8th European Conference on Applications of Polar Dielectrics (ECAPD '8)
Metz/France, September 2006, paper ECAPD '8 – 161006

Depth-resolved analysis of ferroelectric domain structures in Ti:PPLN waveguides by nonlinear confocal laser scanning microscopy

GERHARD BERTH,[1] VIKTOR QUIRING, WOLFGANG SOHLER AND ARTUR ZRENNER

Department of physics, Universität Paderborn, Warburger Str. 100, D-33098 Paderborn, Germany

We have analyzed the ferroelectric domain structure of periodically poled $LiNbO_3$ by nonlinear confocal laser scanning microscopy. Annealed samples exhibit domain wall width below 300 nm, which more or less corresponds to the diffraction limit. In contrast, domain walls in "as poled" samples appear considerably broader and tentatively structured. In periodically poled Ti:$LiNbO_3$ waveguide structures we find a pronounced enhancement of the nonlinear signal from the domain walls, which appears within the Ti-diffused range of the waveguide including the surface region.

Keywords: Ferroelectric domain structures, nonlinear microscopy, Ti:$LiNbO_3$ waveguides

1. INTRODUCTION

Due to its outstanding properties the ferroelectric crystal lithium niobate ($LiNbO_3$) is widely used in the field of nonlinear and integrated optics. Its piezoelectric, electrooptic and nonlinear properties are applied in integrated optical devices. Of particular interest here is periodically poled lithium niobate (PPLN), which allows for quasi-phase matching (QPM) and corresponding applications in the area of frequency conversion (e.g. second harmonic generation SHG, sum frequency generation, optical parametric oscillation) [1,2]. Crucial for these applications is the control of domain fabrication for high quality devices. The area of application for such periodic structures substantially depends on sharpness, homogeneity, depth expansion (overlap wave-prominent range), periodicity, and homogeneity of the periodic ferroelectric domain structure.
In the current contribution we present our investigations on PPLN samples fabricated by

[1] Corresponding author. E-mail: berth@physik.upb.de

electric field assisted poling with structured electrodes. For our studies we use nonlinear confocal laser scanning microscopy (CLSM), a method which allows for a direct and non invasive study of the domain walls with high resolution in 3 dimensions [3].

2. SAMPLE FABRICATION

Congruent Z-cut $LiNbO_3$ wafers (from Crystal Technology, Inc.) have been used for sample preparation. Optical waveguides have been fabricated on the –Z-face, along the crystallographic X-axis, by indiffusion of photo lithographically defined Ti-stripes of 7 µm width and 98 nm thickness. The diffusion was performed at ~1060 °C for 7 h in an argon atmosphere, with subsequent postdiffusion at the same temperature for 1h in oxygen, in order to reoxidise the sample completely. To periodically pole $LiNbO_3$, a photoresit grating with a period of 16.9 µm was photo lithographically defined on the surface. Liquid electrolyte electrodes (DI-water/LiCl) enabled electric field assisted poling of the ferroelectric domain structure. A microscope image of the Ti:PPLN –Z-face and a schematic presentation of the sample geometry are shown in Fig. 1a) and 1b). The indicated coordinate system corresponds to the crystallographic axes.

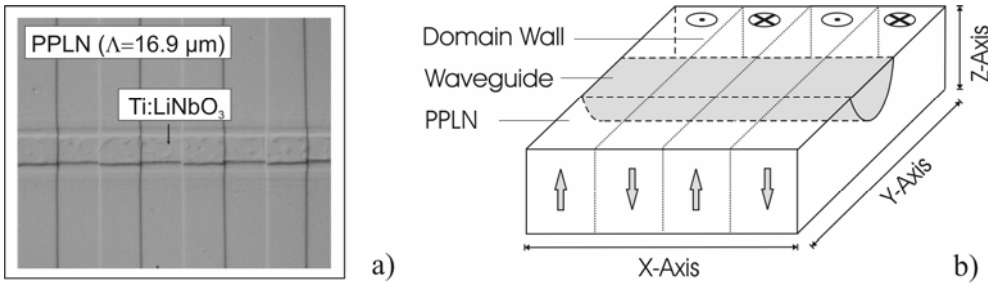

Figure 1 a) Optical micrograph of the surface of a Ti:PPLN waveguide. The specimen has been selectively etched (HF/HNO_3) to reveal the domain structure of 16.9 µm periodicity. b) Schematic of the investigated structure geometry. The domain dipoles are indicated by arrows.

After fabrication the single mode waveguides were characterized in terms of mode intensity distribution and propagation losses at 1550 nm wavelength. The propagation losses of the modes were determined by the Fabry-Perot method [4] to be less than 0.05 dB/cm.

3. EXPERIMENTAL SETUP

For the nonlinear studies a confocal laser scanning microscope was developed. Our special setup depicted in figure 2 allows for microscopy in reflection and transmission mode. In both cases infinity-corrected microscope objectives (Leica N-Plan 100x/0.9/0.27 or Leica PL-Fl-L 100x/0.75/4.7) were used to focus the incoming laser beam to a diffraction-limited spot.

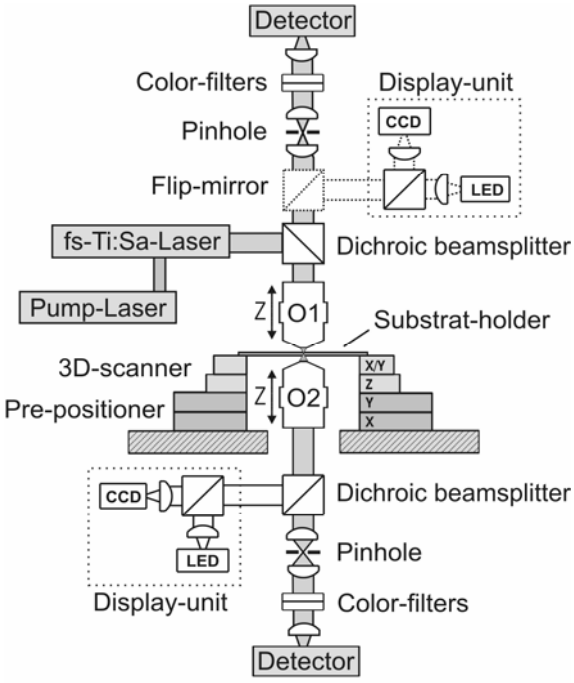

Figure 2 Schematic of our experimental setup of the CLSM. O1, O2: Infinity-corrected objectives. A complex system of positioning-units (3D-scanner, Pre-positioner) allows analyzing areas of up to 5" x 5" in size.

Light detection can be accomplished either in reflection (objective-1) or transmission (objective-2) geometry. For spatial filtering in the confocal imaging mode a pinhole of typically 2 µm diameter is used. Most of the information which does not originate from the focal plain of the microscope objective is suppressed by this arrangement [5]. Light from the focal plane passes the pinhole and is detected. A confocal microscope setup therefore allows for a depth-dependent optical tomography. Image acquisition in 3 dimensions is accomplished by scanning the sample via closed loop nano-positioners with respect to a fixed laser focus. In linear mode the spatial resolution of our confocal microscope is ~300 nm in the transverse and ~500 nm in the axial direction.
For the nonlinear confocal microscopy a mode-locked Ti:sapphire laser (Femtosource C-20) with pulse duration of 20 fs at a repetition rate of 80 MHz and a center wavelength of λ=800 nm was used for optical excitation. By means of an autocorrelation setup we

determined the pulse width after passing through the objective O1 to be about 100 fs. Under these conditions (average power on the sample ~100 mW) the samples were probed with a peak pulse power density of the order of 1 x 10^{12} W/cm^2. The separation of the excitation light and the frequency-doubled signal was accomplished by dichroic beam splitters (Layertec FS 101495). In addition color glass filters (BG39) were used. The detection of the nonlinear signal was done by a single photon counter (SPCM-AQR) utilizes a silicon avalanche photodiode.

4. RESULTS AND DISCUSSION

In our investigations we have concentrated on differently processed PPLN specimens and different measurement geometries. In previous work [6] nonlinear microscopy for ferroelectric domain wall visualization has been performed in reflection geometry. Up to now, however, the mechanisms for second harmonic generation (SHG) light emission in this particular geometry remains unclear. One of the most basic experiments in the context of nonlinear microscopy is therefore a comparison between the SHG emission in reflection and transmission mode, which can be performed experimentally with our special microscope setup. For experiments in transmission geometry the crystals were also backside polished. Figure 3 shows typical second harmonic (SH) images of area scans of the PPLN surfaces.

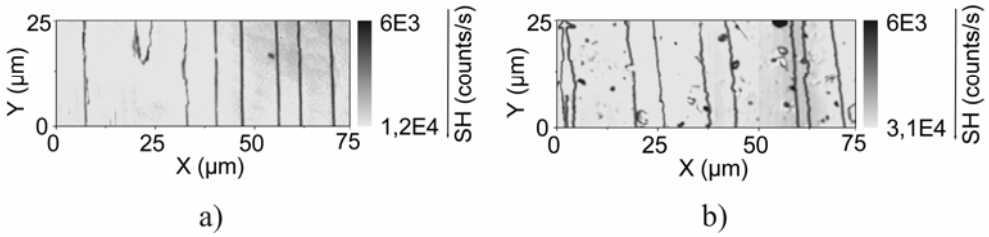

Figure 3 SH area-scans from the surface (Z-face) of PPLN in reflection (a) and transmission (b) mode with a scan increment of 250 nm.

Dark lines indicate the domain walls with reduced intensity of the SH signal. In both modes the second harmonic surface signal from the domain walls appears reduced with respect to the domains. Comparing the detected intensity levels of the SH signals in reflection and transmission geometry reveals that the transmission mode provides higher intensity and contrast. Nevertheless there is also a detectable SH signal generated in backward direction.

In our case we consequently expect that SHG appears mainly in collinear direction to the fundamental wave. For a backward-propagating SH wave, the wavenumber mismatch is $\Delta k_{ba} = k_{SH} + 2k_F$ and for the forward-propagating SH wave $\Delta k_{fo} = k_{SH} - 2k_F$, where k_{SH} is the wavenumber of the SH wave and k_F the wavenumber of the

fundamental wave. The coherence lengths of both processes are given by

$$L_C = \frac{\pi}{\Delta k}, \text{ with } \Delta k_{ba} = \frac{2\pi}{\lambda_{SH}}\left(n(2\omega) + n(\omega)\right) \text{ or } \Delta k_{fo} = \frac{2\pi}{\lambda_{SH}}\left(n(2\omega) - n(\omega)\right)$$

respectively. The coherence length of the backward-propagating SH wave ($L_{C,ba}$ ~ 43 nm) is much smaller as compared to the collinear direction ($L_{C,fo}$ ~ 1µm). For nonlinear microscopy operation in transmission mode is therefore clearly favourable because of the longer coherent interaction length. In addition, chromatic errors of the objective lenses (800 nm vs. 400 nm) are irrelevant, because excitation (fundamental wave) and detection (SH wave) can be focused independently.

If we change the propagation direction of excitation and detection from Z-face to Y-face, the SH intensity increases to a higher level (see figure 4). It's well known that the SHG contrast depends on the direction of propagation with respect to the crystal orientation and the corresponding nonlinear coefficients [7].

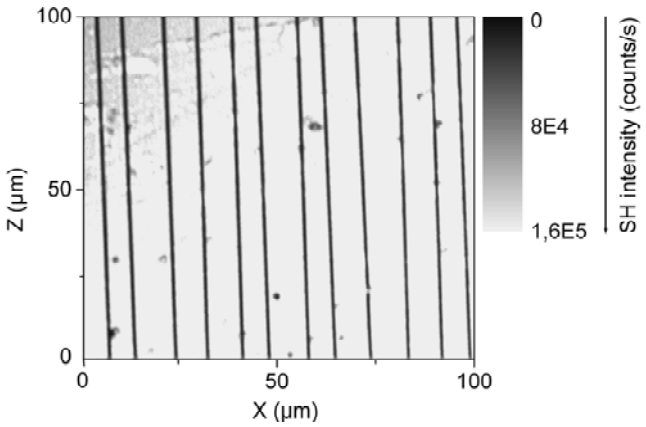

Figure 4 Second harmonic area-scan from the surface (Y-face) of PPLN in reflection mode with a scan increment of 250 nm. Dark lines represent here the walls with high contrast in the SH signal.

For all investigated surface orientations the SH signal from the domain walls is reduced in relation to the domain signal. Annealed samples (2h, 120°C) exhibit apparent "domain wall width" of less than 300 nm, which corresponds more or less to the spatial resolution of our technique.

In contrast to the behavior of the surface signal, a different SH signature is found inside the crystal. Here the nonlinear signal from the domains is reduced with respect to the wall signal. An example for a sub-surface PPLN signal is shown in Figure 5, where the bright lines represent the domain walls. The observed inversion of the contrast of the domain walls was verified by systematic depth scans.

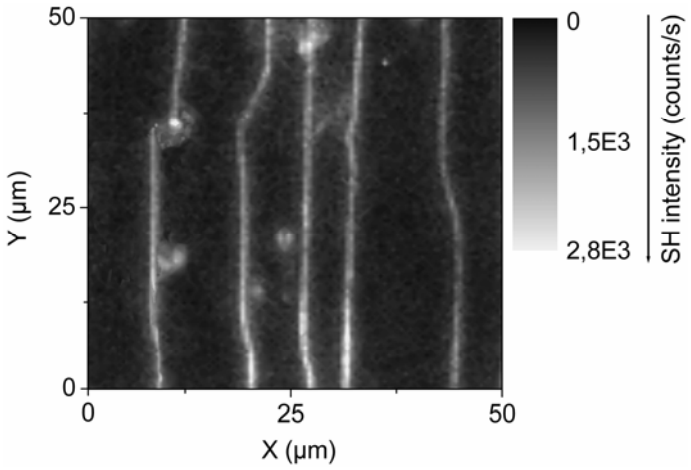

Figure 5 Second harmonic area-scan of the X/Y-plane 20µm below the PPLN surface-region in reflection mode.

Furthermore “hexagonal defects” with a specific orientation with respect to other defects and the crystal structure can be observed [8].
To learn more about the physical character of the domain walls, poled samples have been investigated also prior to sample annealing. In both cases (annealed and as grown) within the surface region the same wall structure was observed. Inside the crystal, however, we found a different behaviour. Figure 6 shows an X/Y-scan in a depth of 20 µm. Bright lines represent the domain walls.

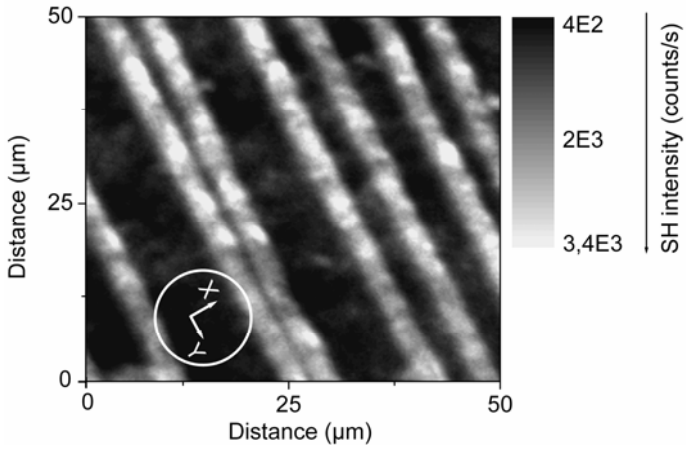

Figure 6 SH X/Y-scan of a PPLN-plane 20 µm below the surface done prior of sample annealing (reflection mode, scan increment = 250 nm).

In contrast to annealed samples, here, the wall signal inside the crystal shows a spatial broadening with an apparent “wall width” of about 3 µm. Studies of the domain distribution of PPLN using scanning nonlinear dielectric microscopy have shown, that a strong residual stress or an internal electric field substantially reduces the nonlinear

dielectric constant of $LiNbO_3$ [9]. Based on these facts, we commend annealing of PPLN structures to improve the efficiency of QPM.

In addition we have performed also a nonlinear analysis of the ferroelectric domain signals in Ti:PPLN. In contrast to bulk PPLN, we find in Ti:PPLN waveguide structures a strong and distinct enhancement of the nonlinear signal from the domain walls within the surface region, peaked at a depth of ~ 3 µm (see figure 7). The nonlinear contrast of the domain walls is found to be inverted and clearly enhanced within the surface region as a consequence of Ti incorporation.

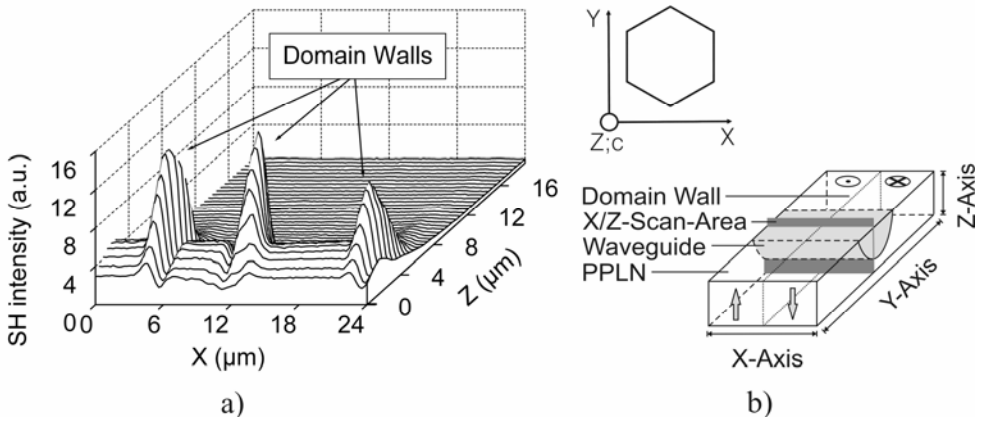

Figure 7 a) SH depth scan across a waveguide in the X/Z-plane of Ti:PPLN. b) Geometry of the scan environment. The indicated coordinates corresponds to the crystallographic axes. (X: Line-scans along the waveguide across walls, Z: Focus was moved downwards in the waveguide)

The depth resolved studies show that the observed enhancement in fact appears throughout the whole Ti-diffused range of the waveguide. Furthermore we find, that the nonlinear response around the domain wall no longer appears as a symmetric peak. This is shown more in detail in figure 8, were a line-scan at maximum enhancement (3 µm depth) and a contour plot of the whole depth scan are shown.

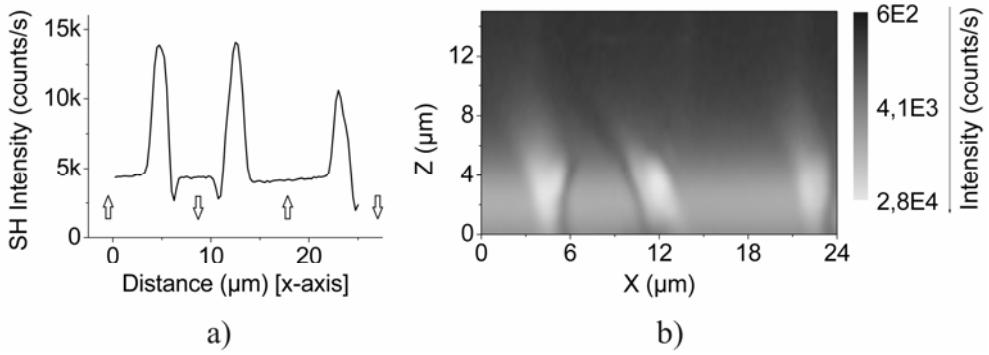

Figure 8 SH scans of Ti:PPLN structures in reflection mode. a) X-line-scan in a depth of 3µm (domain dipoles are indicated by arrows). b) Contour plot of the X/Z depth scan.

Domains of different orientation show also different behaviour of the SH signal at the domain boundaries. In similar structures it has been found by selective etching, that (+)-domains are tentatively narrower that (-)-domains. Based on this assignment we have indicated the domain orientations for our present experiment in figure 8a. In the transition region from (+)-domains to (-)-domains we always find a SH signal which consist of a sharp dip followed by the strong enhancement peak. The domain walls therefore appear no longer as sharp features, but as broader, structured regions. Since the poling procedure is performed after the indiffusion of Ti, we tentatively conclude, that the observed behaviour might be linked to peculiarities of the internal electric field distribution at the domain boundaries. This hypothesis is supported by the long range character of the observed phenomenon and by the fact, that structural modifications during the final annealing procedure (at 120 °C) seem very unlikely. A more detailed analysis of this phenomenon will be subject of further systematic investigations.

5. CONCLUSION

Summarizing, we have analyzed the ferroelectric domain structure of PPLN by nonlinear CLSM with excitation by a fs-Ti:Sa-laser and detection of the frequency-doubled signal. It has been shown that CLSM allows for a direct, depth-resolved, and non invasive study of the domain walls. In our investigations we have analysed PPLN structures by nonlinear CLSM as a function of depth in reflection and transmission mode. The nonlinear analysis has shown that in both cases the second harmonic surface signal from the domain walls is reduced in relation to the domain signal. Inside the crystal the opposite effect is observed. From a comparison of both microscopy modes we consequently expect that second harmonic generation is mainly in collinear direction to the exaction. Furthermore both annealed PPLN-samples and “as poled” specimens have been investigated. Annealed samples exhibit domain wall width of less than 300 nm, domain walls in “as poled” PPLN appear considerably broadened and show a specific sub-structure, which might be linked to preferential crystallographic orientations. In Ti:PPLN waveguide structures we find a strong and distinct enhancement of the nonlinear signal from the domain walls. Depth resolved studies have shown that the observed enhancement appears within the Ti-diffused range of the waveguide including the surface region.

ACKNOWLEDGMENT

We express sincerer thanks to Dr. H. Suche, Dr. H. Herrmann, Dipl. Phys. S. Orlov (Universität of Paderborn, Germany) and V. Gopalan (Pennsylvania State University, USA) for useful discussions on ferroelectric materials. The authors would also like to acknowledge financial support by the DFG research unit 295.

REFERENCES

1. Fejer M.M.: Nonlinear Optical Frequency Conversion. Physics Today. 1994; 47: 25.
2. Myers, E., Miller G. D., Eckardt , Fejer M. M., Byer ., R. L., Bosenberg W. R.: Quasi-phase-matched 1.064-mm-pumped optical parametric oscillator in bulk periodically poled LiNbO3. Optics Letters. 1995; 20:1
3. Flörsheimer M., Paschotta R., Kubitscheck U., Brillert Ch., Hofmann D., Heuer L., Schreiber G., Verbeek C., Sohler W., Fuchs H.: Second-harmonic imaging of ferroelectric domains in LiNbO3 with micron resolution in lateral and axial directions. Applied Physics B. 1998; 67: 593-599.
4. Regner R., Sohler W.: Loss in Low-Finesse Ti:$LiNbO_3$ Optical Waveguide Resonators. Applied Physics B. 1985; 36: 143-147.
5. Sheppard C. J. R., Shotton D. M.: Confocal Laser Scanning Microscopy. Oxford: IOS Scientific Publishers Ltd.; 1997.
6. Rosenfeldt A., Flörsheimer M.: Nondestructive remote imaging of ferroelectric domain distributions with high three-dimensional resolution. Applied Physics B. 2001; 73: 523.
7. Yariv A.: Quantum Electronics. New York: Wiley; 1989
8. Jungk T., Soergel E.: Depth-resolved analysis of ferroelectric domain structures in bulk $LiNbO_3$ crystals by scanning force microscopy. Applied Physics Letters. 2005; 86: 242901.
9. Cho Y., Kazuta S., Ito H. PA: Scanning-nonlinear-dielectric microscopy study on periodically poled $LiNbO_3$ for a high-performance quasi-phase matching device. Applied Physics Letters. 2001; 79: 2955-2957.