Characterisation of periodically poled materials using nonlinear microscopy

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Abstract: Periodically poled crystalline materials are extremely attractive for processes such as second harmonic generation and optical parametric generation due to their very high conversion efficiency. For optimal performance, fabrication of poled regions with sub-micron tolerance is required. In this paper we introduce multi-photon laser scanning luminescence microscopy as a powerful minimally-invasive measurement technique which provides information about internal device structure with high spatial resolution that cannot be easily obtained with existing methods. A comparative study of confocal and multi-photon imaging of periodically poled crystalline materials is also performed.

OCIS codes: (180.4315) Nonlinear Microscopy; (100.6890) Three Dimensional Image Processing; (190.4180) Multiphoton processes.

References and links

1. L.E.Myers, W.R.Bosenberg, "Periodically Poled Lithium Niobate and Quasi-Phase-Matched Optical Parametric Oscillators", IEEE Journal of Quantum Electronics 33 (10): 1663-1672 (1997).
2. E.J.Lim, M.M.Fejer, R.L.Byer, "2nd-harmonic generation of green light in periodically poled planar lithium-niobate wave-guides", Electronics Letters 25 (3): 174-175 (1989).
3. L.E.Myers, G.D.Miller, R.C.Eckhardt, M.M.Fejer, R.L.Byer, "Quasi-phase-matched 1.064µm-pumped optical parametric oscillator in bulk periodically poled $LiNbO_3$", Optics Letters 20 (1): 52-54 (1995).
4. M.M.Fejer, G.A.Magel, D.H.Jundt, R.L.Byer, "Quasi-phase-matched 2nd-harmonic generation – tuning and tolerances", IEEE Journal of Quantum Electronics 28 (11): 2631-2654 (1992).
5. V.Dierolf, C.Sandmann, "Inspection of periodically poled waveguide devices by confocal luminescence microscopy", Applied Physics B 78: 363-366 (2004)
6. A.Rosenfeldt, M.Florsheimer, "Nondestructive remote imaging of ferroelectric c domain distributions with high three-dimensional resolution", Applied Physics B, 73, 523-529 (2001).
7. R.W.Boyd, "Nonlinear Optics-Second Edition", Academic Press, 107-111, (2003)
8. Y.Uesu, H.Yokota, S.Kawado, J.Kaneshiro, "Three-dimensional observations of periodically poled domains in a $LiTaO_3$ quasiphase matching crystal by second harmonic generation tomography", Applied Physics Letters 91, 182904 (2007)
9. D.L.Wokosin, V.Centonze, J.G.White, D.Armstrong, G.Robertson, A.I.Ferguson, "All-solid-state ultrafast lasers facilitate multiphoton excitation fluorescence imaging", IEEE Journal of Selected Topics in Quantum Electronics 2 (4): 1051-1065 (1996).
10. W.Denk, J.H.Strickler, W.W.Webb, "2-photon laser scanning fluorescence microscopy", Science 248 (4951): 73-76 (1990)
11. K.Moutzouris, F.Sotier, F.Adler, A.Leitenstorfer, "Highly efficient second, third and fourth harmonic generation from a two-branch femtosecond erbium fiber source", Optics Express 14 (5): 1905-1912 (2006)
12. K.Kawase, J.Shikata, H.Ito, "Narrow-linewidth tunable terahertz-wave sources using nonlinear optics", Topics in Applied Physics 89: 397-423 (2003).
13. L.E.Myers, R.C.Eckhardt, M.M.Fejer, R.L.Byer, W.R.Bosenberg, J.W.Pierce, "Quasi-phase-matched optical parametric oscillators in bulk periodically poled $LiNbO_3$", Journal of the Optical Society of America B, 12 (11): 2102-2116 (1995)
14. Y.Wang, V.Petrov, Y.J.Ding, Y.Zheng, "Ultrafast generation of blue light by efficient second-harmonic generation in periodically-poled bulk and waveguide potassium titanyl phosphate", Applied Physics Letters, 73 (7) 873-875 (1998)

15. Hyoun Woo Kim, Seung Hyun Shim and Jong Woo Lee, "Growth of MgO thin films with subsequent fabrication of ZnO rods: Structural and photoluminescence properties", Thin Solid Films, Volume 515, Issue 16, 4 June 2007, Pages 6433-6437
16. Xiangke He[a] and Dongfeng Xue, "Doping mechanism of optical-damage-resistant ions in lithium niobate crystals" Optics Communications, Volume 265, Issue 2, 15 September 2006, Pages 537-541.

1. Introduction

Periodically poled crystalline materials are extremely attractive due to the very high conversion efficiencies possible [1] and processes such as second harmonic generation (SHG) [2] and optical parametric generation [3] are much improved. In order to optimise these devices, the micro-domain structures made by the application of very high electric fields at the design stage have to be engineered with sub-micron tolerance [1]. The initial method employed to visually measure the success of QPM crystal manufacture was conventional brightfield microscopy [4] but this method gave very little information about internal structure, and will only provide information on the domain period length and damage from near the surface of the device. Defects or inhomogeneities at depth cannot be observed. Subsequent studies with confocal laser scanning microscopy (confocal LSM) [5] and second harmonic generation laser scanning microscopy (SHG LSM) [6] were later performed to provide structural information at depths of up to several tens of μm inside the crystal, thus further improving internal inspection capabilities. There are, however, limitations with these LSM techniques. Confocal LSM can be employed to generate luminescence from the doped regions, and the signal can be captured to create a stack of contrast images that can be used to study periodic structures across the length of the device. Unfortunately, this method is capable of visualizing only thin periodically poled crystal samples (i.e. <100μm) due to the single-photon nature of the excitation. Alternatively, SHG LSM is a complex imaging process which requires the correct orientation of the crystal relative to the incident radiation, as well as an optimized crystal temperature to induce efficient SHG via phase-matching [7]. In the absence of phase-matching, the SHG signal intensity is extremely low and therefore the detection method must be carefully chosen in order to increase the signal-to-noise ratio. For example Uesu et al [8] employed interference techniques to discriminate the low intensity SHG signal from noise to create optical contrast images of periodically poled domains in $LiTaO_3$, but this introduces additional cost and complexity to the nonlinear imaging system.

Multi-photon laser scanning luminescence microscopy (multi-photon LSM) offers a novel solution approach for imaging thick periodically poled nonlinear materials. There are no strict conditions on the crystal orientation or temperature required to generate a multi-photon-excited signal. There are also no strict requirements upon the excitation/detection wavelength, as luminescence excitation is broadband in comparison with SHG LSM, which requires excitation at a specific wavelength and limits the collection wavelength range to exactly half the excitation wavelength. By comparison, multi-photon excitation of periodically poled media can be performed using conventional multi-photon imaging technology and laser sources, and detection over >10nm spectral bandwidths yields a high signal intensity to create very high-resolution, high contrast images. Furthermore, multi-photon excitation allows improved depth imaging over single-photon excitation, enabling high contrast imaging of periodically poled media at depths of several hundred μm [9,10]. By using multi-photon luminescence LSM to generate high-resolution 3D reconstructions, we present a novel method of domain characterisation and damage analysis for periodically poled materials. We report the application of this method for magnesium-oxide doped periodically poled lithium niobate (MgO:ppLN) characterisation but this flexible approach can also be employed to study other periodically poled materials.

2. Background

Quasi-phase-matching (QPM) provides an alternative to conventional birefringent phase-matching in bulk nonlinear media, which places a severe requirement of a zero phase mismatch ($\Delta k=0$). In QPM, $\Delta k\neq 0$ is permitted but after one coherence length the mismatch is reset to zero. This is performed by reversing the sign of the nonlinear coefficient, which has the effect of adding π to the relative phase shift of the interacting waves. This is illustrated in Figure 1. Quasi-phase-matching of nonlinear materials to create periodically poled structures has proven extremely effective in nonlinear optical frequency conversion experiments, permitting the use of the highest *d* component via phase-matching in any direction and enabling phase-matching in isotropic media or that which has inadequate birefringence for conventional phase-matching. The overall conversion efficiency can be maximised by definition of the domain period Λ, which is twice the coherence length. The length of the domain period Λ is determined at the point of manufacture

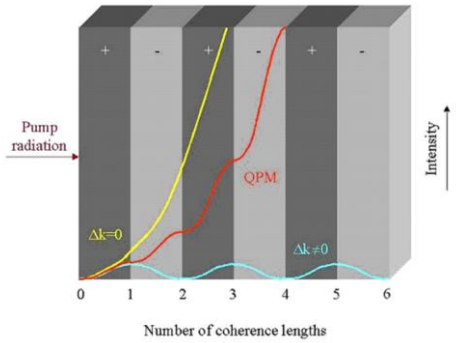

Fig.1. The effect of reversal of the nonlinear coefficient in a periodically poled structure

In the case of a ferroelectric material, domain period length Λ is determined by the application of an electric field. For example, magnesium oxide-doped lithium niobate ($MgO:LiNbO_3$) becomes magnesium oxide-doped periodically poled lithium niobate (MgO:ppLN). MgO:ppLN has a very high damage threshold and >50mm crystals are commercially available. Due to the wide spectral transmission, MgO:ppLN has found applications in the generation of short, UV wavelengths through to the terahertz region of the EM spectrum [11,12]. Under the application of a critically high electric field for >50ms, the lithium and niobium ions in $MgO:LiNbO_3$ will shift relative to the oxygen layers thus changing the polarity of the crystal, thus creating the domain periods [13]. Typically Λ>10µm although shorter period structures do exist [14]. The length of the domain period Λ must be controlled with sub-micron wavelength accuracy to maximise the efficiency of the required conversion process. Analysis of the QPM crystal structure is therefore critical to assess the quality of the resultant poled crystal to ensure that the domain period is within the sub-micron tolerance required. Furthermore, it is necessary to verify that the crystal is free from damage or defects both on the surface and deeper within the device. This greatly limits the quality assessment of QPM devices.

3. Experiment

Figure 2 shows the experimental setup in both the confocal LSM and multi-photon LSM cases, where an upright microscope was attached to a laser scanning system and used a 20x/0.5NA objective lens. For the confocal LSM luminescence study, we employed a confocal laser scanning system (Leica SP5) coupled to an upright microscope (Leica DM6000). We used 1mW average power output from a λ = 488nm, emitting Krypton/Argon laser source to generate single-photon luminescence from the crystal.

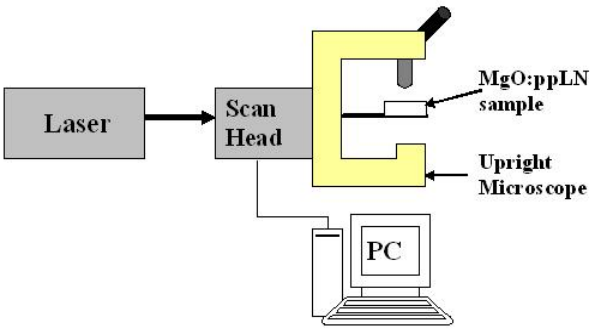

Fig.2. Experimental setup for both the confocal LSM and multi-photon LSM methods. An upright microscope was attached to a laser scanning system and a 20x/0.5 N.A. objective lens was used.

This source was chosen peak single-photon absorption wavelength to generate luminescence from MgO at room temperature is 465 nm [15] and the λ=488nm source was the closest available match for this excitation. The spectral detection feature of the Leica SP-5 detection system enabled the choice of the optimum luminescence spectral detection range while blocking the λ=488nm excitation source. For this crystal, the λ= 500-600nm spectral range offered the best signal detection. For the multi-photon LSM imaging we employed a laser scanning system (Bio-Rad Radiance MP2000) coupled to an upright microscope (Nikon E600FN). For two-photon excitation of luminescence, we used an average laser power of 6mW from a heavily-attenuated 110fs-pulsed Ti:Sapphire laser system (Chameleon, Coherent) with a pulse repetition rate of 90MHz and a wavelength of λ=860nm. Unlike the Leica SP-5 confocal system, the Radiance MP2000 utilises direct detectors that circumvent the need for the generated luminescence signal to travel back through the scanhead, thus limiting unnecessary optical loss of the luminescence signal intensity. A Ti:Sapphire blocking filter (700SP, Chroma Technologies) was implemented to ensure that only the luminescence signal intensity from the crystal was detected and used to create the contrast image. Optical sectioned images of 512^2 pixels were captured at a rate of approximately 1Hz using a 20x/0.50 N.A. objective lens on both systems throughout the entire 250μm depth of the crystal. Image analysis software (Volocity 4, Improvision) was then used to build full three-dimensional reconstructions of the crystal, using either the single- or multi-photon luminescence signal arising from excitation of the MgO dopant within the crystal. Measurements of the domain period length were made using the same software. By scanning through the optical sections we could investigate the crystal for regions of photo-induced damage and crystal inhomogeneities at depth. Using a 20x/0.5NA objective lens, the axial and lateral resolution for both systems at 488nm were calculated to be 1.64 um and 0.39 um respectively.

4. Results

The imaging parameters on both systems were adjusted to give the best contrast image. With single-photon luminescence excitation, the laser power was set to its maximum value and the PMT gain settings were set to achieve to greatest signal-to-noise ratio. For multi-photon luminescence excitation, only 20% of the available laser power was required along with a low PMT gain setting. Figure 3 shows a typical three-dimensional reconstruction created from images obtained using a) confocal LSM and b) multi-photon LSM On inspection, the structural detail using multi-photon LSM is far greater than that of confocal LSM..

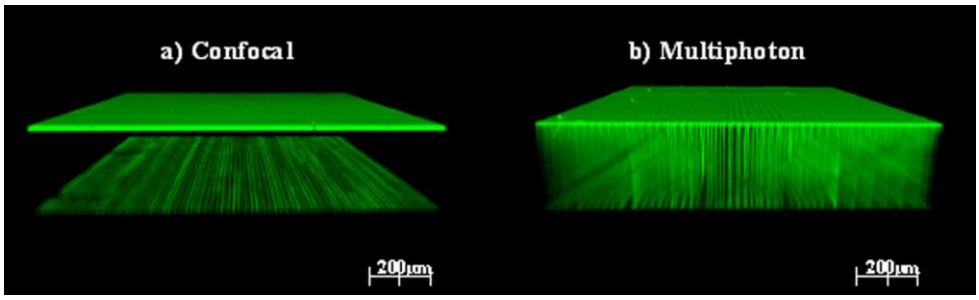

Fig.3. Comparison of 3D reconstructions obtained using a) confocal and b) multi-photon laser scanning luminescence microscopy.

This is attributed to (1) less absorption of the longer wavelength excitation source in the crystal above the plane of focus [10] (2) the signal being quantified by direct detectors on the multi-photon LSM system, which enabled greater signal detection performance (3) highly localized excitation circumventing the need for an iris to reject out-of-focus signal which results in a modicum of luminescence signal loss [10] and (4) less scattering of the longer wavelength excitation light [10].

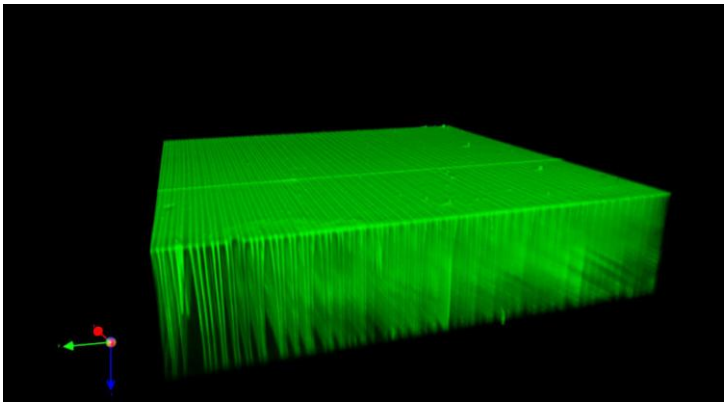

Fig.4. A movie of the 3D reconstruction of the luminescent MgO regions within the MgO:ppLN crystal obtained using multi-photon LSM and Volocity 4 software (2.32MB).

A movie of the 3D reconstruction of the MgO:ppLN crystal using multi-photon LSM and Volocity 4 software is shown in figure 4. This is a powerful analysis method and can be used to thoroughly inspect the crystal visually from different angles, internally and externally.

Figure 5.a. displays a comparison between the lateral view (XY) and the axial view (XZ) from the 3D reconstruction of the MgO:ppLN imaged using multi-photon LSM. The XY section was imaged 20µm below the top surface of the crystal. For discussion purposes, three columns have been highlighted which illustrate different features observed during the study. Column 1 shows a near perfect match between XY and XZ which is indicative of successful fabrication, whereas the XY section of Column 2 highlights non-periodicity on the surface of the crystal structure. Additionally, the image of XZ in Column 2 shows a large change in Λ throughout the depth of the crystal. Similarly, the XY image in Colum 3 shows good periodic structures but the cross sectional image XZ draws attention to internal deformation of the crystal. Regions of damage are clearly visible in the XY luminescence

images of Columns 2 and 3, as well as in other regions of the crystal,. Furthermore, it is clear from the variation in luminescence signal intensity at different regions of the crystal that the MgO dopant is inhomogeneous across the crystal, with higher dopant concentrations at the period boundaries in all cases. This is a consequence of the fabrication process, where the dopant migrates relative to the direction and strength of the electric field [16].

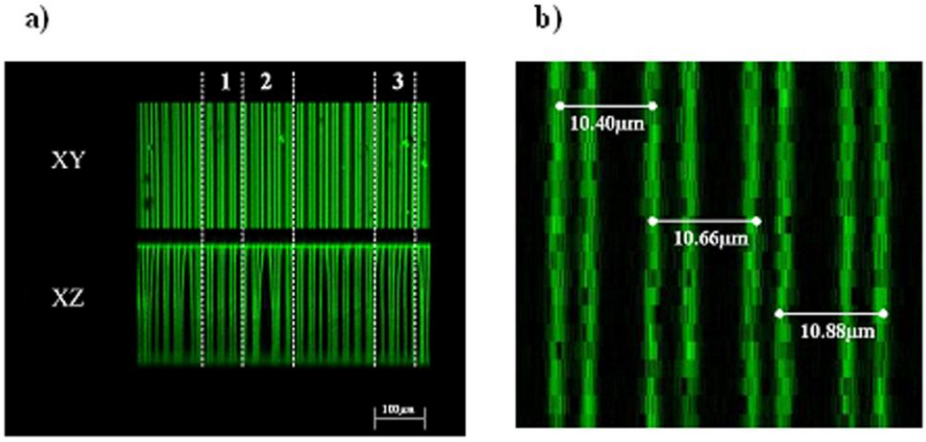

Fig.5.a) Comparison of the MgO:ppLN crystal surface view (XY) with the cross sectional view (XZ). Images were taken from the 3D reconstruction of the crystal obtained using multi-photon LSM excited luminescence. b) Close-up image taken from Column 1 in fig 5.a.. Measurements of the crystal periodicity were easily obtained using Volocity 4 image analysis software.

As well as obtaining visual information about the structure of the crystal, image analysis enables accurate measurement of the MgO:ppLN crystal period length, which contributes towards the performance of the structure for nonlinear frequency conversion. Figure 5.b. features a close-up image from Column 1 for close inspection of the luminescence excited within the crystal Three measurements of the location of this high intensity luminescence signal were taken to illustrate the difference in periodicity within the crystal, from which the difference in periodicity was measured to be as large as 0.48µm. The lack of periodicity and the damage observed in Figure 5.a. and 5.b. reduces the efficicacy of the QPM device for nonlinear frequency conversion. This study highlights the importance of imaging the internal structure of QPM crystals such as MgO:ppLN, since inspection of the near surface alone cannot give an accurate indication whether the fabrication process was successful.

5. Conclusion

From this study, it has been shown that multi-photon LSM provides important information that cannot be obtained via brightfield microscopy, confocal LSM or SHG imaging techniques. Using multi-photon luminescence LSM, internal deformation of the crystal structure and dopant inhomogeneity can be identified at depth within thick samples, with sub-micron resolution. Such imaging could potentially be used as an inspection tool to aid in the fabrication and subsequent enhancement of the nonlinear characteristics of both MgO:ppLN and alternative QPM devices.

6. Acknowledgements

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