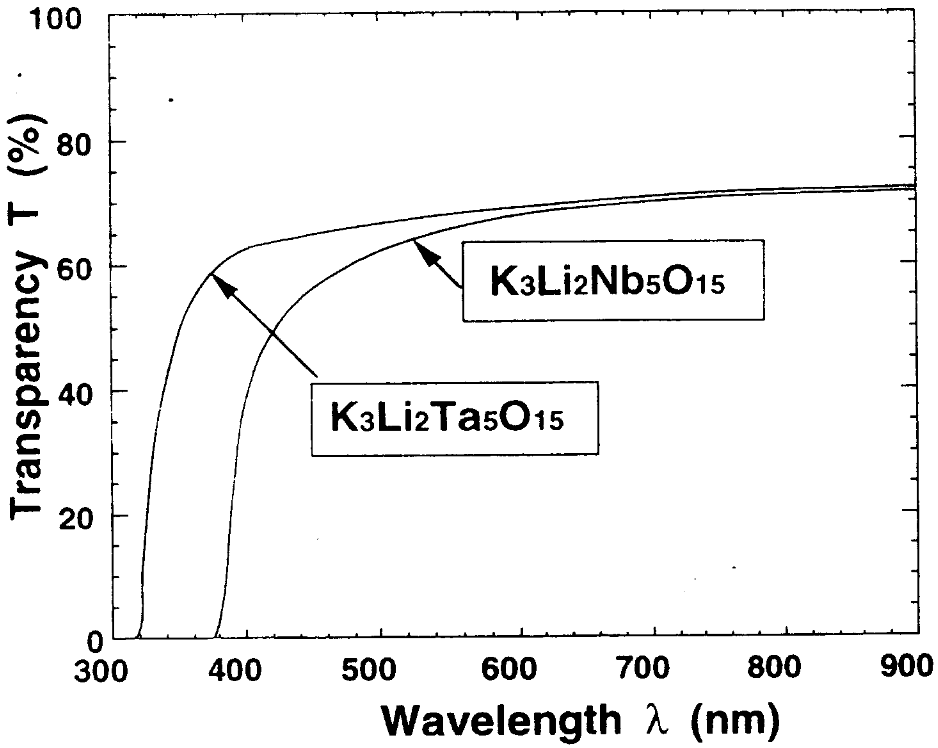
$K_3Li_2(Ta_xNb_{1-x})O_{15}$ crystal for Blue SHG

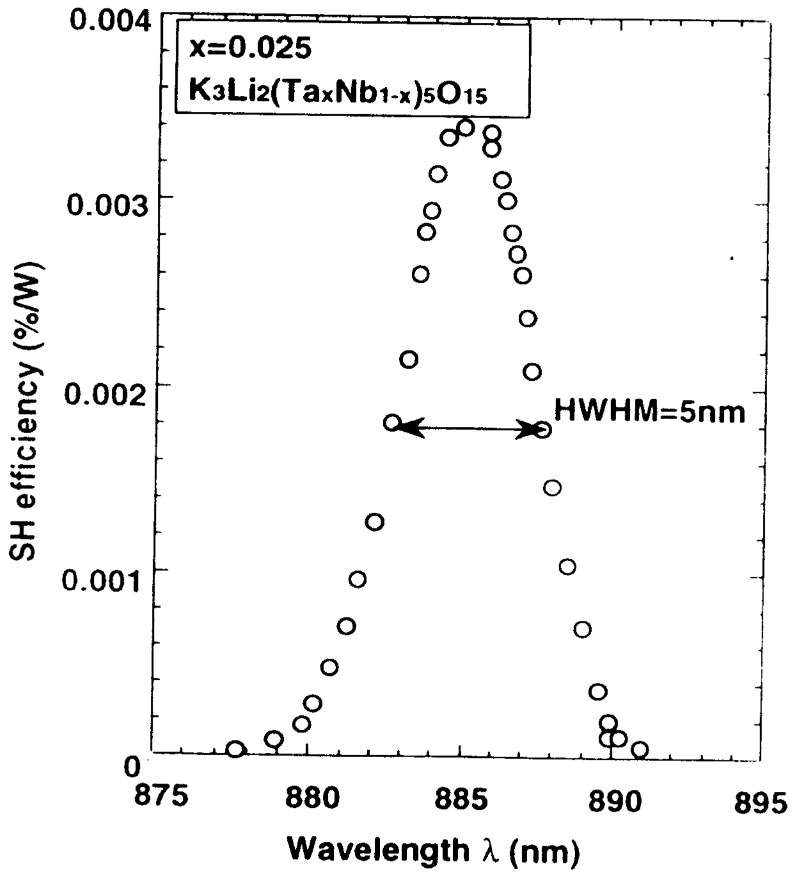


$K_3Li_2(Ta_xNb_{1-x})O_{15}$crystal
($x=0.8$, 10mmϕ x 45mm)
grown by double crucible method

- short absorption-edge : 320nm
- wide SH phasematching region:
  790 - 920nm
- large nonlinear coefficient : 14pm/V



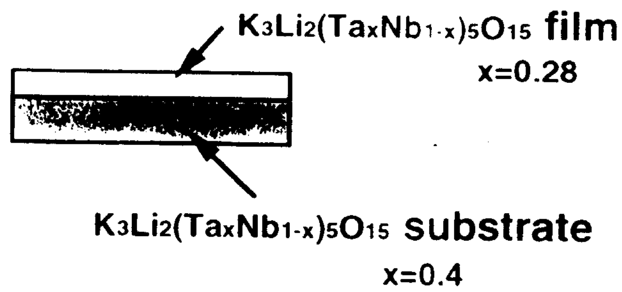
Characterization of KLTN crystal as a frequency doubler



< Issues >

- optical inhomogeneity
- difficulty in crystal growth
- grown from terminal solid solution in ternary eutectic system

1 mmϕ fiber crystal or LPE thick film



Summary

1. We have demonstrated all solid state blue SHG laser.

   20 mW of blue output at 427 nm was obtained with intracavity frequency doubling of a LD pumped Cr:LiSAF laser used by LBO crystal.

2. We are currently developing nonlinar crystals to improve device performance.

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