Noncritically phase-matched second harmonic generation of blue light in Si[O]N–calix[4]arene slab type waveguides

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Abstract

Phase-matching ($TM_0^\omega \rightarrow TM_1^{2\omega}$) devices for generation of blue light using Si[O]N and optically nonlinear calix[4]arene layers are designed, fabricated and tested. The devices show second harmonic peak power of 17 mW at 472.9 nm and 4.5 W at 481.6 nm after 10 mm propagation length of a 1 mm broad beam using fundamental peak power of 200 W and 500 W, respectively. The strong requirements of noncritical phase match are fulfilled, in both design and realization, by the excellent process controllability of the passive Si[O]N waveguides and the insensitivity of the devices towards technologically critical parameters, mainly the calix [4]arene layer thickness.

1. Introduction

Due to the advantages of short wavelength for different applications [1], e.g.: data storage, laser printing, medical application, chemical analysis, etc., the interest and research on efficient compact blue light sources increased strongly within the last years. The final aim of our research consists in second harmonic generation (SHG) of blue light from low cost AlGaAs laser diodes. Planar optical waveguides may be the low cost alternative for expensive frequency doubling crystals [2] and II–VI semiconductor laser diodes [3]. Next to highly efficient second harmonic generation in waveguides of inorganic material, mainly LiNbO₃ [4] and KNbO₃ [5], attention is paid to the use of organic optical nonlinear material. Conversion efficiencies of 1064 nm fundamental light up to $3 \times 10^{-4}\%$ W⁻¹ have been reached with highly nonlinear MNA [6,7]. Furthermore, SH-efficiencies as high as $1.6 \times 10^{-3}\%$ W⁻¹ ($\lambda_\omega = 1064$ nm) and $2 \times 10^{-2}\%$ W⁻¹ ($\lambda_\omega = 926$ nm) have been measured in VDCN-VAc [8] and in

DCANP [9], respectively. In this paper, noncritically phase matched (PM) SHG in tetranitro-tetrapropoxy-calix[4]arenes (calix) in combination with passive LPCVD grown silicon oxynitride waveguides is reported for the first time. Due to a high, stable nonlinear optical coefficient of $d_{33} = 12$ pm/V after corona poling, calix is suitable for SH application [10,11]. Because of transparency of calix into the UV [12,13], the material has the potential for generation of SH light down to 430 nm. Since the layer quality of calix, mainly layer thickness uniformity and reproducibility, is not sufficient for the strong requirements of PM SHG, the devices are designed insensitive towards this parameter. The tuning of the device sensitivity is enabled by introduction of a well defined passive layer into the waveguide structure. In order to fulfil the remaining, high requirements towards this passive waveguide layer, there is need of a thin film material with excellent thickness and refractive index uniformity, process controllability and reproducibility. LPCVD grown silicon oxynitride is known for its excellent properties from

IC and integrated optics application [14,15]. Therefore, we decided for optimization and application of this type of material as passive waveguide in the PM SHG devices.

2. Material properties

Next to process reproducibility and controllability, LPCVD grown silicon [oxy]nitride (Si[O]N) is known for its excellent optical properties [15–17]. These mainly are, high transparency down to UV (low optical losses), flexibility of the refractive index (between $n=1.46$ for SiO₂ and $n=2.0$ for Si₃N₄), wavelength dispersion dependent on the refractive index, excellent uniformity of layer thickness and refractive index across the wafer, etc. For growth of Si[O]N by LPCVD, the processing circumstances (gases, gas flow, temperature, pressure, etc.) can be varied across a broad range. The aim of our work on LPCVD of Si[O]N consisted in optimization of the processing circumstances with respect to the requirements of phase matched SHG [18]. This mainly includes minimization of layer thickness and refractive index non-uniformity.

For our LPCVD set-up, the optimum processing circumstances were found for growth of Si[O]N from SiH₂Cl₂, NH₃ and O₂ at 900°C and 100 mTorr [19]. Thereby, the gas flow of SiH₂Cl₂ and NH₃ is kept constant at 20 sccm and 120 sccm, respectively, and the flow of O₂ is varied between 0 and 5 sccm. In Fig. 1,

Fig. 1. Dependence of the refractive index n and the growth rate R of Si[O]N on the amount of O₂ in the gas flow.

(a)

(b)

Fig. 2. Measurement results and accuracy (error bars) of the thickness non-uniformity δd (a) and the refractive index non-uniformity Δn (b) versus the refractive index n at $\lambda = 632.8$ nm of available Si[O]N layers; the non-uniformities have been measured across an area necessary for SHG application (length 1–2 cm).

the dependence of the growth rate (R) and the refractive index (n) on the oxygen flow is given for the above mentioned processing circumstances. All measurements have been carried out by prism coupling using a wavelength of 632.8 nm. For calculation of the thickness and refractive index non-uniformity, the minimum and maximum values, measured across an area on the wafer of 20 mm long and several mm wide, are considered. The measured thickness non-uniformity (δd) and the refractive index non-uniformity (Δn) in dependence on the refractive index of the film is shown in Fig. 2 (a) and (b), respectively. From Fig. 2a is obvious that for an up to 500 nm thick Si[O]N layer with $n > 1.8$, the thickness deviation across the device area will be less than 1 nm. For a SiON layer with a refractive index around 1.7, this thickness deviation will typically be 3 nm. As it is obvious from Fig. 2b, the refractive index non-uniformity is in the order of the measurement accuracy of the prism coupling method. Therefore, the refractive index deviation will further not be taken into account. The wavelength dispersion behavior of the

Si[O]N waveguides, an important parameter for the design of the devices, has exactly been characterized in the for SHG relevant wavelength region [19]. Furthermore, the optical losses of slab type waveguides applying visible light were determined to be below the detection limit of approximately 0.2 dB/cm. Further, the maximum available layer thickness (d) of the LPCVD grown films is limited to 500 nm for silicon nitride (Si_3N_4) and to approximately 1 μm for silicon oxynitride (SiON) with a refractive index around 1.7.

Calix is an organic material with large hyperpolarisability and suitability for thin film application [12,13]. For thin film deposition, 75 wt.% of calix in PPMA dissolved in chloroform is spin-coated on a substrate [10,13]. Layer thickness uniformity of spin-deposited calix is typically 3%, randomly distributed across the wafer. Further, the maximum available layer thickness is limited to approximately 800 nm. Optical non-linearity is introduced by application of an electric field (corona poling) [10,13]. Poling is carried out for 15 minutes at 100°C applying 6 kV. Further, the layer is cooled down to room temperature, while the high voltage is kept. Due to the highly insulating properties of Si[O]N, calix layers on this type of substrate are insufficiently poled [11]. Therefore, the optical non-linear coefficient of corona poled calix on LPCVD grown Si[O]N is $d_{33} = 2 \pm 1$ pm/V, only.

3. Noncritical design

Two devices, based on phase match between the fundamental TM_0^ω mode and the second harmonic $\text{TM}_1^{2\omega}$ mode, are designed for operation in the 940 to 960 nm fundamental wavelength region. The general layer structure is schematically given in Fig. 3. Clearly,

calix NLO waveguide
SiON / Si_3N_4 passive waveguide
SiO_2 buffer layer
Si-substrate

Fig. 3. Schematic layer structure used for noncritically phase matched SHG devices.

the combined waveguide consists of an optical nonlinear calix layer and a well defined silicon nitride or oxynitride layer. Taking into account relevant material properties and the restrictions with respect to the layer thickness, the sensitivity towards calix and Si[O]N thickness variations of PM devices is calculated as a function of Si[O]N layer thickness. While the fundamental wavelength and the refractive indices of the layers are kept constant, the calix layer thickness is varied in order to achieve phase match ($\Delta n_{\text{eff}} = |n_{\text{eff}}^\omega - n_{\text{eff}}^{2\omega}| \sim 0$). The coherence length l_c of the phase matching device is determined by Δn_{eff} , where $l_c = \lambda_\omega / (4\Delta n_{\text{eff}})$. The field overlap between the fundamental and second harmonic mode at given mode and wavelength is purely defined by the used layer parameters (thickness and refractive indices of the used waveguide material). For effective second harmonic generation, both the coherence length and the field overlap should be high. Anyway, in a two layer system variations of both layers contribute to the Δn_{eff} value, and in our case, the contribution of the layer in which the high field overlap is required is due to technological restrictions more critical. Therefore it might be clear that a high field overlap does not always result in efficient device performance. It is essential to consider the case together with the potential coherence length. As it will be shown, this can result in a compromise in order to achieve higher efficiency on the expense of field overlap. Since silicon nitride layers show next to excellent reproducibility a thickness non-uniformity across 2 cm of less than 0.2%, the design of the first device is based on an Si_3N_4 passive waveguide layer. The simulation results of this device with respect to the PM layer thickness of calix and the sensitivity is shown in Fig. 4. Using silicon nitride, phase match can be achieved at nitride thicknesses below 150 nm (part a) and between 400 and 438 nm (part b). The limitation is given by the maximum calix layer thickness. Part a of Fig. 4 shows phase match at high field overlap, while in part b the field overlap is low due to low evanescent field of fundamental and second harmonic in the calix layer. Further, it should be taken into account that 1 nm thickness variation of silicon nitride is a worst case value, while by 10 nm variation of calix thickness the minimum value is given. The worst case of this variation is not exactly known, but in practice it can be several times higher. Therefore, the lowest sensitivity of Δn_{eff} towards calix thickness variation should

Fig. 4. Phase matching ($TM_0^{\omega} \rightarrow TM_1^{2\omega}$) calix layer thickness (solid line) versus silicon nitride layer thickness using $\lambda_{\omega} = 945$ nm, and the dependence of Δn_{eff} at 10 nm calix and 1 nm silicon nitride thickness (d) away from phase matching thickness (d_{PM}).

Fig. 5. Phase matching ($TM_0^{\omega} \rightarrow TM_1^{2\omega}$) calix layer thickness (solid line) versus silicon oxynitride layer thickness using $\lambda_{\omega} = 958$ nm, and the dependence of Δn_{eff} at 10 nm calix and 3 nm silicon oxynitride thickness (d) away from phase matching thickness (d_{PM}).

always be chosen. According to the high overlap simulation, the best achievable Δn_{eff} with respect to calix thickness variation is more than three orders of magnitude higher than in the lower overlap case. Therefore,

realization of the lower overlap design should be chosen, although on the expense of approximately two orders of magnitude lower overlap. Although this choice is the best approach for this type of device, the conversion efficiency between the fundamental and the second harmonic is expected to be low. In order to increase the conversion efficiency, a second type of device has been designed. This device is optimized with respect to high field overlap, keeping sufficient device insensitivity towards layer thickness variation. For optimization, next to the layer thicknesses, the refractive index of the passive waveguide is varied. The best result was achieved using a silicon oxynitride layer with a refractive index around 1.685. Considering this refractive index and SiON layer thickness within the technological limitation, the corresponding PM calix thickness and the sensitivity of the device towards thickness variations is calculated. The simulation result is shown in Fig. 5. For the same reason as mentioned for the first device, the design with the lowest sensitivity towards the calix layer thickness is chosen. Anyway, in case silicon oxynitride is used, a stronger evanescent

Fig. 6. Field distribution of fundamental and second harmonic mode of device 1 (a) and 2 (b) designed for phase matched SHG.

Table 1

Design and important device parameters of PM SHG devices around 950 nm fundamental wavelength.

device	λ_ω [nm]	d [nm]		n at λ_ω ($\lambda_{2\omega}$)		$\frac{\delta\Delta n_{\text{eff}}}{\delta d \text{ (calix)}}$	$\frac{\delta\Delta n_{\text{eff}}}{\delta d \text{ (Si[O]N)}}$	Overlap ^a [W ⁻¹ m ⁻¹]
		calix	Si[O]N	calix	Si[O]N	[nm ⁻¹]	[nm ⁻¹]	
1	945	700	438	1.563(1.607)	1.983(2.033)	1E-7	2.8E-4	1.8E-4
2	958	645	591	1.563(1.606)	1.672(1.698)	1.4E-6	3.6E-5	1.2E-3

^a Overlap between the fundamental and the second harmonic field in the calix layer.

field is present in the nonlinear layer. The difference in overlapping fields of both devices is obvious from their field distribution shown in Fig. 6. Due to higher overlap, a conversion efficiency of approximately one orders of magnitude higher is expected in case of device 2, while insensitivity towards thickness variation is kept. The parameters chosen for realization of both devices are summarized in Table 1.

4. Device fabrication and testing

The devices are realized on thermally oxidized <100> Si wafers. In case of device 1, on the buffered wafer, silicon nitride is grown during 89 minutes. The layer is characterized ellipsometrically and by prism coupling using a wavelength of 632.8 nm. The centre thickness and refractive indices of the 10 mm long device area are 440.5 ± 0.2 nm and $n_{\text{TE}} = 2.0125 \pm 0.0002$ and $n_{\text{TM}} = 2.0038 \pm 0.0003$, respectively. A thickness deviation of approximately 0.4 nm is measured. In order to realize device 2, silicon oxynitride is grown during 192 minutes using an oxygen

flow of 3.6 sccm. The layer is characterized by prism coupling. A layer thickness of 589.8 ± 0.5 nm and refractive indices of $n_{\text{TE}} = 1.6863 \pm 0.0001$ and $n_{\text{TM}} = 1.6834 \pm 0.0001$ are measured at the centre of the 10 mm long device area. The thickness deviation across this area is 3 nm.

After Si[O]N deposition and measurement, films of 75 wt.% calix in PPMA are spin coated on top of the passive waveguide layers. The calix layer thickness is checked ellipsometrically and by a profilometer. In case of device 1, thicknesses between 700 and 750 nm are measured, a deviation which is considered to be sufficient for the application. In case of device 2, thicknesses between 560 and 640 nm are measured. Then, the calix layers are corona poled over an area including the device area.

For SHG, a frequency doubled *Q*-switched Nd:YAG pumped dye laser with 6 ns pulses at 10 Hz repetition rate is used. The dye laser is tunable within a wavelength range between 910–970 nm. The fundamental TM_0^w mode with a beam width of 1 mm is coupled into the slab type waveguide structure by an SrTiO_3 prism. The prism coupling is schematically shown in Fig. 7.

Fig. 7. Use of prism coupling for measurement of device 1 (a) and 2 (b).

Fig. 8. Measured SH-power of mode match ($TM_0^\omega \rightarrow TM_1^{2\omega}$) for device 1.

Since there is zero field at the calix-air interface in case of device 1 (see Fig. 6), the nonlinear material under the prism is removed in order to achieve sufficient coupling. In case of the second device, this step was not necessary. The fundamental peak power inside the waveguide is determined to be 200 W and 500 W in case of device 1 and 2, respectively. After 10 mm propagation, the fundamental TM_0^ω mode and the generated second harmonic $TM_1^{2\omega}$ mode are coupled out by a second $SrTiO_3$ prism. The fundamental is filtered out, and the SH-intensity is measured by a photomultiplier (device 1) and a photodiode (device 2).

In case of device 1, the wavelength of the dye laser is tuned around 946 nm. The result of this measurement is shown in Fig. 8. A maximum of 17 mW SH output power at 945.77 nm fundamental wavelength is obvious. A conversion efficiency of $4.25 \times 10^{-5} \% W^{-1}$ is calculated. The FWHM of the SH-peak is 0.62 nm. Furthermore, the asymmetric dependence of the SH-peak on the wavelength is remarkable. A steeper decay at the shorter wavelength side is obvious. This dependence can be explained by the thickness deviation of the silicon nitride layer, which might be considered as a parabolic taper with an asymmetric distribution around the phase matching thickness, across the device interaction length. This assumption is in agreement with the measured thickness distribution across the used device area. For measurement of device 2, the wavelength of the dye laser is tuned around 963 nm. The result of this measurement is shown in Fig. 9. The maximum of the SH-peak power is about 4.5 W, measured at the fundamental wavelength of 963.3 nm. This is a conversion efficiency of $1.8 \times 10^{-3} \% W^{-1}$. The SH-peak at

Fig. 9. Measured SH-power of mode match ($TM_0^\omega \rightarrow TM_1^{2\omega}$) for device 2.

FWHM is 3.3 nm, which is mainly due to the SiON thickness deviation. Further, a small distribution due to calix thickness non-uniformity is expected, too. Furthermore, the dependence of the SH-peak on the wavelength is almost symmetric. This indicates a symmetric distribution of the SiON thickness variation around the phase matching thickness, which agrees with the measurement result achieved during characterization of the device area.

5. Conclusion

In conclusion, we have succeeded in realization of waveguide structures consisting of calix and silicon oxynitride for SHG of blue light with a conversion efficiency up to $1.8 \times 10^{-3} \% W^{-1}$. The experimental results agree with the theoretical expectations. For future experiments, there are two main steps: firstly, generation of blue light down to 430 nm, and secondly, efficiency enhancement. In order to realize the first step, devices have been designed using a fundamental wavelength around 870 nm. With respect to the second step, potential for efficiency enhancement is present in several ways. For example, the nonlinear optical coefficient of corona poled calix on Si[O]N should be optimized from 2 pm/V to 12 pm/V. An other possibility is given by the use of channel waveguides instead of slab type waveguides. Thereby, the width of the interacting beam, which reciprocally acts on the SH-output power, can be decreased from 1 mm (laser beam width) to a few μm (channel width). We believe that combining all possible enhancement steps, the conver-

sion efficiency of our devices can be increased several orders of magnitude, and finally, there is potential for SHG from laser diode input using this type of devices.

References

- [1] R.J. Gambino, MRS Bulletin (1990) 20.
- [2] R.F. Belt, G. Glashurov and Y.S. Liu, Laser Focus/Electro-Optics (1985) 110.
- [3] N. Nakayama, S. Itoh, T. Ohata, K. Nakano, H. Okuyama, M. Ozawa, A. Ishibashi, M. Ikeda and Y. Mori, Electron. Lett. 29 (1993) 1488.
- [4] X. Cao, B. Rose, R.V. Ramaswamy and R. Srivastava, Optics Lett. 17 (1992) 795.
- [5] D. Fluck, B. Binder, M. Küpfer, H. Looser, Ch. Buchal and P. Günter, Optics Comm. 90 (1992) 304.
- [6] O. Sugihara, T. Kinoshita, M. Okabe, S. Kunioka, Y. Nonaka and K. Sasaki, Appl. Optics 30 (1991) 2957.
- [7] O. Sugihara and K. Sasaki, J. Opt. Soc. Am. B 9 (1992) 104.
- [8] Y. Azumai, M. Kishimoto and H. Sato, Japan. J. Appl. Phys. Part 1 31 (1992) 1358.
- [9] M. Flörsheimer, M. Küpfer, Ch. Bosshard, H. Looser and P. Günter, Advanced Mater. 4 (1992) 795.
- [10] O.F.J. Noordman, N.F. van Hulst and B. Bölger, J. Opt. Soc. Am. B, to be published.
- [11] O.F.J. Noordman, K. Wörhoff and N.F. van Hulst, proceedings of ECIO '95, Delft, The Netherlands, p. 89.
- [12] E. Kelderman, G.J.T. Heesink, L. Derhaeg, T. Verbiest, P.T.A. Klaase, W. Verboom, J.F.J. Engbersen, N.F. van Hulst, K. Clays, A. Persoons and D.N. Reinhoudt, Advanced Matter. 5 (1993) 925.
- [13] G.J.T. Heesink, N.F. van Hulst, B. Bölger, E. Kelderman, J.F.J. Engbersen, W. Verboom and D.N. Reinhoudt, Appl. Phys. Lett. 62 (1993) 2015.
- [14] F.H.P.M. Habraken, ed., Research Report ESPRIT, Project 369, Volume 1 (Springer Verlag, Heidelberg, 1991).
- [15] W. Gleine, Ph.D. thesis, University of Hamburg-Harburg, VDI Verlag, Düsseldorf, 1990.
- [16] Yi-Ming Xiong, P.G. Snyder, J.A. Woollam, G.A. Al-Jumaily, F.J. Gagliardi and L.J. Mizerka, Surf. and Int. Anal. 18 (1992) 124.
- [17] L. Zambov, G. Peev and V. Shanov, Vacuum 43 (1992) 227.
- [18] K. Wörhoff, O.F.J. Noordman, N.F. van Hulst, H. Albers and P.V. Lambeck, Technical Digest CLEO Pacific/Rim '95, July 10-14 1995, p. 199, Chiba, Japan.
- [19] K. Wörhoff, H. Albers, P.V. Lambeck, N.F. van Hulst and Th.J.A. Popma, to be published.