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論　文　内　容　要　旨

CAPTER 1　Introduction

Activated ferroelectric crystals, ferroelectrics that are doped with rare earth atoms, allow us to utilize not only the nonlinear or electro-optic properties of the host material but also the different fluorescence spectra resulting from the dopant. This will allow nonlinear frequency conversion, electro-opric effect and the lasing action to be established all within a single crystal. The advantage of this type of frequency cpnversion over conventional external second harmonic generation (SHG) or optical parametric generation is that it establishes a resonant condition for the 'pump' wavelength which directly involves in nonlinear frequency conversion process, providing higher efficiency of frequency conversion, without the need of any precise adjustment of the cavity length.

Growth of these kinds of doped nonlinear ferroelectric crystals or activated ferroelectrics opens up a wide range of scopes and possibilities for scientific and technical applications by performing frequency conversion and lasing within the same crystal. However, several interactions between the host crystal and the additive ions restrict the number of useful material combinations. These include size disparity, valence and spectroscopic properties. There are several reports on the growth of rare earth doped ferroelectric lithiom niobate ($LiNbO_3$), but with only a few of them have been lased successfully. Despite the high nonlinear coefficient and ease of handling and polishing, the need for operation at elevated temperatures combined with a relatively lower damage threshold have severely limited the practical applications of this material. On the other hand, lithium tantalate, $LiTaO_3$ which is an isomorphs of $LiNbO_3$, has been proven to have higher damage threshold and the same order of nonlinear and electro-optic coefficient. Until recently, in was not popular for freuyency conversion because of the phase matching problems which arose from its lower birefringence. However the advent of quasi phase matching (QPM), has made the nonlinear material very attractive for second harmonic and optical parametric devices.

As regards to the dopant ions, the exceptionally favorable characteristics of the trivalent neodymium ion for laser action were long recognized. This atom exhibits a satisfactory fluorescence lifetime and narrow fluorescence linewidths in crystals with ordered structures and possesses a terminal state for laser transition sufficiently high above the ground state allowing cw operation at room temperature. Furthermore, it exhibits absorption band which matches well with commercially available high power laser diodes.

This thesis is devoted to the construction of integrated multifunctional laser devices from doped nonlinear ferr oelectrics, namely, neodymium doped lithium tantalate, Nd:$LiTaO_3$ (Nd:LT) by modulating the domain structure. Studies are first concentrated on the selection, growth and processing of the doped Nd:LT crystals and on the optical and lasing characteristics. Modulation of the domains of the activated ferroelectric host is then carried out with an aim to obtain SHG by the method of QPM. Novel methods are proposed and implemented for integrated Q-switched operation and SHG employing the electro-optic and nonlinear properties. The thesis describes in detail all the ideas and techniques that have been developed in the course of achieving this kind of integrated device.

CHAPTER 2 Technical background

The necessary technical background required for constructing the integrated nonlinear device from doped ferroelectric has been summarized here. This chapter has briefly described the optical effects in activated ferroelectric laser crystals. Spectroscopic properties of some other activated ferroelectric laser crystals have been mentioned as a reference. Phase matching necessary for efficient frequency conversion within the crystal has been pointed out. Two techniques for phase matching—birefringent and quasi phase matching have been described. In order to determine the domain period necessary for QPM, an equation has been formulated from coupled wave equation. Finally, examples of operation of several interesting frequency conversion device based of QPM on undoped ferroelectric have been cited.

CHAPTER 3 Doped nonlinear opical material

It has been shown that $LiTaO_3$ is a very suitable host from several points of view, such as its high nonlinear and electro-optic coefficient, its high damage threshold and its ability to accept Nd ions without any quenching. The growth of the Nd:LT crystal, annealing and poling of these materials have also been described here. Poling near Curie temperature required the application of a DC voltage of about 15V/cm whereas poling at room temperature required a considerably higher voltage of about 16.8kV/mm.

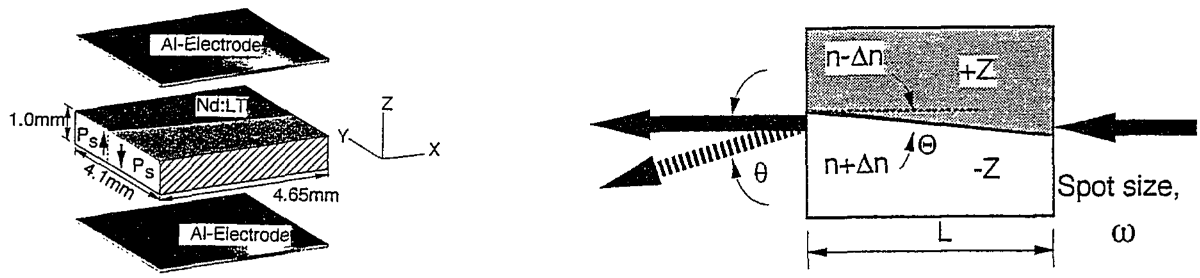
Asymmetric behavior of the hysteresis loop has also been studied in context to the domain inversion at room temperature. It has been observed that the coercive force is reduced by a half, when domain reversal is performed to restore the spontaneous polarization direction back to the original direction. The pryoelectric properties of $LiTaO_3$ has also been studied.

CHAPTER 4 Continuous wave laser oscillation in Nd:$LiTaO_3$

Thorough investigation of the optical properties of the Nd:LT crystal has been carried out to aid the design of integrated laser devices. Absorption spectrum showed that it has high absorption (5.6cm^{-1}) around 806nm for σ polarization. The fluorescence spectra, associated energy levels and the lifetime have also been measured for different concentration of Nd_2O_3. It showed moderate lifetime of about 100μs. Annealing and poling of the material was found to be very effective in improving the optical qualities of the material and in oscillating with π polarization. Finally this chapter describes the demonstration of cw laser oscillation at room temperature in Nd:LT at 1.092μm and 1.082μm with ordinary (σ) and extaordinary (π) polarization, respectively, by pumping at 806nm. Threshold values of about 11mW, 8mW and slope efficiency of 16.8%, 11.7% are obtained from uncoated samples with π and σ lasing, respectively.

CHAPTER 5 Integrated electro-optic Q-switching in a domain inverted Nd:$LiTaO_3$ laser

In this chapter a novel method for electro-optic Q-switching that integrates the laser gain and the Q-swiching action within a single Nd:LT crystal has been presented. The single lasing crystal has been uniquely designed to possess two domains of oppositely directed polarization. The σ of the cavity has been controlled electro-optically by exploiting the deflection caused by the boundary of these two oppositely directd domains under an applied electric field.



(a) (b)

Fig. 1 Geometry of the integrated electro-optic Q-switching and lasing crystal. The axis of optical propagation is x, and the electric field is applied along z. The angle of inclination Θ of the boundary is 3°. (a) Exploded view. (b) Schematic explaining the deflection caused by the domain boundary under the applied field.

The geometry of the Q-swithched lasing crystal is shown in Fig. 1 (a) and (b) for explaining the principle behind its operation. Two oppositely directed domains are formed using dc field-poling technique within the x-cut crystal such the the boundary between them makes an angle Θ with the optical propagation direction. When an electric field of amplitude E_3 is applied along the optical axis (z), the magnitude of the excursion, Δn_3 of the extraordinary refractive index, n_3 will be giben by,

$$\Delta n_3 = r_{33} n_3^3 E_3/2$$

where r_{33} is the electro-optic tensor component. The sign of this deviation will depend on the relative orientation of the spontaneous polarization to the electric field. For a uniform field E_3 applied to both the regions, the refractive indices at the two regions will be $n_3 + \Delta n_3$ *and* $n_3 - \Delta n_3$.

For small angle of inclination Θ of the boundary, the total deflection θ of the beam can be approximated by,

$$\theta = 2\Delta n_3/\Theta = r_{33} n_3^3 E_3/\Theta$$

Although Δn_3 in small compared to n_3 under practically applicable electric fields, for small angles of inclination Θ, it can been shown to be enough to cause deflection of several millirads from the propagation axis and to inhibit the oscillation.

When the crystal having this unique shape of domain structure is placed inside a resonator, it is, therefore, possible to electro-opticaly switch the cavity Q between the low-loss state and the high-loss state to achieve Q-switching. The Q-switched laser based on this technique could generate 24.7ns pulses at 150Hz with peak power of 148W under LD pumping. The operationg voltage necessary for this Q-switching was found to be about 600Volts.

CHAPTER 6 Theoretical analysis of domain period for quasi-phase matching

This chapter is devoted to the derivation of a temperature dependent dispersion relation of the extraordinary refractive index of $LiTaO_3$. Also a temperature independent dispersion relation has been formulated for the ordinary refractive index. Based on these equations, the necessary periods for quasi phase matching in Nd:LT have been calculated and the effect of temperature on the period has been studied. The bandwidth and the temperature sensitivity have also been studied. During the theoretical study, retracing behavior has been found to exist in the plot of domain period vs. wavelength.

CHAPTER 7 Intracavity quasi-phase matched second haemonic generation and Q-switching in a periodically poled Nd:LiTaO$_3$

This chapter describes an LD pumped integrated intracavity QPM second harmonic generation based on periodic poling of Nd:LT. The Nd:LT crystal when pumped near 806nm by a source, it establishes lasing at 1.082 μm with π polarization.

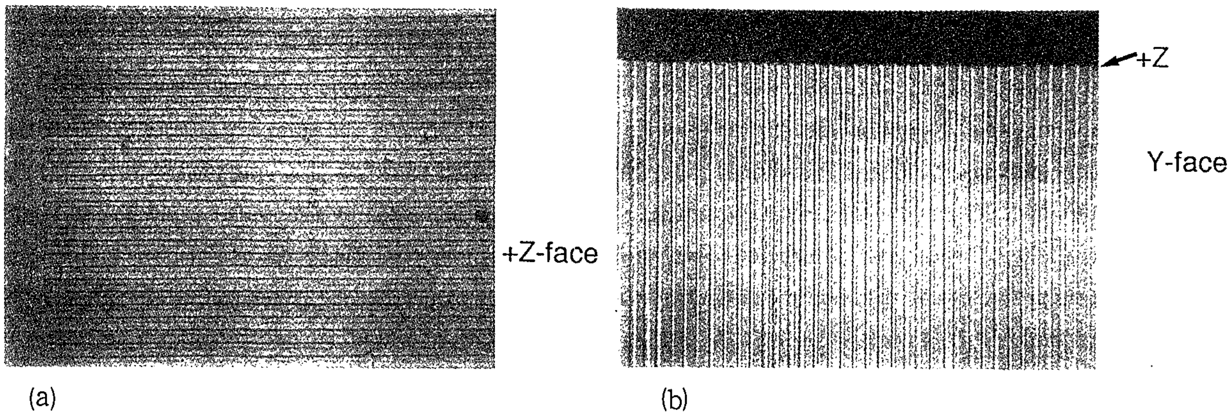


Fig. 2 Photograph of the etched facs of the periodically domain inverted Nd:LT laser. (a) show the +z face (b) the y face, the wider strips are those under the electodes and experience domain reversal. Thickness is 500 μ m and period is 8.1 μ m.

Bacause of the periodically poled structure of the Nd:LT crystal, the lasing will be simultaneously converted to green SHG at 541nm by quasi-phase matching. The periodic poling has been performed by applying controlled high voltage to patterned electrode. Domain period is precisely defined by a lithographic musk by use of standard microfacrication technique. The domain inversion voltage of Nd:LT sample with doping 0.44wt% is 16.8kv/mm. A phot graph of such a periodically poled Nd:LT sample is shown in Fig. 2.

Furthermore, an intracavity QPM-SHG and the simultaneous Q-switched operation using a single periodically poled Nd:LT crystal has also been shown to obtain Q-switched green pulses. This has become possible by means of the combined exploitation of both the nonlinear and the electro-optic property of the activated host. In this way 39ns green pulses at 541nm with a peak power of 2.4W as shown in Fig. 3 have been obtained by pumping with a 0.7W fiber LD.

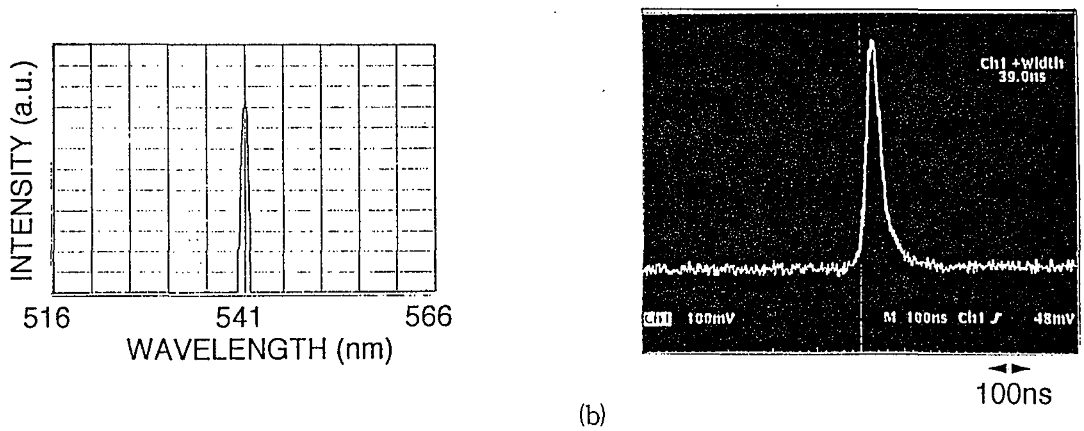


Fig. 3 Spectrum (a) and waveforn (b) of the Q-switched, π polarized quasi phase matched second harmonic, generated by the integrated, domain modulated Nd:LT laser.

CHAPTER 8 Summary and conclusion

The results obtained in the present study can be summarized as follows:

1) Activated ferroelectric Nd:LT is proved to be very suitable for use in integrated multifunctional laser device because of its high nonlinear and electro-optic coefficient, its high damage threshold and its ability to accept Nd ions without any quenching.

2) Annealing and poling are found effective in improving the optical proprties of the material and in establishing the extraordinary lasing within the meterial.

3) Domain structure of the material has been engineered to utilize the electro-opric properties in conjunction with the lasing properties and to establish integrated Q-switched operation. A novel and compact integrated Q-switched laser generating 148W peak, 24.7ns Q-switched pulses, has been devised based on this unique structure.

4) Integrated QPM-SHG and simultaneous Q-switching has been demonstrated by successful periodic modulation of the domain of Nd:LT and by novel electrode structure. This was made possible by the use of both the nonlinear and electro-optic properties of the host in conjunction with the lasing properties of the active ion. In this way 39ns green pulses at 541nm with a peak power of 2.4W has been obtained at room temperature by pumping with a 0.7W fiber LD.

審 査 結 果 の 要 旨

非線形光学は，光波による光波の超高速制御の基礎技術であり，多彩な機能を利用した光デバイスへと展開する可能性を有している。一方，固体レーザはレーザ活性イオンを固体結晶中に含み，高出力半導体レーザ励起によって，スペクトル線幅の狭い高出力でビーム品質の良いレーザ出力を得ることができる。一般に非線形材料とレーザ材料は別個であり，レーザ出力を非線形光学材料に入射して相互作用を実現する。本論文は，非線形光学材料である $LiTaO_3$ 結晶に Nd イオンをドープしレーザ結晶と非線形媒質を一体化することにより，集積化した新機能を発現させるための基礎研究を行った結果をまとめたもので，全8章よりなる。

第1章は序論である。

第2章は，レーザ活性イオンをドープした強誘電体材料に関する従来の研究結果をまとめ，その発振特性が強誘電体のドメインを周期的に反転し，その周期を制御して位相整合を実現する疑似位相整合デバイスとの整合性に優れていることを示している。これらの検討結果から，本研究では最適な材料として Nd イオンをドープした $LiTaO_3$（以下 Nd:LT と略す）を取り上げる。

第3章では，CZ 法で成長させた Nd : LT 結晶について，熱処理および単一分極処理を施し，材料の強誘電体特性を測定したを述べている。第4章では，動作の基礎となる吸収スペクトルや発光スペクトル特性を詳しく調べている。さらに，レーザチップに加工してチタンサファイヤレーザを励起光源として，レーザの連続発振特性を測定している。この結果，Nd:LT はレーザ材料として優れた特性を持つことを明らかにした。

第5章では，LT 材料の電気光学効果とドメイン反転構造を利用した光偏向素子を Nd:LT レーザチップに集積化し，高速に電気的にスイッチすることにより，Qスイッチ動作を実現している。半導体レーザを励起に用いた動作において，発生した光パルスはパルス幅 24.7ns，ピーク出力 148W と，連続動作時の約千倍のピーク出力を得ており，この結果は高く評価される。

第6章では，Nd:LT に対する疑似位相整合条件を詳細に検討している。このために，屈折率を光波長および温度の関数としてあらわす精度良い近似式，LT について導出している。第7章では，ドメインを約8 μm 周期で反転した Nd:LT を製作し，レーザ発振とともに第2高調波の同時発生を初めて実現した。また，第5章で述べたQスイッチ動作をこの素子に組み込み，541nm の第2高調波をピーク出力 2.4W で発生した。非線形光学結晶に，レーザイオンとともに周期ドメインやQスイッチ構造を集積化した本研究は，新たな高機能光源の可能性を示したものとして高く評価される。

第8章は結論である。

以上要するに本論文は，レーザ活性イオンをドープした強誘電体の非線形光学材料を用い，ドメイン特性の制御により新たな機能が実現できることを示したもので，実用上有用であり，量子電子工学に寄与するところが少なくない。

よって，本論文は博士（工学）の学位論文として合格と認める。