SPECTRALUS

WHITE PAPER

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Periodically-poled Lithium Niobate (Tantalate) Crystalline Components for Generation of Blue-Green Light from InGaAs/GaAs Diode and DPSS Lasers

Introduction

Since the invention of the first laser many years ago, the frequency conversion of laser radiation by nonlinear optical crystals has become an important technique widely used in quantum electronics and laser physics for solving various scientific and engineering problems. The fundamental physics of three-wave light interactions in nonlinear optical crystals is now reasonably well understood. This has enabled the production of various harmonic generators, sum- and difference- frequency generators, and optical parametric oscillators based on the nonlinear optical crystals that are now commercially available. At the same time, scientists continue an active search for improved nonlinear optical materials. In early 1980's it was recognized that in ferroelectric materials, optical second harmonic generation (SHG) efficiency can be dramatically enhanced by devising of the periodically poled domain (PPD) based quasi-phase matching (QPM). In recent years congruent and stoichiometric Lithium Niobate (LN), Lithium Tantalate (LT), and Magnesium Oxide doped LN and LT (MgO:LN and MgO:LN), KTP crystals have been used for application to QPM SHG. These materials are ferroelectric, which means that below Curie temperature they exhibit a spontaneous electric polarization and domain structure. Creation of the periodical domain structure by periodic inversion of the spontaneous polarization is called "periodical poling" and provides a means for producing the 180˚ phase shift required to implement QPM. Periodically poled ferroelectric based nonlinear optical materials are suitable for use in optical devices to convert near infrared (NIR) radiation from a diode or other lasers to light in the blue-green (visible) or near UV region of the optical spectrum.

Further information on the applications and properties of nonlinear crystals, especially LN and MgO doped LN, are given in "Handbook of Nonlinear Optical Crystals" by V.G Dmitriev, G.G Gurzadyan and D.N. Nikogosyan, Springer-Verlag (1999) ISBN3-540-69354-5 292.

Laser light in the blue-green (visible) wavelength region (i.e., 400-550nm) is used in a wide variety of analytical techniques. Currently the argon ion laser is the only generally available source of coherent blue-green light. Argon ion lasers are relatively bulky, delicate and expensive. There is a great need for a solid state blue-green laser. None is currently available, which emits in this spectral region. However there are solid state (semiconductor diode and DPSS) lasers, which emit in the near infra-red (NIR) region.

Frequency doubling (nonlinear frequency conversion) would enable a NIR laser to provide blue-green light. An extensive discussion of blue-green laser technology and applications, including biomedical engineering, spectroscopy, semiconductor wafer inspection, display science, optical data storage, reprographic, color display and undersea communication is contained in "Blue-Green Lasers" by W. Risk, T. Gosnell and A. Nurmikko, Cambridge University Press (2003) ISBN 0-521-52103-3. The discussion of nonlinear frequency conversion using QPM in nonlinear crystals contained therein is incorporated herein by this reference (see especially pages 77-84 and 101-104 and 108).

Conversion of NIR laser source light into light in the green-blue spectral range can be carried out by using SHG, also known as frequency doubling techniques, a technology generally known in the laser-based optical industry. A reasonable goal for single-pass conversion efficiency is that it should be in ~25% range to avoid excessive laser cost. Conversion efficiency is proportional to input power, the square of the effective nonlinear coefficient of the nonlinear element (crystal) and the length of the nonlinear element. As crystal length is increased, conversion efficiency increases, but the frequency doubling process becomes more sensitive to changes in temperature, strain and other factors affecting the uniformity of the refractive index of the nonlinear element. As a result, length alone cannot be used to compensate for an element's low nonlinearity. Therefore, the material with the maximum non-linear coefficient should be used to enhance the efficiency of conversion.

A technique that compensates for the difference in phase velocity between the fundamental wave and its harmonic in a nonlinear crystal caused by natural dispersion is known as birefringent phase matching. In this case, the optical anisotropy of a nonlinear crystal is used to find a unique propagation direction, where fundamental and harmonic waves have the same phase velocity. For most of the commercially available nonlinear optical materials (LN, LT, and KTP) the maximum conversion efficiency is about 1.25%. LN and LT are particularly attractive materials because of their status as commodity materials, and they have a high nonlinear coefficient (normally referred to as d_{33}) and also are available in relatively large size crystals.

QPM provides the mechanism for an efficient way to generate second harmonic frequencies. The general concept of using QPM as a mechanism for doubling optical frequencies has been known for about forty years. Essentially it is a technique that compensates for the difference in phase velocity between the fundamental wave and its harmonic in a nonlinear crystal caused by natural dispersion. In QPM, the two waves are allowed to have different phase velocities, and they shift out of phase relative to one another over a distance called the coherence length.

At present the most efficient way to create a QPM structure is to use periodically poled single-crystalline ferroelectric materials. In these materials, creating specific micrometer scale domain configurations with a periodically alternating direction of spontaneous polarization are used for this purpose. Due to the polar character of these materials the sign of the non-linear coefficient (d_{33}) can be changed by switching the direction of spontaneous polarization. If the period (Λ) of the periodically polled domain structure is equal to double the coherent length, the phase difference due to natural dispersion is compensated for by the change of the sign of the non-linear coefficient ($d_{33} \Rightarrow - d_{33}$) at the domain boundaries, causing the continuous transference of power from the fundamental beam to the harmonic beam throughout the entire length of the crystal.

Effective nonlinear coefficient value, poling period and absorption edge (the range of limited absorption) are the factors which influence the choice of periodically poled materials for SHG applications.

There is considerable interest in compact lasers operating in the blue-green spectral region (0.4-0.55μm) for various applications in science and technology such as biotechnology, data storage, optical image recording and displays. The preferred laser source would be a blue GaN-based diode laser. Although such devices are now commercially available, their output powers are limited (~20mW) and cost is relatively high. Thus, in the present circumstances, the frequency-doubling of near-infrared (0.8-1μm) commercial, low-cost diode lasers represents a competitive technology. Displayers for consumer application required multi-watt sources that can be manufactured in high volumes for less than $100/watt. To meet the needs of display applications, a new class of nonlinear optical materials with high optical-to-optical efficiencies would be required, and their development becomes the focus of vendors producing nonlinear-optical components.

Two of the most widely used nonlinear materials employed for frequency conversion of near-infrared diode lasers are Lithium Niobate ($LiNbO_3$ or LN) and Lithium Tantalate ($LiTaO_3$ or LT) partly because of their modest cost, but more importantly because they can be periodically poled to create quasi-phase matching conditions (QMC) that can increase the second harmonic generation (SHG) efficiency up to 80% for the a pulse laser. There is a lot maturity with these materials; however cost-effective manufacturing methods that produce stoichiometric LN and LT crystals do not exist yet. Standard congruent material is grown with strong deviation from the perfect stoichiometric composition. The lithium content is in range of 48%, whereas the ideal chemical formula for stoichiometric balance would be 50% lithium. The fact that the crystal is missing some lithium means that the crystal is rich in defects. Stoichiometric crystals exhibit low absorption, a low coercive field (allowing shorter poling periods) and lower susceptibility to photorefraction. Due to difficulties with the growth of stoichiometric crystals, high crystalline element processing (element orientation, cutting/dicing and fine optical polishing), and high labor cost, the price of a periodically poled LN or LT (PPLN, PPLT) element in USA is ~$1000.

In order to reduce the cost of PPLT/PPLN element to ~$100 it is necessary:

1. To investigate and develop appropriate material-processing procedures and fabrication techniques to allow wafers level fabrication of PPLN/PPLT elements with grating periods from a few microns to 30 microns;

2. To develop the techniques to make fabrication of high-quality PPLN and PPLT-based nonlinear elements a reliable and routine procedure.

Technical Description

1. Material selection

Tab. 1 shows that the most effective material for SHG application is LN crystal having biggest value of d_{33}. Due to QPM, it is possible to create viable bulk single-pass blue-green light sources for display applications, using LN, since it provides a way to obtain a normalized room temperature conversion efficiency of ~4% /(watt·cm) for 1064nm⇒532nm SHG and 5%/(watt·cm) efficiency for 920nm⇒460nm SHG, more than twice the minimum requirement.

LT has a normalized room temperature conversion efficiency of 0.85-1% /(watt·cm), below the display requirement for bulk single-pass 1064nm⇒532nm SHG. However, for 920nm⇒460nm SHG, LT has a normalized conversion efficiency of 2% /(watt·cm) and would be suitable for that application. It is important to point out that for the wavelengths of ≤410nm LT has higher transparency compared to LN as well and therefore it is material of choice for 300nm-410 nm wavelength conversion (Fig.1).

Two other potential materials in which QPM has been demonstrated for BG light generation are KN and KTP. A normalized conversion efficiency for these materials are ~2.5% /(watt·cm) for KN and 1.5%/(watt·cm) for KTP, for the 852nm⇒426nm SHG. To achieve 25% single-pass conversion efficiency, a KN crystalline element of 2.5 cm and KTP crystalline element of 3.5 cm length are required. However the maximum crystal length in production is 2cm for KN and 3cm for KTP. Therefore, KN and KTP have not been chosen because both crystals are not available in sufficiently long devices size, are prohibitively expensive and suffer from a number of quality issues.

The Spectralus's strategy for material development is to advance the technology of bulk and waveguide electrical field periodic poling in LN and LT 5-50 mm long, 0.5-2mm thick devices that could be routinely fabricated with good uniformity over an entire 3"-4" diameter wafer. Lithographic technique is used to produce QPM structure and to assure the periodicity, where even small errors in periodicity can substantially degrade conversion efficiency. The ability to define QPM/domain structure with lithographic precision created an opportunity to fabricate SHG-based conversion devices with performances not achievable using non-lithographic techniques.

Tab. 1. Parameters of perspective ferroelectrics for QPM SHG applications

Crystal	d_{33} pm/V	Curie Temperature °C	Coercive Field, KV/mm	Growth method and max. size
congruent $LiNbO_3$ 5%MgO:$LiNbO_3$ near stoichiometric $LiNbO_3$ stoichiometric $LiNbO_3$	34 44 	1145 -1133 1209 1200 	21 5 4 2-3	CZ, **100Φx60mm** CZ, **100Φx60mm** **DCCZ, 75Φx60mm** **VTE, 75Φx4mm**
congruent $LiTaO_3$ MgO:$LiTaO_3$ near stoichiometric $LiTaO_3$ stoichiometric $LiTaO_3$	2**6** 30 	**600** - **603** **690** **695**	21 ≤2 1.7 0.1	CZ, **100Φx50mm** **CZ, 100Φx60mm** **DCCZ, 75Φx60mm** **VTE, 75Φx4mm**
$KNbO_3$ (KN)	25±5	225	≤**0.6**	CZ, **15Φx20mm**
$KTiOPO_4$ (KTP)	15±2	**670**	**2,6**	HT, **20Φx25mm**

In LN and LT crystals QPM structure is fabricated by creating of the ferroelectric-domain-inverted grating. Electric field poling is used to create the domain grating. Fabrication of LN and LT QPM devices involves first lithographically patterning the domain grating structure. Then the domain-inversion process is produced by applying voltage to lithographically defined periodic electrode structure when the polar component of electric e field is larger than the coercive field (Tab.1). This technique is referred to as electric field periodical poling or simply periodical poling (PP).

Domain periods (Λ) between 6 μm and 8 μm are required for green light generation, and blue light generation requires domain periods between 2 μm and 5 μm. To reach a maximal efficiency of conversion, engineering of uniform domain grating with a duty cycle close to 50% in 0.5-2mm thick LN and LT single-crystalline elements is required.

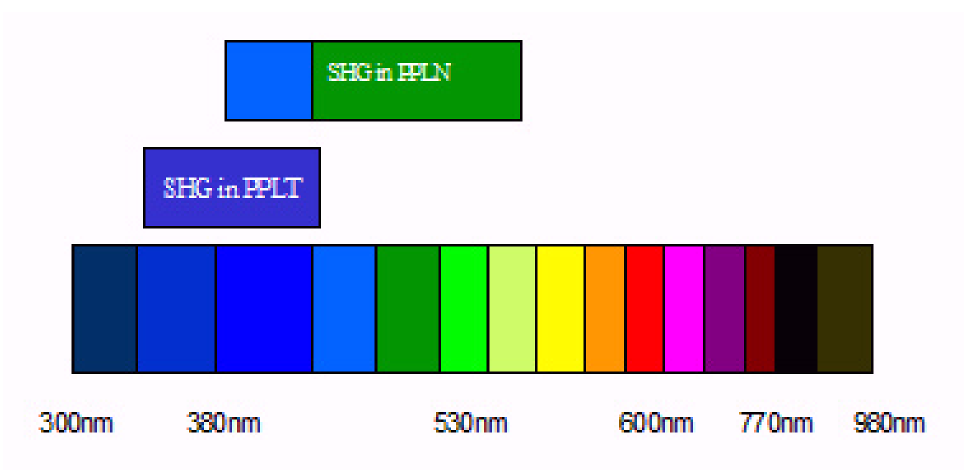

Fig.1. Wavelength accessible using GaInAs/GaAs lasers diodes and periodically LN and LT crystals

2. Engineering Periodic Domain Structure

Crystalline defects play major role: it is easier to create uniform domain grating in the stoichiometric LN, LT that have minimal defects, than in congruent LN and LT.

The kinetics of the domain structure in spatially nonuniform electric field produced by the electrode pattern depends on the number of factors: 1) shape of individual electrode, 2) electrode material, 3) variant of electrode structure (Fig.2), 4) geometry of the electrode pattern, 5) parameters of the dielectric layer, 6) poling waveform, 7) switching current limit, and 8) temperature. Moreover, the spatial uniformity of the switching characteristics, conductivity and thickness of the crystalline wafer is of crucial importance. The optimization of all technological factors can be done only the basis of the deep knowledge of the foundations of domain engineering in ferroelectrics.

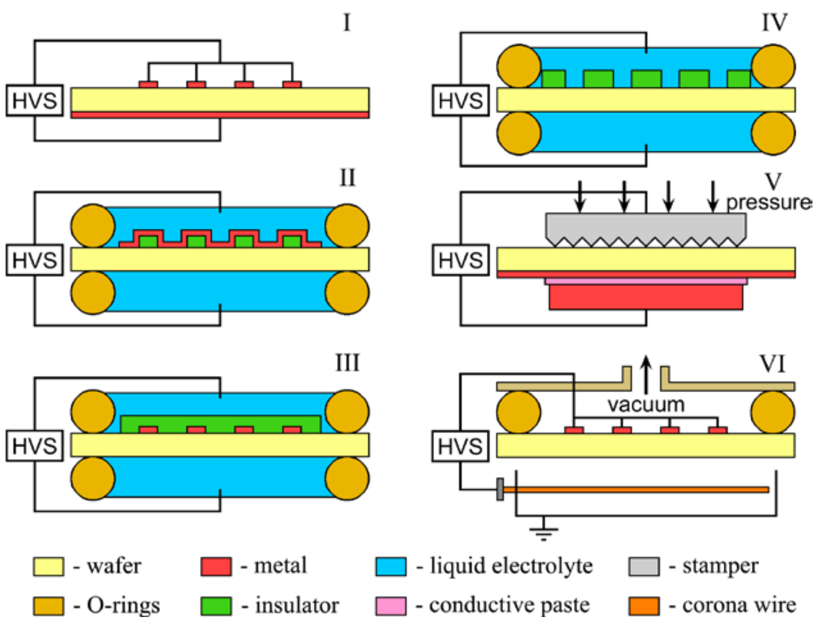

Fig. 2 Variants of electrode structures for periodical domain patterning by electric field poling. I – metal electrode pattern, II – metal electrode over insulator pattern, III – metal electrode pattern covered by insulator, IV – liquid electrode over insulator pattern, V – stamper electrode, VI – corona discharge method.

Table 2. Electrode structures for periodical domain patterning by electric poling

#	Method	Periodic electrode	Uniform electrode
I	Metal electrode pattern	Patterned metal	Metal
II	Metal electrode over insulator pattern	Patterned insulator covered by metal	Metal or liquid electrolyte
III	Metal electrode pattern covered by insulator	Patterned metal covered by insulator	Liquid electrolyte or metal
IV	Liquid electrode over insulator pattern	Patterned insulator covered by liquid electrolyte	Liquid electrolyte
V	Stamper electrode	Patterned stamper electrode	Metal
VI	Corona discharge method	Patterned metal and vacuum as an insulator	Corona electric discharge

Spectralus's efforts are directed towards identifying and optimizing those parameters contributing most significantly to repeatable, good-quality periodic domain pattern in LN and LT crystals.

An extensive discussion of domain structure development including investigations of the domain engineering aspects in $LiNbO_3$ is contained in: Ferroelectrics, V.221, pp157-167 (1999) by V. Shur, E. Rumyantsev, R. Batchko, G. Miller, M. Fejer, R. Byer

The domain kinetics during periodical poling from the single domain state in a spatially inhomogeneous field can be divided into five main stages: 1) nucleation of new domains at the surface, 2) forward growth of nucleated domains in polar direction with subsequent coalescence, 3) broadening of the strip domains by sideways domain wall motion, 4) stabilization of the domain structure in external field, and 5) backswitching after removing of external field (Fig.3).

All stages have to be carefully optimized to produce a specified domain period and duty cycle with acceptable uniformity throughout the volume of the wafer.

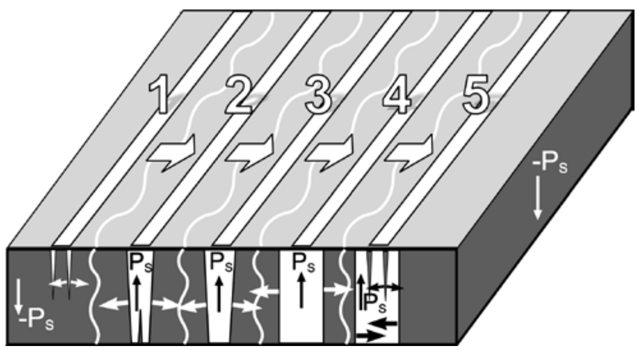

Fig.3. The main stages of the domain evolution during periodical poling

After detail analysis of the published data and basing on our experience we have chosen for the poling of CLN to use the most simple and easier for realization design of the electrode structure, so-called "photoresist only" (Fig. 2 IV). In this case the photoresist pattern is deposited on one side. The liquid electrolyte in spatial sample holder was used for application of electric field.

The scheme of used experimental setup is shown on Figure 4. The setup allows us to apply arbitrary shape poling pulses from TREK 20/20 pulse source to the sample located inside the holder made from acrylic resin. Accuracy of the poling pulse: time resolution 10 ns (100 MHz sampling rate), voltage resolution 14 bit. The lowered voltage and switching current are monitored via the digital storage oscilloscope Tektronix TDS1002, triggered by the generator. Oscilloscope is connected to the computer, thus downloading the measured data via RS232 protocol. Optical polarizing microscope with video camera allows us to record simultaneously the video and instantaneous pictures with view area about 1 mm^2.

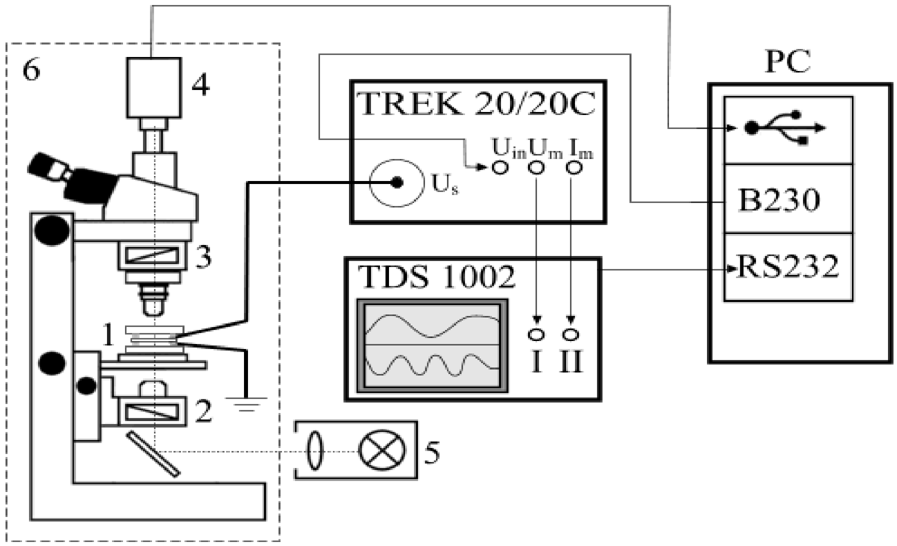

Fig. 4. Scheme of the poling setup: 1 – sample in the holder, 2 - polarizer, 3 - analyzer, 4 - video camera, 5 - light source, 6 - acrylic resin box.

3" wafer with patterned photoresist is placed in the sample holder for poling with liquid electrolyte – saturated water solution of LiCl. Two holders different in the rubber pads shape: (1) circular and (2) rectangular with rounded corners were used during the poling process.

The periodical poling of the MgO doped LN can be achieved only at the elevated temperatures. As a result we use another variant of the electrode structure with the metal electrodes (Fig.2 I).

The whole poling process contains several operations.

1. Optical inspection of the wafer using polarized microscope to reveal the bulk macro-defects and residual domains (deviations from single domain state).
2. Creation of metal electrode pattern by lithography with controlled heating/cooling rates during baking of photoresist
3. Inspection of the domain structure in the wafer just before poling to avoid appearance of residual domains by polarizing or phase contrast microscopy without application of the external field.
4. Periodical poling at elevated temperature above 100^{O}C in silicon oil with observation of domain kinetics in transmitted or reflected light with subsequent low cooling rate to avoid wafer cracking and to uncontrolled change of the tailored domain structure.

The quality of a periodically-poled (PP) structure is mainly determined by two factors: periodicity and duty cycle (DC). The periodicity of the PP structure strongly affects the phase-matching wavelength of a conversion device, while the DC of the PP structure affects conversion efficiency. Maximum conversion efficiency can be achieved for a perfect uniform DC of 0.5. Therefore, to estimate the uniformity of the DC, the optically microscope images of the etched (in pure hydrofluoric acid) periodically poled surface were carried out (Fig. 5). The widths of domain inverted region were measured on the Z+ and Z- surfaces in the fabricated PPLN element. Uniform PP structure with a 6.75 μm period and 0.5$\pm$0.1 DC has been fabricated from the Z+ surface to more than 400 μm depth over an area of 5 mm by 10 mm.

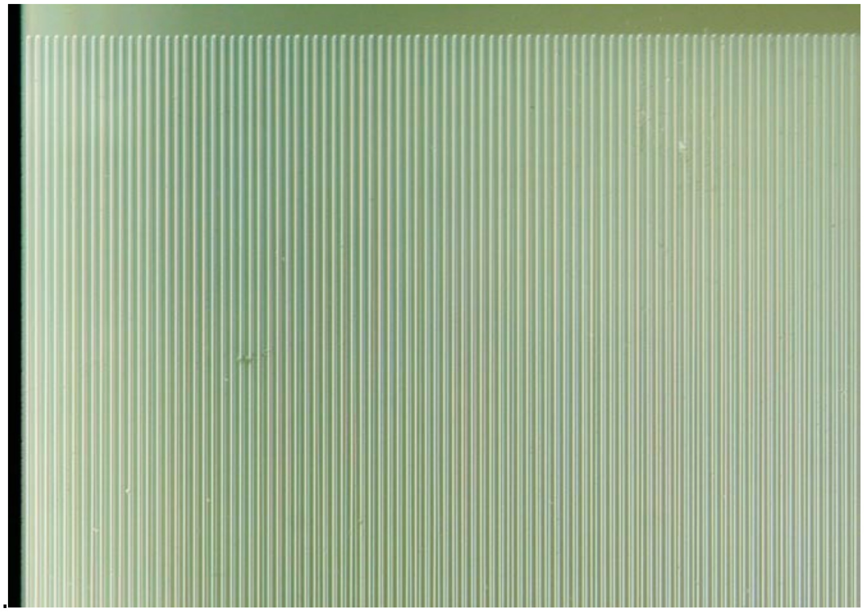

Fig. 5. Optical microscopic image of the etched PP structure for the 1.0 mm thick PPMgOLN element with a 6.95 μm domain inverted period on Z+ surface.

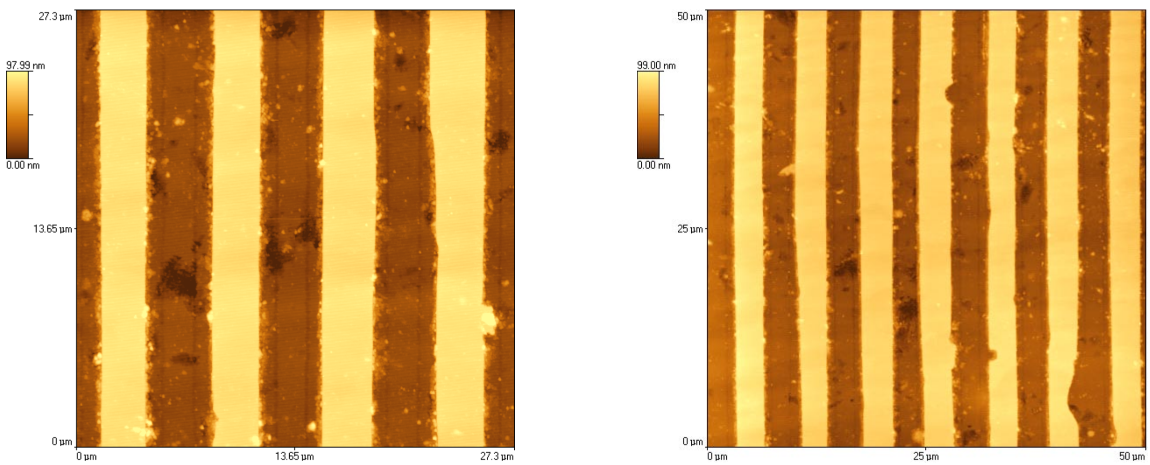

Fig. 6. Scanning probe microscopy observation of the periodical domain structure revealed by etching in 1.0 mm thick MgO:LN

3. Applications

An important application of periodically poled ferroelectrics is wavelength conversion of commercial diode and compact DPSS near-IR lasers to blue, green, and medium-IR spectral regions. Periodically poled LN and LT have clear advantages over the birefringence phase-matched KTP, LiB_3O_5 (LBO) and BaB_2O_4 (BBO) nonlinear crystals in terms of 3-5 times lager effective nonlinearity and a possibility to phase-match any second-order interaction within the transmission band of material. Due to high conversion efficiency, PPLN devices are well suited as a nonlinear component for SHG from compact, low power consumption (<10W) lasers, capable of producing 0.1-1W optical power. For the testing of our PPLN elements, the YAG:Nd DPSS laser, that produces 300mW cw at 1064±0.1nm in single-transverse mode, has been used.

Figure 7 present the dependence of the SHG (532nm) outputs on temperature. Excellent fitting of experiment and calculations demonstrates higher quality of fabricated periodically-poled grating, presented in Fig. 5, 6.

A doubling of frequency of diode laser by high-efficient PPLN or PPMgOLN nonlinear-optical components is likely to be the winning technology for low-power blue-green laser market needs.

The diode laser used in present work was InGaAs VECSEL surface-emitting laser . Both their optical mode characteristics and wavelength were controlled by an extended compound optical cavity. The laser produced ~0.15 W cw at 1064±0.2 nm in single mode operation.

The PPLN components have narrow acceptance bandwidths of about 2 nm/mm and therefore 5mm of length conversion elements are optimal for doubling of VECSEL diode lasers. On the other hands, conversion efficiency of periodically-poled elements is proportional to the fundamental optical power of laser. To increase the conversion efficiency, pulsed laser, modulated by an injection current, has been used. By using 2 A current pulse, 0.5 W optical power was generated in single-transverse mode.

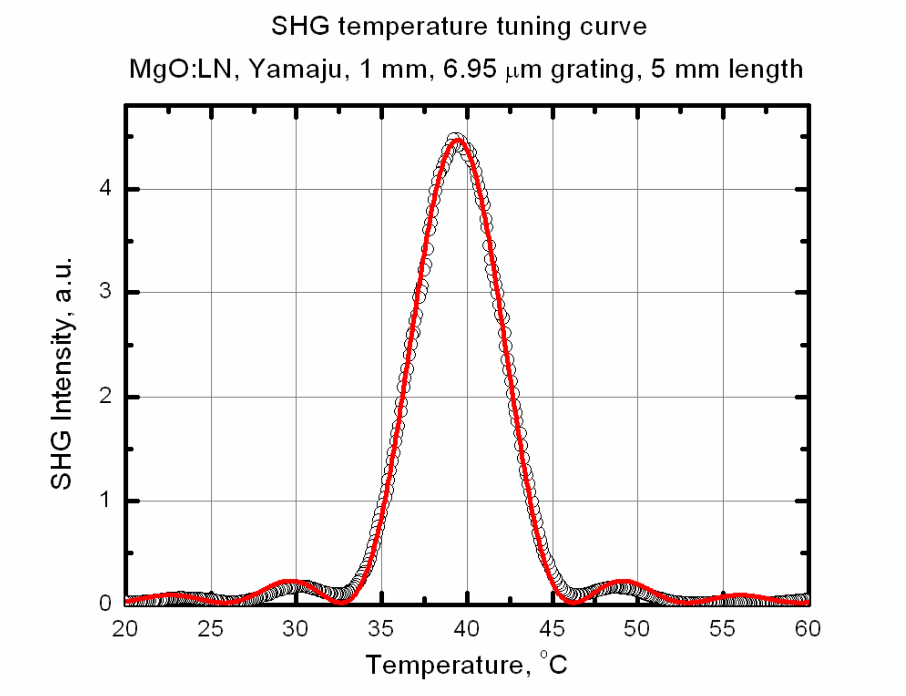

Fig. 7. Temperature dependence of the green light maximum intensity (532 nm output power) for MgO PPLN period 6.95 microns. The dots are experimental data and the solid curve is theoretical curve for a 5 mm length.

Detail analysis of pulse regime has shown that a wavelength chirp of 5nm/μs rate takes place.

Therefore, for optimization of SHG process, pulse duration and length of PPLN element was restricted to 100 ns and 3mm. We have demonstrated 10mW average and 1W peak green power light generation from the intracavity-doubled VECSEL laser (Fig. 8).

No degradation of green light power was observed during 100 hours of operation.

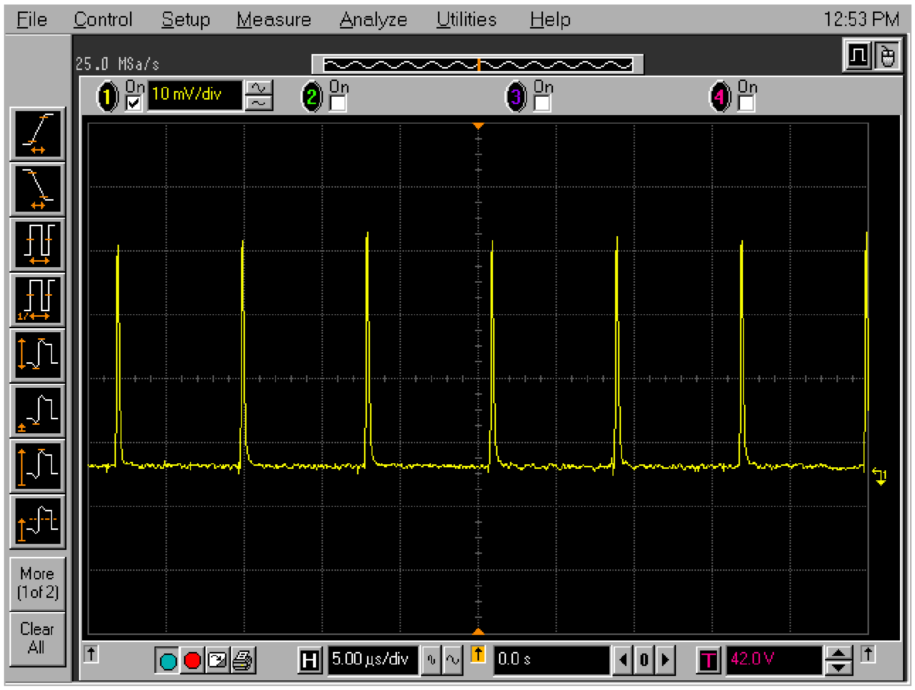

Fig. 8. Pulsed 532nm output generated by the intracavity-doubled of VECSEL (1064nm) laser.