NON-COLINEAR FREQUENCY DOUBLING IN LITHIUM NIOBATE

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Abstract   Induced non-colinear frequency doubling (INCFD) is presented as a characterization technique for electrooptic crystals. Using the interaction of two non-colinear laser beams, the method yields a three-dimensional spatial resolution. First experimental results on several samples of lithium niobate are presented. The limitations of the technique are discussed.

INTRODUCTION

In the last decades various ferroelectric materials have been shown to be suitable for electrooptic applications. One of the most important is still lithium niobate which is widely used in electooptical and electroacoustic device fabrication.

It is possible to influence the properties of these crystals - especially in the case of lithium niobate - by varying the composition or by adding different dopants. Many methods for checking the crystal composition have been developed, for example: measuring of the Curie temperatur [1], optical absorption edge [2], refractive indices [10], birefringence [3], second harmonic generation. The last three methods use the fact that the refractive indices depend on the crystal composition and - in the case of second harmonic generation - vary with temperature.

Conventionally applied, second harmonic generation is a colinear process which means that phase matching can be achieved when the quasi-scalar relation for momentum conservation $|\vec{k_2}| = |\vec{k_1}| + |\vec{k_1'}|$ (1) is fulfilled, where $|\vec{k_1}|$ and $|\vec{k_1'}|$ are the wave vectors for the fundamental beams and $|\vec{k_2}|$ for the generated harmonic wave; usually noncritical phase matching is applied. Using spatially resolved second harmonic generation (SRSHG) [4, 5] two dimensional topographies of the crystal composition with good spatial resolution can be achieved.

In contrast to this, non-colinear frequency doubling utilizes two light beams inclined to each other to produce second harmonic light. The *vectorial* phase matching condition $\vec{k_2} = \vec{k_1} + \vec{k_1'}$ (2) has to be applied which yields an additional experimental parameter, the angle between $\vec{k_1}$ and $\vec{k_1'}$. Spontaneous non-colinear frequency doubling [6, 7, 8] uses scattered light as a second beam. A cone of second harmonic light results which reflects the properties of the illuminated region in the crystal.
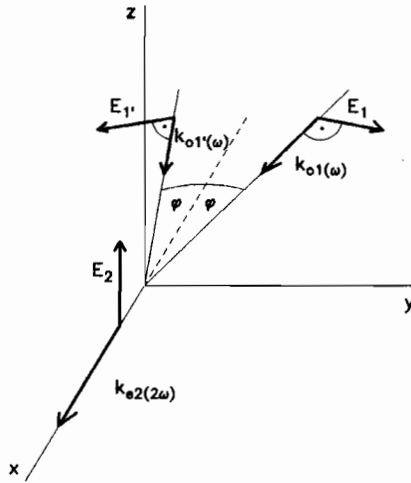
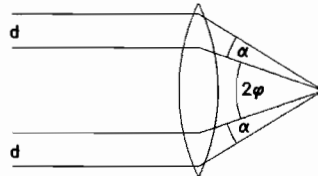FIGURE 1: Wave vectors and polarizations for non-colinear phase matching in LiNbO$_3$ crystals.

FIGURE 2: Geometry of the focussed beams

In this paper we will concentrate on induced non-colinear frequency doubling (INCFD) where two laser beams are crossed inside the crystal. As the interaction volume of the two laser beams is limited in all three spatial dimensions, a three-dimensional non-destructive investigation of crystals is possible.

EXPERIMENTAL

Taking into account the shape of the indicatrix, equation (2) can be transferred to an equivalent condition for the refractive indices. For negative uniaxial crystals like LiNbO$_3$ and a geometry where the fundamental beam polarizations are perpendicular, the harmonic beam polarization parallel to the optic axis, the equation $n_e(2\omega) = n_o(\omega) \cdot \cos\varphi'$ (3) results. n_o and n_e are the ordinary and extraordinary indices of refraction, respectively, and φ' is the angle enclosed by one fundamental and the second harmonic beam inside the crystal. The two fundamental beams are assumed to have the same frequency ω. Eq. (3) can
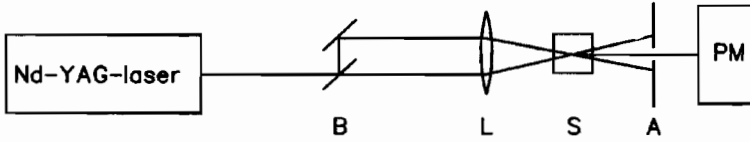
FIGURE 3: Experimental arrangement for measuring INCFD. B: beam splitter, L: lens, S: temperature controlled sample holder, moveable in 3 directions, A: aperture for blocking the fundamental beams.
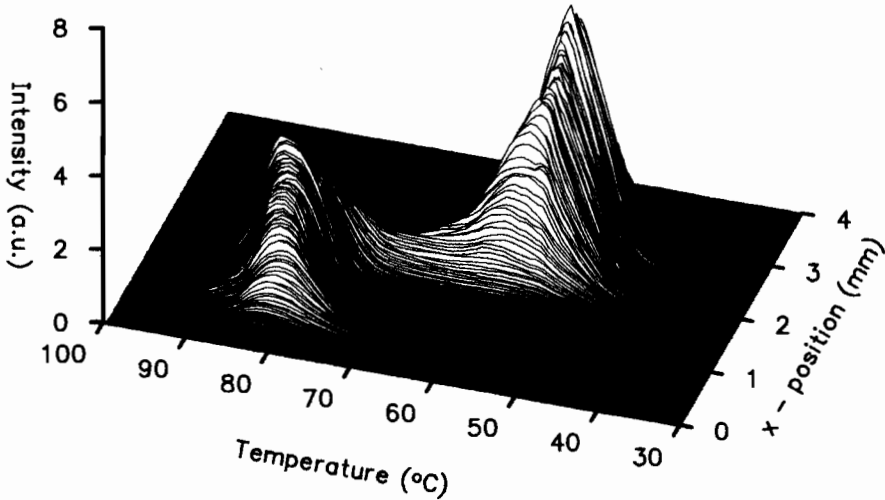

FIGURE 4: SHG-intensity as a function of temperature for different crystal positions. The fundamental and harmonic beams propagate in x-direction.

be satisfied either by chosing an approxiate angle φ' or by variing the temperature thus changing the refractive indices. Angle or temperature can be used as a measure for the crystal composition. The conditions described in figure 1 are achieved in an experimental setup which is schematically sketched in figure 3.

The principal limitations for the resolution of the technique may be derived from figure 2 and equation (3). The ratio $n_e(2\omega)/n_o(\omega)$ limits the useable angle 2φ to a maximum of about 10 degrees. High spatial resolution - achieved by short focal length focussing - has the drawback of a large beam divergence and thus bad temperature resolution. In general one has to find a compromise suited to the crystal under investigation.

RESULTS

The principle of the measuring technique was tested on a model sample consisting of two homogeneous $LiNbO_3$ crystals with different compositions which were positioned one
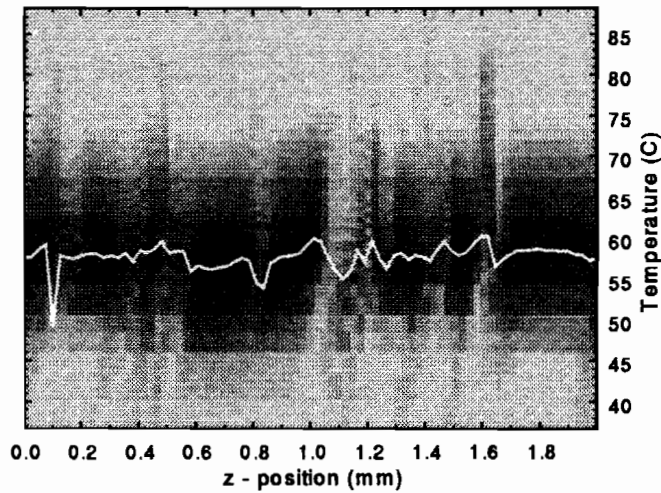
FIGURE 5: SHG-intensity as a function of the crystal position in z-direction and of the crystal temperature measured on a Mg-doped LiNbO$_3$ sample. The white line connects the intensity peaks for the respective positions which reflect the phase matching temperatures. A variation of 1°C in the phase matching temperature corresponds to 0.01 mol% in the Li content.

behind the other. Figure 4 shows the SHG-intensity as a function of the sample position in beam direction (x-direction as shown on figure 1) and of the sample temperature. The two homogeneous regions can be clearly distinguished with good spatial resolution. The intensity peaks reflect the NCFD phase matching temperatures which correspond to the respective phase matching temperatures for colinear SHG.

The method was then applied to investigate a LiNbO$_3$ crystal doped with about 4% Mg [9] which showed up slight variations in its optical quality. In the SHG intensity measured as a function of temperature similar variations are found, their dependence on crystal position in growth direction is depicted in figure 5. The variation of the phase matching temperature in a cross sectional plane through the crystal is shown in Figure 6. This temperature varies by about 12 K which can be referred to a variation in Li content of about 0.12% [10]. In contrast to earlier measurements [4,5] variations not only in growth direction (z) but, albeit less expressed, also perpendicular to it can be stated.

As an example for a composition profile we investigated a LiNbO$_3$ crystal treated by vapour transport equilibration (VTE) technique [11] in Li-rich powder for several hours. The SHG intensity as a function of temperature and crystal position is shown in figure 7. The bulk NCFD phase matching temperature (-60°C) corresponds to congruent LiNbO$_3$, the starting material. At the borders the stoichiometric composition ($\hat{=}190^\circ C$) is approximately reached, the in-diffusion of Li is well observable.
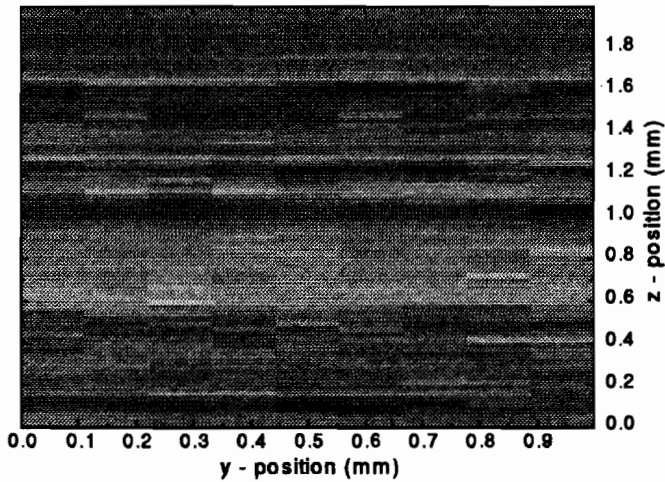
FIGURE 6: Phase matching temperature as a function of the crystal position in y- and z-direction (Mg-doped LiNbO$_3$ sample). The phase matching temperature varies between 48°C (black) and 60°C (white) which corresponds to a variation in Li content of approximately 0.12 mol%.
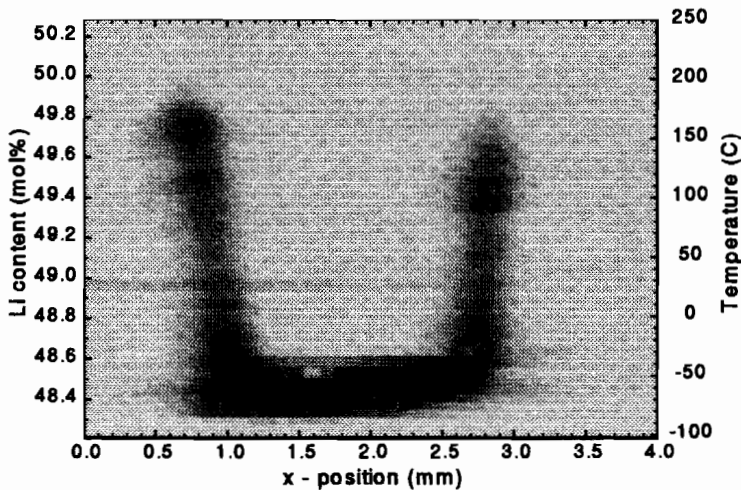

FIGURE 7: SHG-intensity as a function of the crystal position in x-direction, measured on a crystal treated by vapour transport equilibration (VTE) technique. The left axis denotes the Li content which corresponds to the respective NCFD phase matching temperature [10] (linearized scale).

CONCLUSION

Induced non-colinear frequency doubling has been shown to be a powerful technique for the topographic characterization of electrooptic crystals. A three-dimensional nondestructive investigation of compositional or other variations is possible with exellent accuracy and good spatial resolution. For the case of $LiNbO_3$ the accuracy corresponds to approximately 0.01 mol % Li content.

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REFERENCES

1. J.R.Carruthers, G.E.Peterson, M.Grasso, and P.M.Bridenbaugh. J. Appl. Phys. **42**, 1846 (1971).

2. I.Földváry, K.Polgár, R.Voszka, and R.N.Balasanyan. Crystal Res. & Technol. **19**, 1659 (1984).

3. E.Born, E.Willibald, and R.Veith. Proc. IEEE Ultrasonics Symp. (1984).

4. N.Schmidt, K.Betzler, and S.Kapphan. Cryst. Latt. Def. and Amorph. Mat. **15**, 103 (1987).

5. N.Schmidt, K.Betzler, M.Grabs, and F.Klose. J. Appl. Physics **65**, 1523 (1989).

6. J.A.Giordmaine. Physical Review Letters **8**(1), 19 (1962).

7. Harry E. Bates. Journal of the Optical Society of America **61**(7), 904 (1971).

8. A.Reichert and K.Betzler. Ferroelectrics **126**, 9 (1992).

9. B.C.Grabmaier and F.Otto. SPIE (Innsbruck) and ICCG-8 (YORK) (1986).

10. U.Schlarb and K.Betzler. Ferroelectrics (1993).

11. D.H.Jundt and M.M.Fejer. J. Appl. Phys. **72** (1992).