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Imaging of the Ferroelectric Domain Structures by Confocal Raman Spectroscopy

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Confocal Raman spectroscopy was performed as an archetype imaging method to study the ferroelectric domain structure of periodically poled lithium niobate. More precisely, the linkage out of spatial resolution and spectral information proved itself as very useful. Here a specific modulation of the Raman lines by the local variation of polarity and a non-symmetric measuring-signal across the domain structure were found, which allows for imaging of domain boundaries as well as oppositely orientated domains. The high potential of this method is demonstrated by the visualization of the ferroelectric domain structures based on various phonon modes.

Keywords Raman spectroscopy; ferroelectric domains; $LiNbO_3$; confocal imaging

1. Introduction

Nonlinear optical effects play a major role in the field of photonics and integrated optics. A typical example for commercial applications of such phenomena in nonlinear devices is frequency conversion (e.g. sum- and difference-frequency generation, optical parametric amplification and oscillation) [1, 2]. Here a critical parameter is the phase matching, which determines the magnitude of the constructive interaction of involved optical fields. Thus, to achieve high efficiency of such processes, the dispersion for the involved frequencies has to be compensated. For nonlinear components based on optical waveguides one uses the principle of quasi-phase matching (QPM) that micro domain inversion takes advantage of. Here in average rapid phase-changes arising at domain boundaries compensate the various phase velocities of the involved light waves. The efficiency of related processes is largely determined by the homogeneity, sharpness and depth extension of such domain structures. In this context the confocal Raman spectroscopy is a powerful analytic tool for the control of the transferred domain structure, which is extremely sensitive to structural and compositional changes [3, 4].

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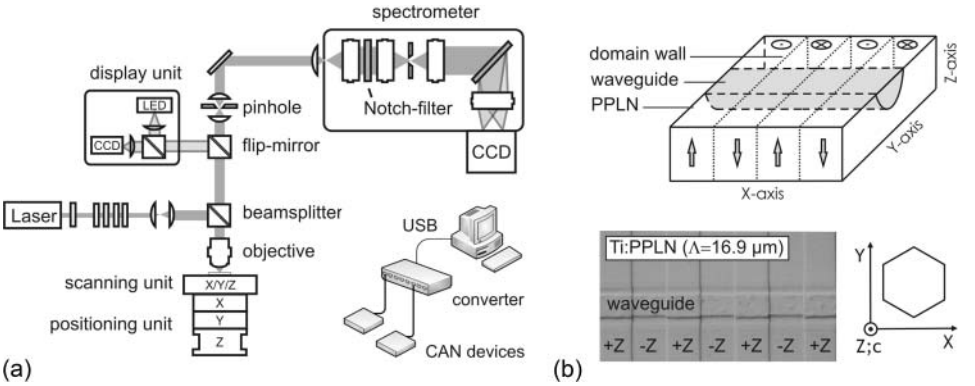

Figure 1. a) Scheme of the modular-engineered confocal Raman setup. b) Schematic of the structure geometry and micrograph of the Z-surface of a Ti:PPLN sample.

2. Experimental

We have applied confocal Raman spectroscopy for imaging and characterization of the ferroelectric domain structure in periodically poled lithium niobate (PPLN). The utilized confocal Raman setup is pictured schematically in Fig. 1. For the optical excitation of phonon modes an infinity-corrected objective (Mitutoyo OBJ Plan Apo SL 100x) focuses the laser light (DPSSL: 532 nm, single line) to a diffraction-limited spot on the sample. The inelastic scattered light originating from a confined detection volume is analyzed by a high-throughput single stage spectrometer (KOSI Holospec f/1.8i) with adapted CCD camera (Andor Newton, BI). The spectrometer includes a holographic transmission grating which offers very short acquisition times as required for tomographic imaging. Suppression of the elastic scattered light is realized by an appropriate notch filter. For spatial filtering a confocal pinhole is placed in front of the detection-unit. Information that does not originate from the focal plane is faded out, thus, solely signal from the focal plane passes the confocal pinhole. Here the confocal mode enables a spatial resolved spectroscopy in three dimensions. Image acquisition is carried out by sample scanning via nano-positioners with respect to a fixed laser focus and a local accumulation of corresponding Raman spectra.

The domain structures were fabricated in congruently melting lithium niobate by electric field assisted poling. A schematic representation of the sample geometry and a optical micrograph of an etched Ti:PPLN sample are depicted in Fig. 1(b). The indicated coordinate system corresponds to the crystallographic axes.

3. Results and Discussion

Raman measurements have been performed on periodically poled Z-cut LN structures at room temperature. The polarization of the excitation light was oriented perpendicularly to the crystallographic Z-axis. In this configuration the reference spectrum includes seven transversal-optical (TO) modes and one longitudinal-optical (LO) in good agreement with the assignment of group theory [5]. Based on these phonon modes we have analyzed the evaluation of the individual spatial intensity distribution with respect to the domain configuration. In Fig. 2 the intensity distribution of the E(TO9)-line resulting from a line-scan at the surface region of a Z-cut PPLN is depicted. Here the scan direction was across the periodic structure (X-direction).

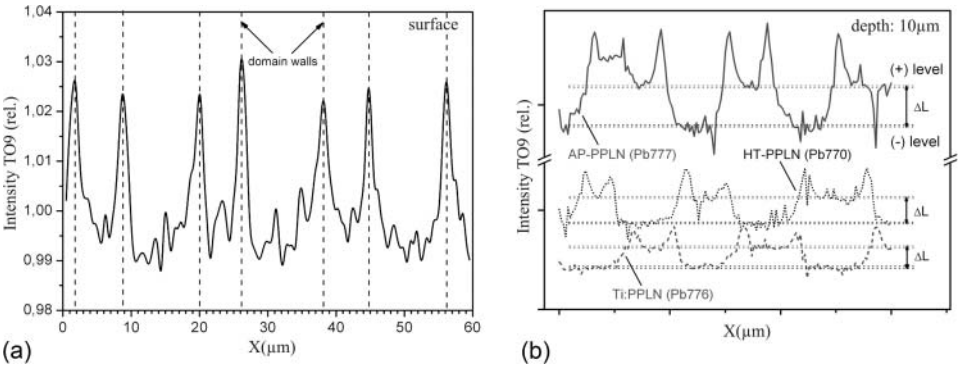

Figure 2. a) Intensity distribution along the X-axis based on the TO9-line a) originated from a PPLN surface and b) originated from a 40 μm depth confocal plane for different processed PPLN samples.

A specific modulation of the intensity distribution, which is linked to the periodic domain structures is visible. It results from a steep increase of the TO9 intensity at domain boundaries. We propose that the enhancement is in principle induced by the changes in the crystal morphology, which, in turn, influence the inner field distribution.

For the surface behavior of the corresponding phonon modes, we suggest that due to the superposition of electric fields in the center of the beam waist (Gaussian focus) and due to the restricted detection volume, almost only one direction of polarization interacts with matter. In contrast to the domain interaction the superposition failed at the domain boundary region, so other polarization directions result in admixed phonon modes, which generate an additional contribution to the detectable signal. As an example, the result of depth resolved measurements are given for the TO9 mode as lines-scans perpendicular to the periodic structure (X-line) in a depth of 40 μm for various PPLN structures in Fig. 2(b). Again the periodic modulation of the Raman signal is observable. Additionally we find specific intensity levels which spatial distribution relate to the contrarily poled domains. Here as poled samples (Pb777) exhibit a higher contrast between the signal of positive and negative domains. For annealed samples (Pb770, Pb776) the signal difference is minimal. Inside the

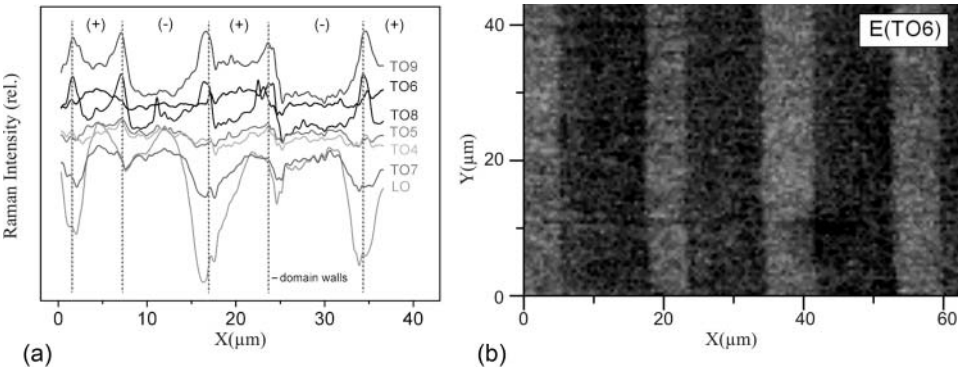

Figure 3. a) Intensity distribution of various Raman modes for a line-scan perpendicular to the domains (depth 40 μm). b) Confocal Image (Z-face) based on the spatial distribution of the TO6 phonon mode.

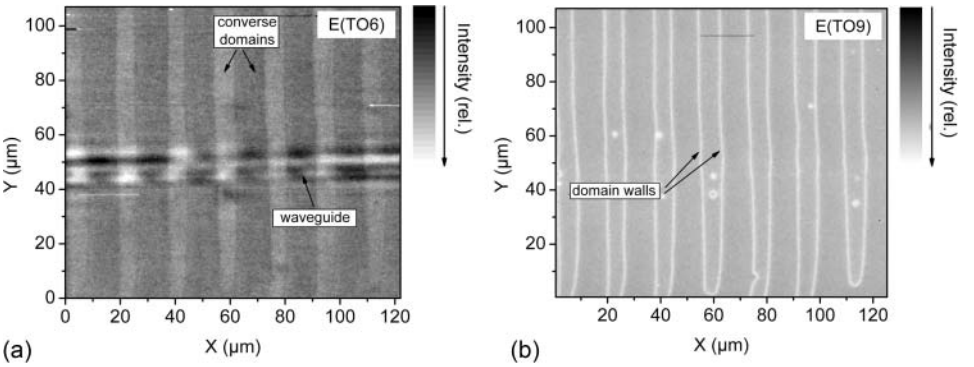

Figure 4. Confocal images of periodic domain structures taken from a) 3 μm and b) 10 μm depth XY-planes based on the spatial intensity variation of the a) E(TO6) and b) E(TO9) phonon mode.

bulk material the crystallographic structure becomes more dominant. So we conclude that structural changes strongly influence the inner electric field distribution and hence the vibrational states. The long ranging character of this phenomenon supports this idea. Because of the changed focus parameters (Rayleigh range inside the crystal) our method becomes more sensitive for other mode contributions. However, these stand to the reason that different phonon modes should be specific modulated. In Fig. 3(a) the spatial intensity distributions of various Raman lines for an X-line-scan are presented (depth 40 μm).

As mentioned before for all phonon modes a specific modulation could be identified regarding the periodic domain structure. Here the TO8 and TO9 modes show a very similar behavior (motions of oxygen octahedron), witch are sensitive to perturbation in the sublattice of both cations [6]. Because of the various modifications for different Raman bands both imaging of contrarily poled domains and the domain borders becomes possible. For imaging Raman spectroscopy a complete spectrum is recorded for every single spatial point thus information deriving from each phonon mode can be visualized by analyzing the spatial variation of the mode intensity. Exemplarily confocal images based on the E(TO6)-mode are shown in Fig. 3(b) (PPLN) and Fig. 4(a) (Ti:PPLN). In both cases the contrarily poled domains become visible due to different intensity levels for the E(TO6)-line regarding the polarity. Visualization of the intensity distribution for the peak intensities of the E(TO9)-line yields a two-dimensional image where the domain walls clearly visible (Fig. 4(b)). In addition appropriate intensity scaling enables also the mapping of the different levels for contrarily poled domains Overall, the applicability of the confocal Raman spectroscopy as an imaging method for monitoring of the transferred domain structures has been shown.

4. Conclusion

Confocal μ-Raman spectroscopy analyses have been performed to study the domain structure in PPLN. Due to the combination of spatial resolution and spectral information, phonon specific imaging becomes possible. We demonstrated the high potential of this method by direct imaging of ferroelectric domain structures, which selectively allows the imaging of domain polarities as well as domain boundaries. To clarify the underlying contrast mechanisms a theoretical modelling will be necessary.

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