

APPLICATION OF NONCOLINEAR FREQUENCY DOUBLING FOR CRYSTAL CHARACTERIZATION

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Abstract Noncolinear frequency doubling (NCFD) is discussed as a sensitive technique for the characterization of acentric crystals. Two methods can be applied: ‘Spontaneous’ NCFD where light scattered at crystal imperfections is used as second fundamental beam and ‘induced’ NCFD where the usage of a second laser beam yields three-dimensional spatial resolution. Experimental investigations of pure and VTE-treated lithium niobate are presented.

INTRODUCTION

For the application of electrooptic crystals a reliable reproducibility of their relevant physical properties is of great importance. Generally these properties can be tuned in a certain range by varying the composition or by adding appropriate dopants. To control fabrication processes, the crystal properties have to be checked by appropriate techniques [1–7]. One of the most sensitive methods is the optical second harmonic generation (SHG) which – by measuring phase matching conditions – can detect crystal composition and its spatial variation [6, 7]. Here we describe two recently developed new SHG methods which utilize the phase matching conditions for second harmonic light generated by two *noncolinear* fundamental beams for crystal characterization.

FUNDAMENTALS OF NONCOLINEAR FREQUENCY DOUBLING

Solving the wave equation for the case of optical second harmonic generation results in

$$S_2(L) \propto L^2 \frac{\sin^2(\Delta k L/2)}{(\Delta k L/2)^2} S_1(0) S_1'(0) \quad (1)$$

for the intensities S_i of the involved beams. L is the length of the interaction volume, the phase mismatch Δk is defined by

$$\Delta k = |\vec{k}_2 - \vec{k}_1 - \vec{k}_1'| = \frac{4\pi}{\lambda_1} \left(n_2(2\omega) - \frac{n_1(\omega) \cos \alpha + n_1'(\omega) \cos \alpha'}{2} \right) \quad (2)$$

with the refractive indices n_i and the angles α and α' enclosed by the fundamental wavevectors (\vec{k}_1, \vec{k}_1') and the harmonic one (\vec{k}_2) , respectively.

The phase matching condition $\Delta k = 0$, which can be detected with excellent accuracy, is generally used to characterize the material.

In the case of conventional – colinear – second harmonic generation, where all wavevectors are mutually parallel to each other, Eq. 2 is simplified using $\alpha = \alpha' = 0$; phase matching is usually achieved by temperature tuning; the phase matching temperature for noncritical phase matching is used as the characterizing parameter [8, 9].

In contrast to this, noncolinear frequency doubling (NCFD) utilized two light beams inclined to each other to fulfill the phase matching condition. Both, temperature and/or angle, can be used in a supplementary way to characterize the material. The second necessary fundamental beam can be taken from scattered light in the crystal (spontaneous NCFD, SNCFD)

or explicitly as second laser beam (induced NCFD, INCFD). In the latter case a fully three-dimensional topographical resolution is possible due to the interaction volume being limited in all three spatial dimensions.

SPONTANEOUS NONCOLINEAR FREQUENCY DOUBLING

When an intense laser beam hits the crystal surface perpendicularly, most of the light passes inside unrefracted (\vec{k}_1). A certain amount of light, however, is scattered by crystal imperfections. If the angle $\alpha + \alpha'$ between fundamental and scattered light (\vec{k}'_1) fulfills the noncolinear phase matching condition $\Delta k = 0$ (Eq. 2), frequency doubled light (\vec{k}_2) is amplified (Eq. 1) [10, 11, 3]. The set of phase matching conditions between fundamental and scattered light beams leads in general to an elliptic cone of second harmonic light.

In LiNbO₃, using an ordinarily polarized fundamental beam with its wavevector perpendicular to the c-axis of the crystal, the cone angle (outside the crystal, \perp c-axis) – the so called SNCFD angle – is given by

$$\varphi = \arcsin \left(n_e(2\omega) \sqrt{1 - \frac{n_e^2(2\omega)}{n_o^2(\omega)}} \right), \quad (3)$$

with n_o : ordinary, n_e : extraordinary refractive index.

Angular Fine Structure The intensity of the second harmonic light depends on Δk like $(\sin x/x)^2$ (Eq. 1). In the case of temperature phase matching this dependence can be measured as a function of temperature due to the temperature dependence of the refractive indices. When spontaneous NCFD is applied, Δk is varying as a function of the the angle φ causing an *angular* fine structure. Fig. 1 shows for the first time a measurement of this angular fine structure of the SNCFD light.

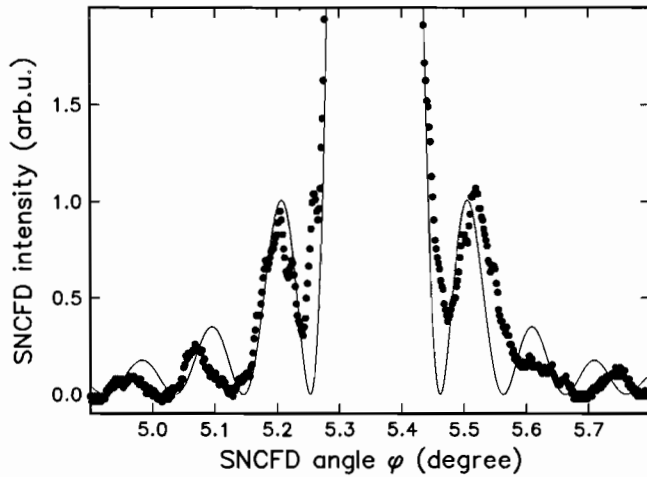


Figure 1: Angular fine structure of the SNCFD ring. The dots mark the measurement while the line is marking the calculated dependence. The intensity of the side maxima is scaled to one.

Crystal Characterization by SNCFD After focussing the beam onto the crystal the emerging cone of frequency-doubled light is projected onto a screen which is scanned by a CCD camera. By moving the sample with two stepper motors a two-dimensional spatial resolution is realized. As a two-dimensional spatially resolved scan of the crystal results in up to some thousand ring pictures, it is necessary to use an automatic evaluation algorithm for the ring parameters [12].

To show the high resolution provided by SNCFD, a highly homogeneous sample crystal was investigated, LiNbO₃ grown from a melt containing a certain amount of potassium [13]. As

can be derived from Fig. 2, composition changes less than 0.01 % Li_2O can be resolved by the method.

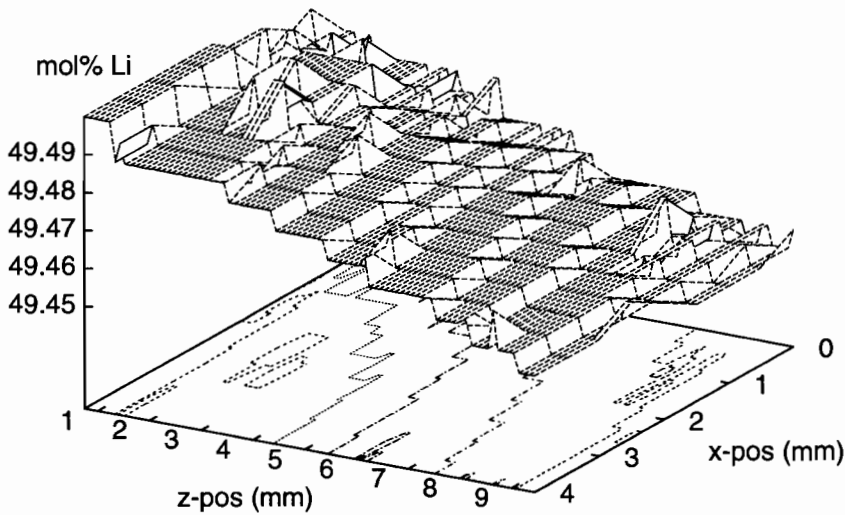


Figure 2: SNCFD topography of a highly homogeneous LiNbO_3 crystal grown from a potassium-enriched melt. The slight variation of the crystal composition in growth direction (z) can be detected with an accuracy of better than 0.01 %.

INDUCED NONCOLINEAR FREQUENCY DOUBLING

In contrast to SNCFD, for INCFD the two fundamental beams are taken in a symmetric way from the laser light, forcing a restricted interaction volume.

The experimental arrangement for measuring INCFD is sketched schematically in Fig. 3. The light of a pulsed Nd:YAG-laser is split into two parallel beams of approximately equal intensity. The two beams are then focussed onto the same spot inside the sample crystal. The

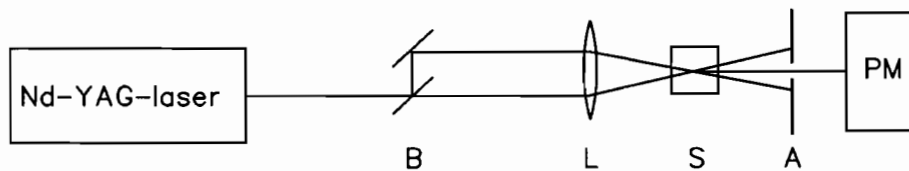


Figure 3: Experimental arrangement for measuring INCFD. B: beam splitter, L: focussing lens, S: temperature controlled sample holder, moveable in all three spatial directions, A: aperture for blocking the fundamental beams, PM: photomultiplier.

sample position can be controlled by a three-dimensional linear translation stage with μm resolution. The second harmonic intensity is measured by a photomultiplier with suitable pulse detection electronics as a function of the four parameters position $\{x, y, z\}$ and sample temperature. An evaluation with respect to the temperature yields the three-dimensional topography of the (noncolinear) phase-matching temperature which can be referred to a topography of the composition or doping of the material [14].

The INCFD-measured topography of a LiNbO_3 crystal treated by vapor transport equilibration (VTE) techniques [15, 16] is shown in Fig. 4 for two spatial directions (x and z). As a measure for the Li-content the (noncolinear) phase-matching temperature is plotted as a function of the spatial coordinates x and z in the central xz -plane of the crystal. The crystal was treated by VTE for about 200 hours at a temperature of 1100°C , indiffusion of Li was permitted from each of the six crystal faces.

CONCLUSION

Noncolinear frequency doubling offers, with the two techniques SNCFD and INCFD, very sensitive methods for the characterization of acentric – especially electrooptic – crystals. Both

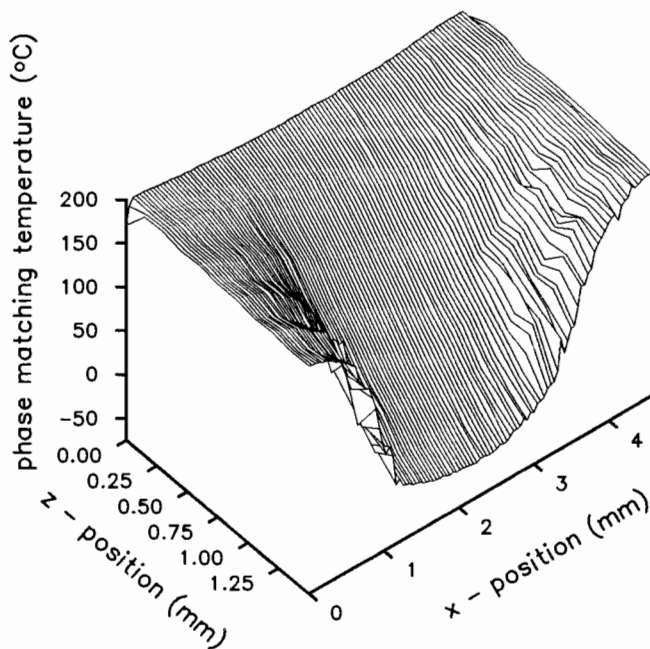


Figure 4: Topography of the INCFD phase matching temperature in the central xz -plane of a VTE treated LiNbO_3 crystal (for a better overview only about half of this topography from $z=0$ – the edge of the crystal – upto $z=1.5$ mm is drawn). The phase matching temperature is a measure for the Li content in the crystal.

techniques enable an accurate two- or three-dimensional topographical inspection of the crystal composition; for the example of lithium niobate the resolution is much better than 0.01 mol % Li_2O . Furthermore, it is possible to locate imperfections in crystals by INCFD, as e. g. domain boundaries.

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