	もられす てらおか　えどがる よしお
氏　　　　　名	モラレス 寺岡　エドガル 芳男
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指　導　教　員	東北大学教授　山田　博仁
論文審査委員	主査　東北大学教授　山田 博仁　東北大学教授　中沢 正隆 東北大学教授　横山 弘之　東北大学准教授　大寺康夫

論文内容要旨

Nonlinear optic devices have emerged as a practical answer to several applications. Ranging from telecommunications, optical computing, metrology, biomedical applications, nonlinear effects have been used to fabricate efficient harmonic generators, optical switches, tunable parametric oscillators, mixers, wide spectrum light sources, etc.

In recent times, there has been a growing concern regarding power consumption and device miniaturization. With the emerging of a multimedia-based society, with huge amounts of data transmitted and processed every second, the great amount of power required for data transmission has been a growing concern. One of the answers to this problem has been to fabricate photonic integrated circuits (PIC), efficient and low power consuming devices that can be integrated in a single chip. Semiconductors play a key role in this technology, and the current interest in nonlinear properties in semiconductors is fueled by this need.

Wide bandgap semiconductors in particular are very attractive for applications in nonlinear optical devices. In general, devices fabricated with wide bandgap materials can work at higher temperatures and transmit higher powers without damage, and also allow the operation in a wider spectrum of wavelengths with minimum loss.

For a material to be considered candidate for nonlinear devices, it has to fulfill certain requirements. It has to have a high nonlinear index, to develop nonlinear effects with minimum power. There has to be advanced thin-film technology, to be easily integrated into an optoelectronic device. Highly nonlinear materials that have received attention and are traditionally employed include $LiNbO_3$, InGaAs, AlGaAs, GaN, chalcogenide glasses, etc. While the technology for these materials is advanced and commercial products are available, they are based on rare earths, which are a very scarce resource, and the technology is expensive and complicated. Therefore, simpler and cheaper alternatives are always needed.

We propose nonlinear optical devices based on ZnO waveguides. ZnO is also a material that lands in the category of highly nonlinear materials, with several qualities that make it a very interesting material. Very high quality bulk and thin films are available, it has a wide bandgap, highly abundant, non toxic and the technology for its growth is relatively simpler and cheap. It has received less attention in this field because high quality thin films are just recently available. The main topic of this thesis is the fabrication and characterization of ZnO waveguides for nonlinear applications.

The first chapter is the introduction. Here, the proposed structure for nonlinear waveguide devices is presented. We propose a channel waveguide with sapphire substrate, ZnO core and air cladding. The channel structure, together with the air cladding will help reducing the cross-sectional area, improving the power density, which is essential for nonlinear optical effects. The main properties of ZnO are also discussed in this chapter. ZnO is a direct band gap semiconductor, with a wide band gap of ~3.3eV. For its qualities, it is regarded as a prospective material for optoelectronic applications. It has a hexagonal wurzite crystal structure, with a refractive index $n \approx 2$. The chapter also presents the outline of the dissertation.

Chapter 2 deals with the theoretical calculations. We perform theoretical analyses on the ZnO channel waveguide platform that lead to the parameters of design for the waveguide fabrication.

The single mode condition is obtained for a variety of waveguide sizes. Considering working at a wavelength of 830 nm, we show that the waveguides of height less than 0.7 μm and width of ~1 μm or less are single mode. High quality single crystal ZnO can be grown until a thickness of 1μm , therefore single crystal quality should not be a critical factor in the design of the waveguides.

The pulse propagation equation is obtained from the Maxwell's equations. The key factors that define pulse propagation are propagation loss, group velocity dispersion and nonlinear effects. The strength of each is determined through 3 parameters, namely the loss constant α, the GVD coefficient β_2 and the nonlinear parameter γ.

We calculate β_2 from the results of simulations based on the finite-element method. The maximum value is for the waveguide 400 nm tall, with $\beta_2 = 2.4\, ps^2/m$. Considering 160 fs pulses, the dispersion length L_D is about 1 cm. Since the raw thin-film samples are 1 cm long, even considering the complete sample we conclude that the dispersion should not be a factor.

The nonlinear parameter γ is calculated by obtaining the effective modal area A_{eff}. The effective modal area showed a gradual decrease with decreasing waveguide size and a sudden increase after reaching a minimum. This suggested optimization of the waveguides for maximum nonlinearity is possible. The highest nonlinearity is for waveguides of height 0.5 ~ 0.7 μm and 0.6 ~ 0.8 μm wide. The maximum γ value corresponds to the 0.6 × 0.6 μm² waveguide, with $\gamma = 22.5\ W^{-1}m^{-1}$.

The nonlinear length L_{NL} is calculated as a function of the input peak power. For an input peak power of over 100 W, the nonlinear length is less than 1 mm, reaching less than 0.1 mm for 300 W. We find that nonlinear effects clearly dominate the propagation regime.

In order to obtain channel waveguides with maximum nonlinearity, the waveguide height is kept lower than 0.7 μm and the width is varied around 1 μm.

Chapter 3 is about the fabrication process of the ZnO channel waveguides from thin-film samples. The thin-film samples were grown at Kawasaki laboratory, using the laser-MBE method.

The fabrication process consisted in the following steps.

a. Putting a resist mask, using EB-lithography.
b. Etching of the samples, using Ar-ion milling.
c. Cleaning of the resist in an ultrasonic bath with acetone and ethanol.
d. Cutting the samples to the desired length, by cleaving the sapphire substrate.

The factors that affect the etching process are: bias power, chamber pressure, Ar flow rate and process time. We find that each factor affects differently.

1. Bias Power. The minimum power at which etching is registered is 100 W. The etching rate rises monotonically with the bias power, from around 35 nm/min for 100 W up to 80 nm/min for 300 W. The maximum allowed power is limited by the thermal damage of the resist.
2. Chamber pressure. The etching rate is maximum at the range of 1~2 Pa. A drastic drop in the etching rate is measured for higher pressures.
3. Ar Flow rate. The maximum etching rate corresponds to a flow rate of 50 sccm. Higher flow rates register lower etching rates.
4. Process time. Thermal damage and wrinkling of the resist limited the maximum process time. The process time is split in 5-minute etching steps to prevent thermal damage.

The final etching conditions are:

Etching conditions	
Inductive power	800W
Bias power	100W
Chamber pressure	1Pa
Ar flow rate	50sccm
Process time	5+···min

As result of this process, we successfully fabricate 500 and 380nm–high waveguides of several widths. They are cut by cleaving the sapphire substrate to achieve a flat face. The chosen lengths are 1, 2 and 4 mm to guarantee nonlinear effects and to allow the cut-back measurements.

In chapter 4, the measurement of the propagation loss of the fabricated ZnO channel waveguides is presented. We apply the cutback method to the waveguides to calculate the propagation loss. A strong dependence of the propagation loss to the waveguide width is verified. The waveguides show a propagation loss of 1.5 – 3 dB/mm for the single–mode waveguides and 1 – 1.5 dB/mm for the multimode case. Also from these measurements, the coupling loss is calculated for the lensed fiber – channel waveguide setup.

Possible propagation loss causes are sidewall roughness, surface roughness and material absorption. Sidewall roughness is the most probable cause of propagation loss. To confirm this, we obtain a model of the propagation loss based on scattering loss caused by the coupling between the fundamental guided mode and radiation modes. We characterized the propagation loss as a function of the standard deviation of the roughness σ and the autocorrelation length L_C.

The measured propagation loss values and the model predictions agree very well. We conclude that the sidewall roughness is the main factor for propagation loss in ZnO channel waveguides.

Chapter 5 presents the observation of nonlinear effects and the calculation of the nonlinear parameters. We propagate 160-fs pulses in the ZnO channel waveguides and measure the output spectra. The pulses show a spectral broadening of up to 9 times the input spectral width. The broadening is caused by nonlinear effects, specifically self-phase modulation. We verify that the nonlinear effects are developed in the ZnO waveguides by moving the output fiber to increase the coupling loss and measuring the same spectral shape at lower power.

We obtain the RMS spectral width of the measured spectra and used it to obtain the phase shift and subsequently the nonlinear parameter gamma. A maximum phase shift of 2.9π was obtained. We verify the validity of the obtained parameters by performing simulations of the pulse propagation equation, using the split-step Fourier method. The results of the simulations closely resemble the measured spectra, corroborating our results.

The analysis is repeated for all the measured waveguides. The nonlinearity of the waveguides shows a strong dependence on the waveguide geometry. In general, the nonlinearity increase as the cross-sectional area decrease. The 380 nm-waveguides show a sudden drop in the nonlinearity for sub-micron widths due to the mode leaking to the substrate. The waveguide with the maximum nonlinearity is the $0.5 \times 0.7\ \mu m^2$ waveguide, but it is not the one that shows the wider spectrum due to the high coupling losses involved. The corresponding value of gamma is $\gamma = 22.2 \pm 4\ W^{-1}m^{-1}$, which represents more than 2000 times the value of a highly-nonlinear waveguide.

The nonlinear refractive index n_2 is also calculated, back-tracking from the nonlinearity γ and the effective area A_{eff}. We obtain a value of $n_2 = 9.94\times10^{-19}\ m^2/W$, in close agreement with values available in the literature.

Finally, chapter 6 presents the conclusions and suggestions for future work.

論文審査結果の要旨

医療診断や光通信応用の分野では，広い波長領域にわたって高輝度の発光が得られるコンパクトな光源が求められている。そのような光源を実現する手法として，ピークパワーの大きな超短光パルスを非線形光学材料に入射させて，自己位相変調効果などの非線形光学効果を用いてスペクトルを広げていく方法が知られている。本論文はこのような応用を目指して，ZnO というワイドバンドギャップ半導体に着目し，それを用いて光導波路を作製し，その非線形光学応答特性について探究したものであり，全編 6 章からなる。

第 1 章は序論であり，本研究の背景と目的を述べている。

第 2 章では，本研究で用いた光導波路の解析手法について述べている。その手法に基づいて，ZnO をコアとするチャネル型光導波路の波長 800 nm 帯での単一モード条件や群速度分散特性について解析している。また，非線形光学効果を解析するためのパルス伝搬方程式についても述べ，これを用いて非線形光学長を計算し，100 W 以上のピークパワーの時，1 mm 程度の導波路長で非線形光学効果を呈することを明らかにした。また分散長についても計算し，4.5 cm よりも短い範囲で光導波路を作れば良いことを明らかにした。これらは有用な知見である。

第 3 章では，ZnO チャネル型光導波路の作製工程について述べている。サファイア基板上に成膜した ZnO 薄膜を光導波路に加工するための条件，特にドライエッチング条件について詳しく述べており，エッチングチャンバー内のガス圧力やガス流量，バイアス電力等とエッチングレートとの関係について調べ，最適エッチング条件を導出している。また，レジストとのエッチング選択比についても検討し，滑らかなエッチング加工を行うためのエッチング条件を導出するなど，有用な知見を得ている。

第 4 章では，作製した ZnO チャネル光導波路の伝搬損失を測定・評価している。カットバック法により伝搬損失を測定した結果，波長 800 nm 帯において 1.5～3 dB/mm という良好な値が得られた。また，伝搬損失の発生要因について解析し，導波路側壁の加工荒さによる光散乱損失がその主因であることをつきとめている。さらに単一モード光ファイバーとの結合損失を評価したところ 8～14 dB/facet であった。このように，ZnO による実用的な光導波路を初めて実現したことは高く評価できる。

第 5 章では，作製した ZnO チャネル光導波路の非線形光学応答特性について測定・評価を行っている。作製した光導波路に約 300 W のピークパワーの超短光パルスを入射させたところ，スペクトル幅が 2 nm 程度であった入射光に対して，出射光のスペクトル幅を約 7 倍の 13.7 nm にまで広げることに成功した。これは約 3π rad の非線形位相シフトが生じたことに相当する。この非線形位相シフトの大きさから，非線形光学定数 n_2 の値を見積もったところ $0.5\sim1.5\times10^{-18}$ m²/W であり，妥当な値であることが確かめられた。さらに光導波路のコアサイズと非線形パラメータとの関係についても調べ，非線形光学効果を最大に引き出すことができる光導波路のサイズについても言及している。これらは極めて重要な知見である。

第 6 章は結論である。

以上要するに本論文は，ZnO を用いたチャネル型光導波路の特性について解析すると共に，その作製手法を確立し，実際に導波路を試作することにより伝搬損失および非線形光学応答特性を明らかにしたもので，非線形光学および光応用分野の発展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。