Strong Second-harmonic generation in PZT thin films

Gilles F. Feutmba*[a,b,c], I. Luntadila Lufungula*[b,c], A. Hermans[b,c] , John P. George[a,c], Tessa Van de Veire[b,c], Bart Kuyken[b,c], Dries Van Thourhout[b,c] and Jeroen Beeckman[a,c]
[a] Liquid Crystals and Photonics Group, ELIS Department, Ghent University, Ghent B-9000, Belgium;
[b] Photonics Research Group, INTEC Department, Ghent University-imec, Ghent B-9000, Belgium;
[c] Center for Nano- and Biophotonics (NB-Photonics), Ghent University, Ghent B-9000, Belgium;
*gillesfreddy.feutmba@ugent.be; phone +32 465 27 78 81

ABSTRACT

Second-order nonlinear processes enable a wide range of applications in research, spectroscopy, microscopy, quantum computing and fiber-optic communication. However, the majority of available second-order nonlinear devices rely on bulk nonlinear crystals and free-space optical components. By exploiting the advancements made in integrated optics, CMOS-compatible materials with large second order nonlinearity could enable on chip nonlinear devices at small cost. However, silicon and silicon nitride which are the most used materials in the silicon-on-insulator platform for photonic integrated circuits exhibit negligible second order nonlinearity and alternate materials have to be investigated. We demonstrate here strong SHG from PZT thin films grown on glass substrates. The nonlinear response of the film is calibrated against a BBO crystal. The dominant tensor component is found to be 60 pm/V with the film poled out of plane. This opens potential and avenues for highly efficient on chip nonlinear devices.

Introduction

Second order nonlinear effects have generated numerous possibilities in different sectors of applications and fields such as research, spectroscopy, microscopy, quantum computing and fiber-optic communication. A lot of devices used in our daily routines are based on second order nonlinear effects. Green lasers, optical parametric oscillators and amplifiers (sold by most laser manufacturers), optical quantum computing, terahertz radiation sources and electro-optic modulators just to name a few, are all examples of devices that rely on second order nonlinear effects such as Second Harmonic Generation (SHG), difference-frequency generation, parametric down-conversion and Pockels effect. However up to date, the majority of these available second-order nonlinear devices rely on bulk nonlinear crystals and free-space optical components.

Silicon (Si) and silicon nitride (SiN) are widely used materials in the silicon-on-insulator (SOI) platform for Photonic Integrated Circuits (PICs) and integrated opto-electronics. By leveraging CMOS infrastructure, smaller, faster, robust and energy-efficient optical devices can be made by integrating a huge amount of bulky and expensive free space optical components. The complexity and functionality of these PICs could greatly be extended by integrating the aforementioned nonlinear devices on Si/SiN. Unfortunately, Si suffers from two-photon absorption, limiting its efficiency in nonlinear applications. Moreover, Si has a centrosymmetric structure and exhibits negligible second-order nonlinear response. Although attempts have been made to break the symmetry of Si by engineering strain fields on silicon waveguides using silicon nitride and enhance the nonlinear response to ~40pm/V [1], the SHG conversion efficiency remains low for nonlinear device applications. SHG has been observed in SiN films and waveguides [2,3] with an effective second-order susceptibility of $\chi^{(2)}$ = 5 pm/V, which is still about two orders of magnitude lower than traditional nonlinear crystals.

Alternative CMOS-compatible materials with large second-order nonlinearity have been investigated as an attempt for on chip optical nonlinear devices. Low-voltage integrated $LiNbO_3$ Pockels based modulators operating at 1550 nm and SHG have been demonstrated [4]. However, the use of thin film $LiNbO_3$-on-insulator requires expensive bonding procedures. Thin films of Lead Zirconate Titanate (PZT), another ferroelectric material has been successfully grown on SiN [5] and Si [6] waveguides for high speed electro-optic modulation by exploiting the Pockels effect and stands as a good candidate for on chip nonlinear devices. In this paper, we demonstrate strong SHG from PZT thin films grown on glass substrates by chemical solution deposition. The nonlinear response of the film is calibrated against a BBO crystal. The PZT films are poled prior to the nonlinear measurements. The dominant tensor component $\chi^{(2)}_{zzz}$ is found to be (60±10) pm/V (averaged over three samples) with the film poled out of plane.

Setup and Measurement

The PZT thin films are grown by chemical solution deposition (CSD) using a lanthanide-based intermediate layer [7]. The PZT films are grown on a 30 nm thick ITO layer deposited on a 1.1 mm thick Corning glass. Samples with different PZT thicknesses are fabricated. A through PZT film via was made by ultrasonic soldering for contact with the bottom ITO layer. Copper strips are rubbed onto the top surface of the PZT film to serve as contact. The PZT film is polycrystalline with an out-of-plane preferential orientation. To maximize the nonlinear response, the samples are poled prior to SHG measurements. Different poling procedures are performed. After poling, the copper strips are removed and the SHG measurement performed with the light beam passing through the poled region. For the SHG characterization, a mode-locked laser (Calmar FPL-03CCFPM) operating at 1550 nm and emitting 0.1 ps pulses is used. A polarizing beam splitter cube (PBSC) is used to maintain the linear polarization of the collimated light beam and a half-wave plate is used to rotate the polarization. The rotation of the half wave plate is controlled by a motorized stage and allows for polarization-dependent measurements. Two parabolic mirrors are used to focus the laser beam on the sample and to collimate it again after passing through the sample. A long pass filter is placed before the sample to filter out any SHG light generated before the sample and a short pass filter positioned after the sample suppresses the laser light at the fundamental wavelength and allow only the SH light through. A second PBSC positioned before the detector is used as an analyzer. A visible single photon detector (ID120) is used for detection. A lens is placed in front of the detector to compensate for beam displacement induced by the rotation of the sample. The set-up is calibrated with a BBO crystal [8].

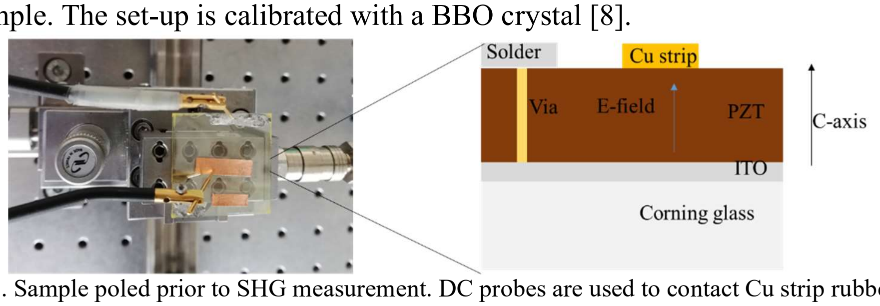

Figure 1. Sample poled prior to SHG measurement. DC probes are used to contact Cu strip rubbed on PZT surface and a solder relaying bottom ITO layer. Sample poled out of plane in direction of C-axis (Z axis).

The PZT thin film has a random in plane polarization and a $C_{\infty v}$ symmetry with respect to the normal.

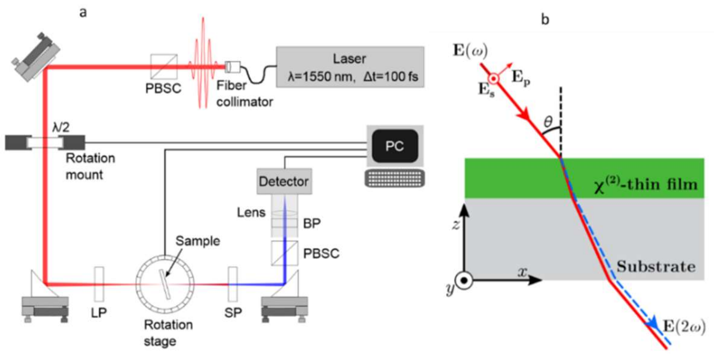

Figure 2. **a** Set-up used for SHG measurements. PBSC, Polarization beam splitter cube; λ/2, Half-wave plate; LP, low-pass filter; SP, Short pass filter; BP, Band pass filter. Infrared light (in red) passes through sample and the Second harmonic (in blue) is recorded. **b** SH measurement principle; The angle of incidence is fixed and SHG signals measured as a function of the rotation angle of half-wave plate that modulates the incident polarization state.

The nonzero second order susceptibility tensor components for SHG are $\chi^{(2)}_{xxz} = \chi^{(2)}_{yyz} = \chi^{(2)}_{xzx} = \chi^{(2)}_{yzy}$, $\chi^{(2)}_{zxx} = \chi^{(2)}_{zyy}$ and $\chi^{(2)}_{zzz}$, where z is the film normal and x and y are the two orthogonal in-plane directions (figure 2b). For measurements performed on PZT samples, the angle of incidence was set at 70°. The fundamental polarization state was modulated by rotating the half-wave plate and the SH recorded as a function of rotation angle. To fit the measurement data and extract the different tensor components, the model described in [9] was implemented in MATLAB. To get better accuracy, the measurement data for both SH polarizations (s and p) are fitted simultaneously.

Results

Three samples with PZT layer thicknesses 885nm (S1), 920.7nm (S2) and 871.9nm (S3) are characterized. Figure 1 shows data fitting of BBO used for calibration and SHG measurements on PZT samples. The first SH measurements on PZT samples are performed without poling the samples. The results show non-zero response which confirms that the crystallites have a net preferential out-of-plane orientation after growth. The nonlinear response is the cumulative response from the individual domains in the PZT thin film. The results on poled samples show an increase in $\chi^{(2)}_{zzz}$ component with increasing poling strength with a maximum average of (60±10) pm/V reached after poling with 13V/µm. The breakdown field appears to be 15V/µm. The results show that the samples can either be poled with a positive or negative voltage after deposition. Although only absolute values of the second order susceptibility tensor components for SHG are extracted here, this is an indication that the sign of these tensor elements can flip with poling unlike $LiNbO_3$ film where the cut sets a fixed crystal orientation. The table below summarizes the extracted tensor components. The $\chi^{(2)}_{zzz}$ value reported here is substantially higher than values previously reported. The value opens an avenue for on-chip second order nonlinear optics using PZT.

	Sample 1 (S1)	Sample 2 (S2)	Sample 3 (S3)
$\chi^{(2)}_{zzz}$	61.06	60.2	63.85
$\chi^{(2)}_{xxz}$	3.33	2.58	3.35
$\chi^{(2)}_{zxx}$	1.53	0.3217	4.95

Table1. The absolute calibrated values of second order susceptibility tensor components in the PZT film

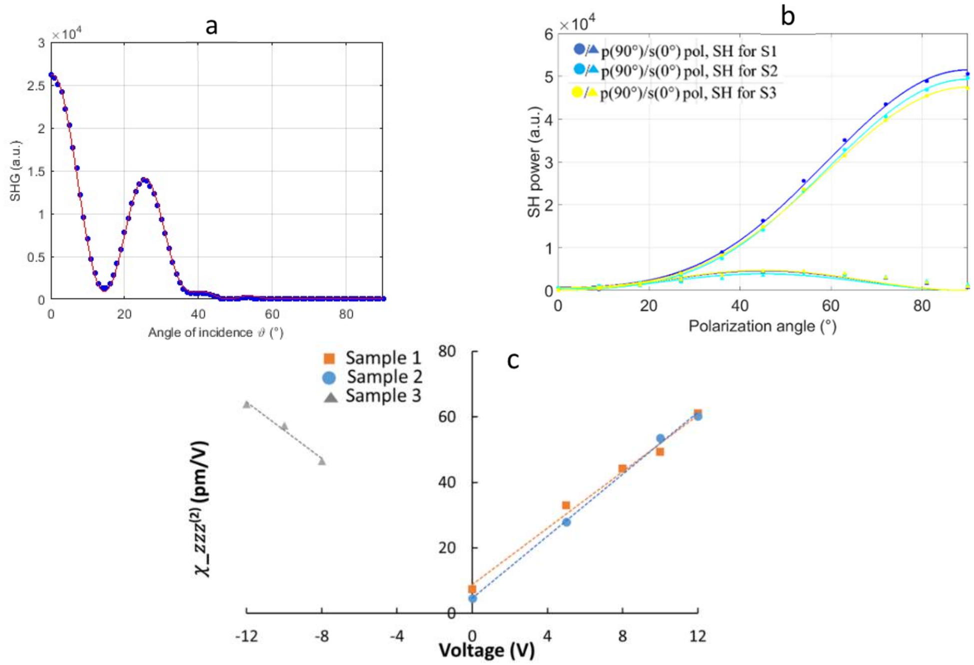

Figure 3: **a.** Measurement and fit for a BBO crystal with known thickness and parameters. Used for calibration. **b.** p-(circles) and s- (triangles) polarized SH power and corresponding fitting curves as a function of the polarization angle obtained from three different samples (S1, S2, S3) poled with 12V for 10 min. The angle of incidence is fixed at 70°. **c.** $\chi^{(2)}_{zzz}$ component increases with increasing with poling voltage. The maximum response of the thin film is obtained when the crystallites are fully aligned in the out-of-plane direction.

Further experiments are planned to better understand the effect and stability of poling over SHG and the $\chi^{(2)}_{zzz}$ sign change. The PZT film can also be poled in plane with a resultant C-axis in plane. Ongoing experiments will help to extract the in-plane second order susceptibility tensor components.

Gilles F. Feutmba acknowledges support and funding as an SB-PhD Fellow of the research foundation–Flanders (FWO 179475/1S68220N -CDR).

References

[1] M. Cazzanelli et al., "Second-harmonic generation in silicon waveguides strained by silicon nitride" Nature Mater. 11, 148 (2011).
[2] J. S. Levy et al., "Harmonic generation in silicon nitride ring resonators" Opt. Express 19, 11415 (2011)
[3] Ning et al., "Strong second-harmonic generation in silicon nitride films", Appl. Phys. Lett. 100, 161902 (2012)
[4] Andreas Boes et al., "Status and Potential of Lithium Niobate on Insulator (LNOI) for Photonic Integrated Circuits"; Laser Photonics Rev. 12, 1700256 (2018)
[5] Alexander, Koen, et al. "Nanophotonic Pockels modulators on a silicon nitride platform." *Nature communications* ***9,*** *3444 (2018)*.
[6] Gilles F. Feutmba et al. "Hybrid PZT/Si TM/TE electro-optic phase modulators", photonics west (2019)
[7] J.P. George, et al. "Lanthanide-Assisted Deposition of Strongly Electro-optic PZT Thin Films on Silicon: Toward Integrated Active Nanophotonic Devices", Acs Appl Mater Inter, 7 (2015) 13350-13359.
[8] A. Hermans et al., `` On the determination of χ(2) in thin films: a comparison of one-beam second-harmonic generation measurement methodologies´´, scientific reports, 7, p.44581 (2017).
[9] D. S. Bethune, "Optical harmonic generation and mixing in multilayer media: extension of optical transfer matrix approach to include anisotropic materials," J. Opt. Soc. Am. B, vol. 8, 367, 1991.