

## CHARACTERIZATION OF ELECTROOPTIC CRYSTALS BY NON-COLLINEAR FREQUENCY DOUBLING

A. REICHERT AND K. BETZLER  
Fachbereich Physik, Universität Osnabrück, FRG

**Abstract** Non-collinear frequency doubling (NCFD) is presented as a technique for characterizing electrooptic crystals. Two methods can be applied: 'Spontaneous' NCFD uses Rayleigh scattered light as second fundamental beam thus yielding a rather simple experimental arrangement; 'induced' NCFD needs a precise second laser beam yet allows 3-dimensional spatial resolution. The principles of the methods and some experimental results for  $\text{LiNbO}_3$  are discussed.

### INTRODUCTION

Electrooptic materials usually exhibit a rather complex crystal structure. Due to this fact most of these materials can be grown in a comparably wide composition range, a large variety of dopants may be added. A typical member in this group of materials is  $\text{LiNbO}_3$  <sup>1, 2</sup>. Both dopants and composition influence the crystal properties in a more or less expressed way. To check the crystal composition, several physical methods have been developed. All of them use one of the crystal properties which are strongly composition dependent. Easily measurable properties include the Curie temperature <sup>3</sup>, the birefringence <sup>4, 5</sup> and the optical absorption edge <sup>6</sup>. A newly presented method makes use of anisotropic holographic scattering <sup>7</sup>. The method of measuring the phase-matching temperature for optical second harmonic generation (SHG) depends on the fact that the extraordinary index of refraction strongly varies with both composition <sup>8, 9, 10</sup> and temperature <sup>11</sup>, whereas the ordinary stays rather constant. Using spatially resolved second harmonic generation (SRSHG) <sup>12, 13</sup>, two-dimensional topographies with a considerable spatial resolution can be achieved. Here we describe non-collinear frequency doubling (NCFD) as an additional technique for characterizing electrooptic crystals. Spontaneous NCFD is a simply-to-apply room-temperature method with two-dimensional resolution, induced NCFD for the first time yields the possibility of nondestructive three-dimensional investigations.

## PRINCIPLES OF THE METHOD

Both SHG methods — SRSHG and NCFD — use the fact that the refractive indices and thus the phase-matching conditions for second harmonic generation are affected by crystal composition and dopants. Inhomogeneities in these crystal parameters show up as spatial variations in the phase-matching conditions. For SRSHG ('collinear' frequency doubling) the phase-matching condition is given by  $|\vec{k}_1| + |\vec{k}'_1| = |\vec{k}_2|$  (1) 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, respectively. Usually noncritical phase matching is applied. The tuning parameter used in SRSHG is the sample temperature. This results in a very high sensitivity for the detection of inhomogeneities but has several obvious drawbacks, especially near phase transitions.

In contrast to that, NCFD uses two non-collinear fundamental beams which have to obey the — vectorial — phase-matching condition  $\vec{k}_1 + \vec{k}'_1 = \vec{k}_2$  (2). So the angle enclosed by the two fundamental beams can be used as an additional probing parameter thus drastically reducing the necessary temperature range. Taking into account the shape of the indicatrix, eq. 2 can be transferred to an equivalent condition for the refractive indices. For negative uniaxial crystals like  $\text{LiNbO}_3$  and a geometry where the fundamental beam polarizations are perpendicular, the harmonic beam polarization parallel to the optical axis, the equation  $n_e(2\omega) = n_o(\omega) \cdot \cos \varphi'$  (3) results where  $n_o$  and  $n_e$  are the ordinary and extraordinary indices of refraction, respectively, and  $\varphi'$  is the angle enclosed by one of the fundamental and the second harmonic beam inside the crystal. Furthermore the two fundamental beams are assumed to have the same frequency  $\omega$ .

## SPONTANEOUS NCFD

A simple method to generate non-collinear frequency doubled light is to shine an intense laser beam onto the crystal to be investigated<sup>14, 15</sup>. Scattered light from the surface or from crystal imperfections supplies the necessary second beam, a cone of second harmonic light results (fig. 1). That scattering is the reason for this effect, can be easily verified by

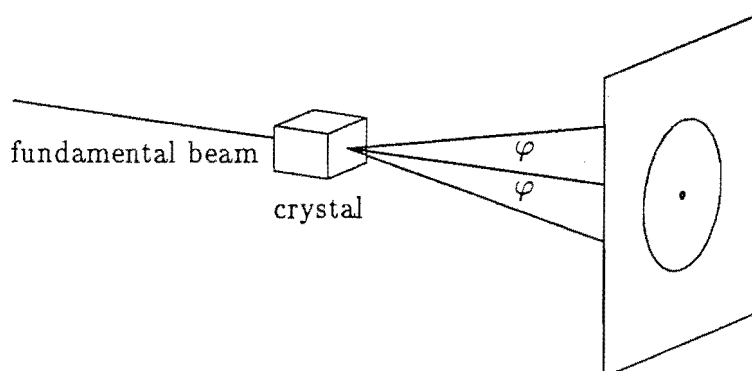


FIGURE 1: SHG-ring generated by spontaneous NCFD. For arbitrary directions an elliptic intensity pattern results. The shape of the ellipse is caused by the refractive indices used, the intensity variations reflect the nonlinear susceptibility tensor.

ing the intensity of the generated scattered light (e. g. by surface treatment).

To test the described spontaneous NCFD for its applicability to characterize crystals applied the method on various  $\text{LiNbO}_3$  samples. As fundamental beam the light of Q-switched Nd:YAG-laser was used (1064 nm, 1 kHz,  $\approx 10$  kW). The generated second harmonic light was detected — after appropriate focussing — by a linear array detector arranged radius-like to the SHG ring. Detector, crystal and fundamental beam were set up in a way that the wave vectors  $\vec{k}_1$ ,  $\vec{k}'_1$  and  $\vec{k}_2$  were directed perpendicularly to the optical axis of the crystal. Fundamental beam polarization was perpendicular, second harmonic beam polarization parallel to the optical axis.

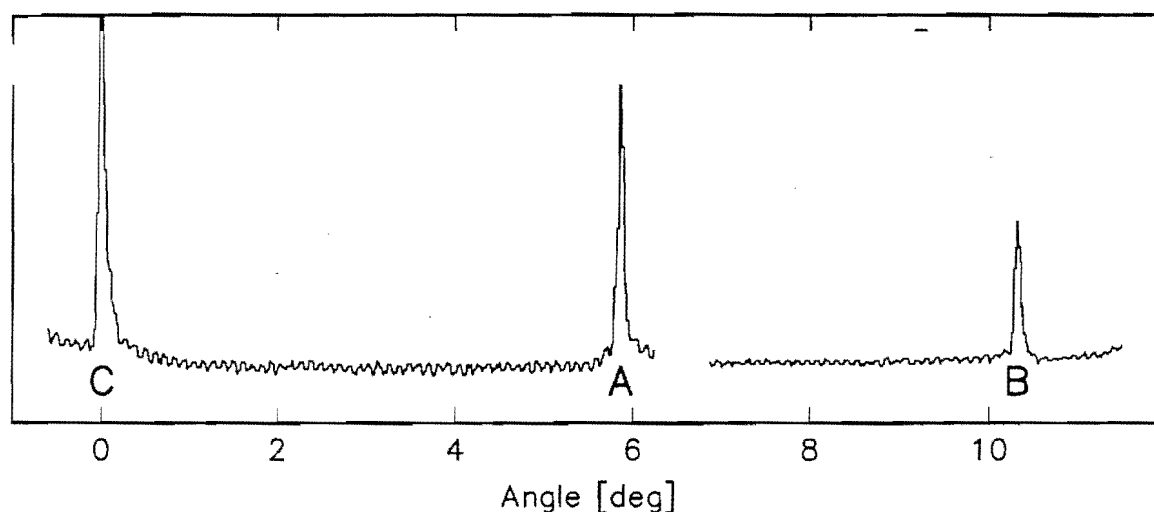


FIGURE 2: Angular dependence of the second-harmonic intensity from spontaneous NCFD at room temperature, angle as measured outside the crystals. A:  $\text{LiNbO}_3$  with 48.25 % Li, B:  $\text{LiNbO}_3$  with 48.4 % Li, C: central spot.

Fig. 2 shows the angular dependence of the generated second-harmonic intensity for two different  $\text{LiNbO}_3$  samples. Besides the central spot (C), a sharp peak for each sample (A, B) at a certain angle  $\varphi$  can be detected. This angle is characteristic for the composition of the respective sample, the lithium content was 48.25 % for sample A and 48.4 % for sample B. From the peak widths and the peak positions for the two samples a sensitivity about 0.003 % for composition measurements of  $\text{LiNbO}_3$  can be estimated.

From the broadening of the NCFD peak, the homogeneity of the sample within the volume illuminated by the fundamental beam can be characterized. An example is shown in fig. 3. The peaks for a rather homogeneous (left) and a slightly inhomogeneous sample (right) are depicted in expanded angular resolution. The peak width of the inhomogeneous sample corresponds to a composition inhomogeneity of about 0.01 %.

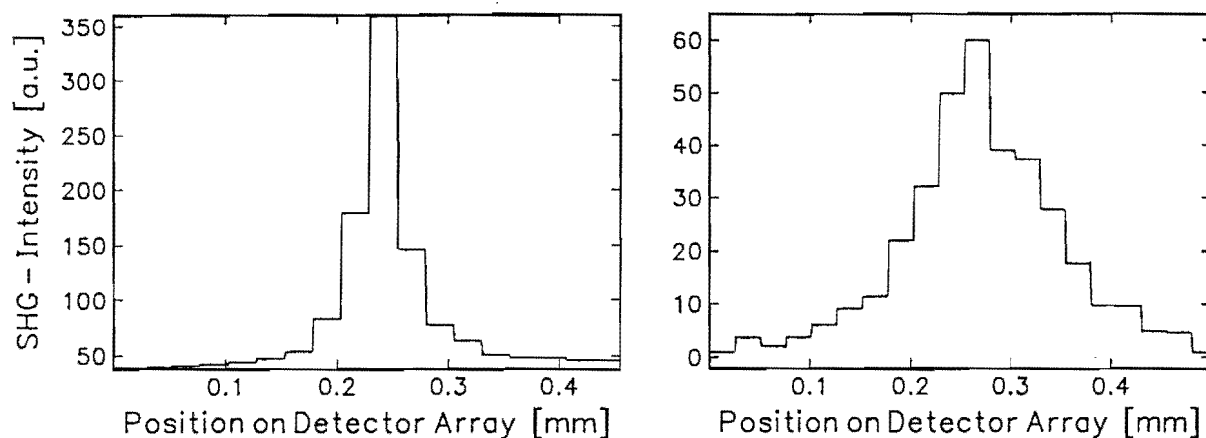


FIGURE 3: Expanded view of the NCFD peak for a rather homogeneous sample (left) and a slightly inhomogeneous one (right)

### INDUCED NCFD

While in spontaneous NCFD one of the fundamental beams is taken from scattered light, so automatically fulfilling the phase matching condition (eq. 2), in the case of induced NCFD each of the fundamental beams is controlled independently. This can be achieved in an experimental setup as sketched in fig.4.

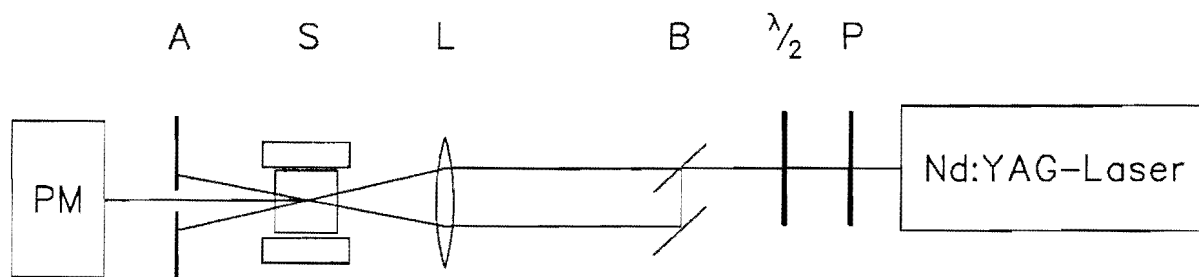


FIGURE 4: Experimental arrangement for measuring induced NCFD. P: polarizer,  $\lambda/2$ : half-wave plate, B: beam splitter, L: lens, S: temperature controlled sample holder, moveable in 3 directions, A: aperture for blocking the fundamental beams.

With the minor drawback that the angle between the fundamental beams and/or the sample temperature has to be adjusted for phase matching, induced NCFD has the obvious advantage that the interaction volume within the sample is limited in all three spatial dimensions. So for the first time a three-dimensional sample characterization is possible. To demonstrate the possibility of spatial resolution in beam direction, we carried out preliminary measurements on a model crystal. The 'crystal' was formed taking two  $\text{LiNbO}_3$  samples of different composition, thus a step-like spatial inhomogeneity was achieved. This model crystal was moved in beam direction, for several positions the temperature depen-

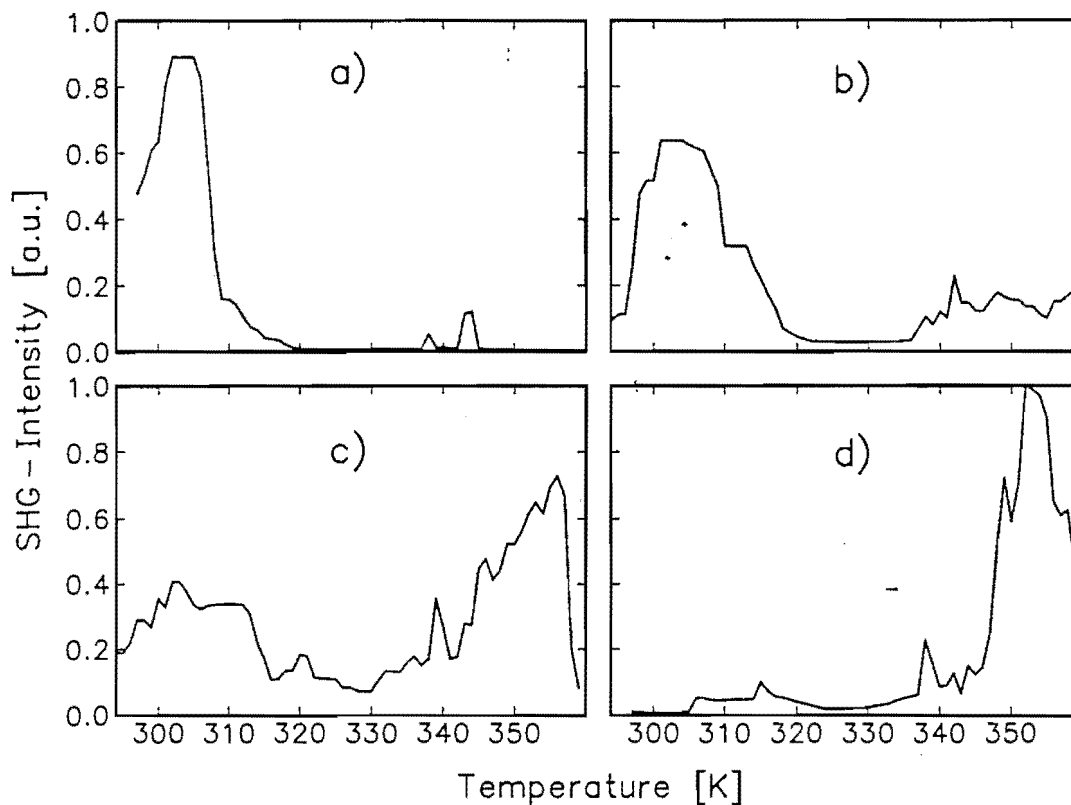


FIGURE 5: Temperature dependence of the second harmonic intensity for a model crystal. From a) to d) the sample was moved 5 mm each.

dence of the second-harmonic intensity was measured. Typical results are shown in fig. 5, the compositional crossover can be clearly detected.

## CONCLUSION

Non-collinear frequency doubling has been shown to be an interesting tool for characterizing electrooptic crystals. In the case of  $\text{LiNbO}_3$ , with 'spontaneous' NCFD a sensitivity of about 0.003 % for composition checks can be achieved. This is comparable to the conventional method of measuring the phase-matching temperature<sup>16</sup>. 'Induced' NCFD offers for the first time the possibility of three-dimensional crystal characterization with a — nondestructive — optical technique.

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