Induced noncolinear frequency doubling: A new characterization technique for electro-optic crystals

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Induced noncolinear frequency doubling (INCFD) is presented as a new characterization technique for electro-optic crystals. Using the interaction of two noncolinear laser beams for the generation of second harmonic light, the method yields a three-dimensional spatial resolution. First experimental results of INCFD measurements on composition gradients in lithium niobate and on the spatial orientations of 90°-domain walls in potassium niobate are shown. © *1996 American Institute of Physics.* [S0021-8979(96)04304-1]

I. INTRODUCTION

In the last decades the demand for materials which are applicable in electrooptics has strongly increased. New materials were developed, 'classic' materials could be improved. The properties of most of these materials can be widely tuned by varying composition and doping.

The great variability in turn necessitates suitable methods for checking composition and doping in order to control the fabrication process. Usually these methods do not measure the desired parameters directly but other physical properties which depend strongly on composition or doping. Typical methods for the case of, e.g., lithium niobate are the measurement of the Curie temperature,[1] optical absorption edge,[2] refractive indices,[3] birefringence,[4,5] or phase-matching temperature for optical second harmonic generation.[6,7]

All these techniques utilize the fact that the dielectric function of the material is influenced by composition and doping in a characteristic way. The high or low frequency, real or imaginary part of the function then is taken as the characterizing property. The sensitivity in general is enhanced when a difference measurement is possible as, e.g., in the case of second harmonic generation where the temperature is tuned such that the difference between the two relevant refractive indices vanishes. With some of the techniques focussed light beams can be used for the measurement thus yielding a topographical resolution in up to two spatial dimensions.

Here we describe induced noncolinear frequency doubling (INCFD) as a new characterization technique which — for the first time — allows three-dimensional spatial resolution. This method utilizes two fundamental light beams inclined to each other to generate second harmonic light. The interaction volume is limited in all three spatial dimensions thus offering the possibility for a three-dimensional characterization with a nondestructive technique.

II. PRINCIPLES OF THE METHOD

A. General

In the well-known case of conventional—colinear—second harmonic generation, phase matching is achieved when the quasi-scalar relation for momentum conservation

$$|\vec{k}_2| = |\vec{k}_1| + |\vec{k}'_1| \tag{1}$$

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 - all parallel to each other.

The phase matching temperature is the parameter which characterizes the crystal composition when noncritical phase matching for second harmonic generation (SHG) is applied. Using a focussed fundamental laser beam allows one to perform a two-dimensional investigation of the crystal composition with a high spatial resolution (spatially resolved second harmonic generation, SRSHG).[8] Similar results can be achieved by using an expanded laser beam and a two-dimensional detector array.[9]

In contrast to this, induced noncolinear frequency doubling (INCFD) utilizes two light beams inclined to each other to fulfill the vectorial phase matching condition

$$\vec{k}_2 = \vec{k}_1 + \vec{k}'_1. \tag{2}$$

Due to the interaction volume limited in all three spatial dimensions a fully three-dimensional topographical resolution is possible. Furthermore, the angle enclosed by the two fundamental beams, i. e. the angle between $\vec{k}_1$ and $\vec{k}'_1$, can be used as an additional experimental parameter which changes the phase matching temperature for this nonlinear process.

For a typical electrooptic material with negative uniaxial birefringence like lithium niobate the principal configuration is sketched in Fig. 1. Two ordinarily polarized fundamental beams with frequency ω and wave vectors k_{o1} and $k_{o1'}$ inclined to each other by 2φ interact generating an extraordinarily polarized second harmonic beam with frequency 2ω and wave vector k_{e2}.

Measuring the phase matching temperature for a fixed angle 2φ as a function of the interaction volume position results in a composition or doping topography of the investigated crystal.

B. Experimental arrangement

The experimental arrangement for measuring INCFD is schematically sketched in Fig. 2. The light of a pulsed Nd:YAG–laser is split into two parallel beams of approximately equal intensity. The two beams then are focussed onto the same spot inside the sample crystal, the interaction angle 2φ is controlled by the distance between the two beams and

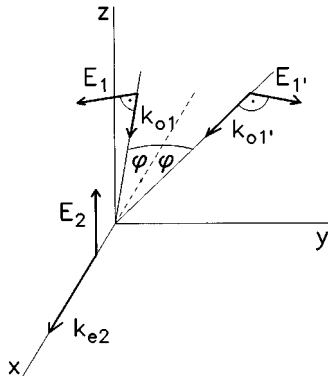---
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FIG. 1. Wave vectors and polarizations for noncolinear phase-matched second harmonic generation in a uniaxial crystal with negative birefringence, z is the optical axis of the crystal.

the focal length of the focussing lens taking into account the refraction at the sample surface. The 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.[10]

C. Limitations

The beam geometry inside the sample is sketched in more detail in Fig. 3: the two fundamental beams control the size of the interaction volume which in principle limits the spatial resolution of the method. The vectorial phase matching condition (Eq. 2) can be rewritten as an equation for the temperature dependent refractive indices of the investigated material and the angle 2φ enclosed by the interacting fundamental beams. For a material with negative birefringence like lithium niobate this results in

$$n_e(2\omega, T) = n_o(\omega, T) \cdot \cos \varphi. \tag{3}$$

Equation (3) shows that the angle φ is limited by the birefringence of the sample, in the case of lithium niobate the maximum usable angle φ is some 0.1 radians. This results in an interaction volume which is more extended in the x-direction than in the y-direction, the ratio X/Y is approximately equal to $\cot\varphi$.

For a laser beam the product of beam waist times divergence is a fixed magnitude, for INCFD that means that
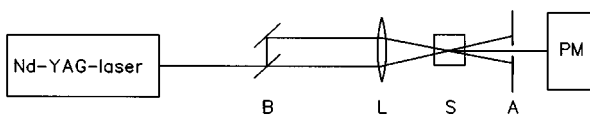

FIG. 2. Experimental arrangement for measuring INCFD. B: beam splitter, L: focussing lens, S: temperature controlled sample holder, moveable in all three spatial directions, A: aperture for blocking the fundamental beams, PM: photomultiplier.
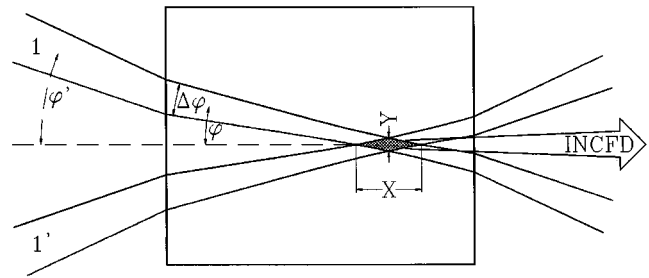

FIG. 3. Detailed beam geometry inside the sample. X and Y are the dimensions of the interaction volume which is formed by the two fundamental beams 1 and 1'. The dimension in z-direction is slightly less than Y.

$Y \cdot \Delta\varphi =$ const. According to this, an improvement of the spatial resolution can only be achieved by increasing the beam divergence which in turn relaxes the phase-matching condition. Such relaxed phase matching reduces the accuracy in the determination of the phase-matching temperature and thus in the determination of composition and doping of the investigated sample. In practice one has to compromise between spatial and compositional resolution and to choose an optimum for the respective sample.

Mathematically the measured intensity field is a special convolution integral over the sample properties and an instrument function which is mainly determined by the interaction volume. In principle this instrument function can be both calculated from the beam geometry and measured using test arrangements like, e.g., steps in the sample properties. A numerical deconvolution then should significantly improve the spatial resolution of the technique. Numerical work aiming in this direction is just being started.[11]

III. RESULTS AND DISCUSSION

As examples for the new technique two applications shall be presented.

A. Composition gradients in lithium niobate

One of the most important anorganic electrooptic materials is lithium niobate. Though generally referred to as $LiNbO_3$, it is usually grown at its congruent melting point exhibiting a significant deficit of Li_2O. To fabricate stoichiometric $LiNbO_3$, several techniques have been developed, e.g., growing in potassium-enriched melt[12] or vapor transport equilibration (VTE).[13,14] The latter, a post-growth heat treatment in $LiNbO_3$ powder, has the ability to produce every desired composition between the congruent (48.4 % Li_2O) and the stoichiometric one (50 % Li_2O).

As VTE is a diffusion technique, it is of great interest to monitor the generated diffusion profiles in order to control the whole treatment. This monitoring task can be done by INCFD.

For our investigations we fabricated a sample crystal using the VTE technique described by Jundt et al.[14] We used an approximately $4\times4\times4$ mm sized $LiNbO_3$ crystal cut from a boule which had been congruently grown by the Czochralsky method. The crystal was treated by VTE for about 200 h at a temperature of 1100 °C, indiffusion of Li was
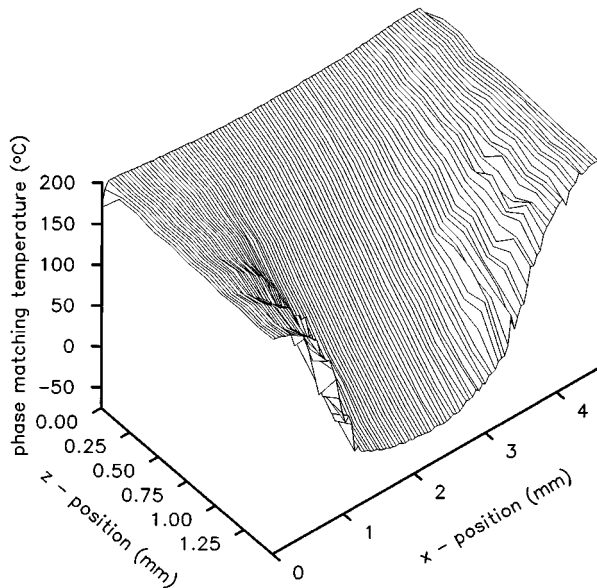
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FIG. 4. Topography of the INCFD phase matching temperature in the central xz-plane of a VTE treated LiNbO$_3$ crystal (for a better overview only about half of this topography from z=0—the edge of the crystal—up to z=1.5 mm is drawn). The phase matching temperature is a measure for the Li content in the crystal.

FIG. 5. SHG-intensity as a function of the temperature and the z-position of a VTE treated LiNbO$_3$ crystal. The white line marks the maximum intensity and thus the phase-matching temperature. The right axis denotes the Li content which corresponds to the respective NCFD phase-matching temperature (see Ref. 15) (as here only an impression of the accuracy shall be given, an approximated linearized scale is used).

permitted from each of the six crystal faces. The resulting diffusion topography in two directions (x and z) as measured by INCFD is shown in Fig. 4. 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.

To get the diffusion profiles in certain crystallographic directions, from the complete picture these directions can be extracted. This is done in Fig. 5 for the z-direction. To get an estimate of the spatial resolution of the measurement, the distribution of the second harmonic intensity is shown as shaded area. Concerning the halfwidth of the second harmonic intensity distribution, the resolution is approximately 30 μm perpendicular to the main beam direction and 200 μm in beam direction. In a simple diffusion geometry like in the VTE example the detection of the intensity maximum—thus the effective resolution—of course can be done with better accuracy.

From the profile measurements the diffusion constants for Li in lithium niobate can be derived.[16] For our treatment temperature of about 1100 °C we find that the diffusion constants parallel and perpendicular to the z-direction (polar axis) are slightly different and vary as a function of the composition[17]:

$$D_\parallel = 1.8 \ldots 6.5 \times 10^{-9} \ \text{cm}^2/\text{s} \quad \text{for } c_{Li} = 48.5 \ldots 49.8\%$$
$$D_\perp = 1.0 \ldots 6.5 \times 10^{-9} \ \text{cm}^2/\text{s} \quad \text{for } c_{Li} = 48.5 \ldots 49.8\%.$$

The exact composition dependence of the two principal diffusion constants is depicted in Fig. 6.

B. Domain walls in potassium niobate

INCFD—as generally second harmonic generation – depends strongly on the orientation of the investigated crystal.
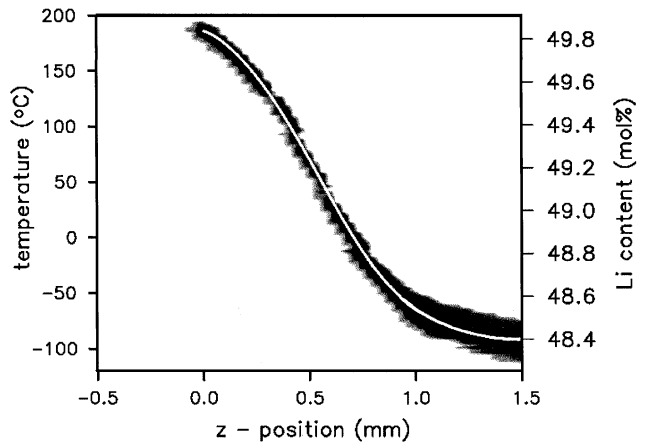
As both the nonlinear susceptibility and the refractive indices abruptly change when the geometrical orientation of the crystal changes, INCFD can be used for a topographical detection of domain boundaries inside a crystal. Domain boundaries will show up as intensity steps in the three-dimensional INCFD intensity field. These intensity steps can be automatically detected using numerical differentiation and evaluating the maxima of the derivative field.

An example is given in Fig. 7. INCFD was measured on a potassium niobate crystal with two domains oriented 90 ° to each other. The overall size of the investigated part of the crystal was about 0.3×0.6×1.1 mm. The measurement was done at constant temperature using highly focussed beams (working with highly focussed beams and thus enlarged beam divergence $\Delta \varphi$ is no drawback in this type of measurements as only a high spatial resolution in the detection of the domain boundaries is of importance). The angle between the interacting fundamental beams was chosen such that phase matching was achieved in the larger of the two domains ($\varphi' = 8.8°$, T=190 °C, usage of the tensor element d$_{32}$). Do-
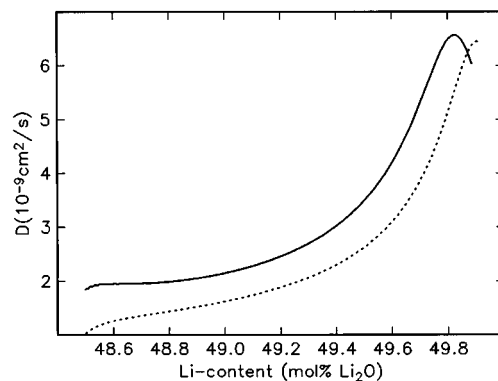

FIG. 6. Composition dependence of the diffusion constants for Li in lithium niobate at 1100 °C parallel (solid line) and perpendicular (dashed line) to the polar axis (z-direction) of the crystal.
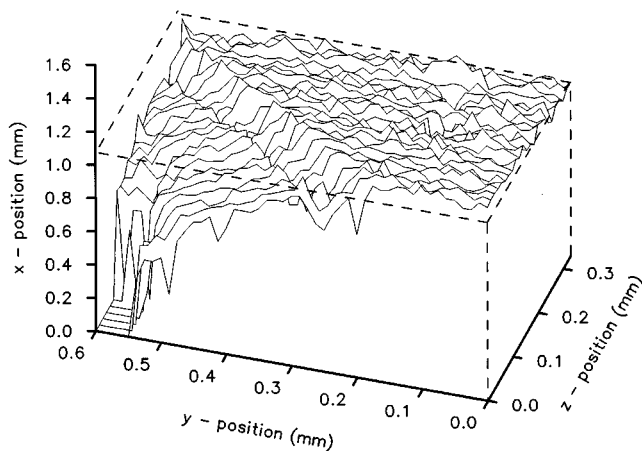
FIG. 7. INCFD measurement on a KNbO$_3$ crystal for detecting the boundary between two ferroelectric domains. The outline of the investigated crystal is indicated by dashed lines. The solid lines connect the maxima of the three-dimensional derivative field which was generated by a numerical differentiation of the INCFD intensity field. At the right the lines indicate the border of the crystal, at the left the domain boundary.

ing this, optimal contrast and – in combination with the strong focussing – a spatial resolution of approximately 10 μm could be achieved.

In Fig. 7 the maxima of the derivative field are connected by solid lines. At the right these lines indicate the border plane of the crystal, at the left the border between the two 90 ° domains in the crystal.

IV. CONCLUSION

Induced noncolinear frequency doubling has been shown to be a powerful technique for the topographic characterization of electro-optic crystals. A three-dimensional nondestructive investigation of compositional or orientational variations is possible with excellent accuracy and high spatial resolution.

In materials like, e.g., lithium niobate, compositional variations can be measured with an accuracy corresponding to approximately 0.01 mol % of Li content. The spatial and compositional resolutions have to be compromised according to problem requirements. Spatial resolutions down to about 10 μm are possible—presently at the cost of relaxed compositional accuracy.

An expansion of the technique by appropriate numerical deconvolution methods—just being started—should significantly improve the resolution.

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