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SPATIALLY RESOLVED SECOND HARMONIC GENERATION AND INTERFEROMETRIC INVESTIGATIONS OF $LiNbO_3$ CRYSTALS†

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Two methods of spatially resolved second harmonic generation (SRSHG) – (1) focused beam and moving crystal, (2) uniformly illuminated crystal and detector array – and an interferometric method are used to investigate the optical homogeneity of various $LiNbO_3$ samples. The experimental results of a crystal with Li-outdiffused layer show up typical differences which can be described by a beam propagation in a profile with variable refractive index. The detector array method is also applied to characterize imhomogeneities in samples with different Mg-contents. A comparison with Mach–Zehnder interferometry (MZI) shows that SRSHG yields more information especially in very inhomogeneous crystals.

Extrinsic impurities and the Li/Nb ratio are important for optical devices of $LiNbO_3$ crystals because of their influence on the refractive indices. While impurities like Ti or H are used to fabricate waveguides,[1–3] Mg greatly reduces optical damage,[4] and, together with the Li/Nb ratio varies the phase-matching conditions for nonlinear optical devices.[5] In turn, the optical properties of $LiNbO_3$ crystals – e.g. the refractive indices and their variations – can serve as a measure for other crystal properties – doping homogeneity etc. Index variations in a crystal can be measured interferometrically[6] or – as $LiNbO_3$ shows noncritical phase matching for second harmonic generation from Nd–YAG wavelength at convenient temperatures – by means of temperature dependent spatially resolved second harmonic generation (SRSHG).[7] We applied both the interferometric and the SRSHG method to two types of $LiNbO_3$ samples:

1 undoped congruent samples with Li-outdiffused surface layer,
2 various Mg-doped samples grown from congruent melt.

The interferometric investigations were carried out in a modified Mach–Zehnder interferometer briefly sketched in Figure 1. By means of the polarizer, the ordinary or the extraordinary index of the crystal could be selected. For the SRSHG measurements the two experimental setups shown in Figure 2 were used: (a) the beam of a pulsed Nd–YAG laser was focused onto the sample crystal which could be moved perpendicular to the beam direction by a computer controlled translation table, the generated second harmonic signal being detected by a photodiode; (b) the second harmonic signal generated by the crystal from an expanded Nd–YAG laser beam was detected spatially resolved by means of an optical multichannel analyzer. In both setups the temperature could be controlled by Peltier cooling or oven heating within a range of −25 to 200°C with an accuracy of about 0.3 degrees.

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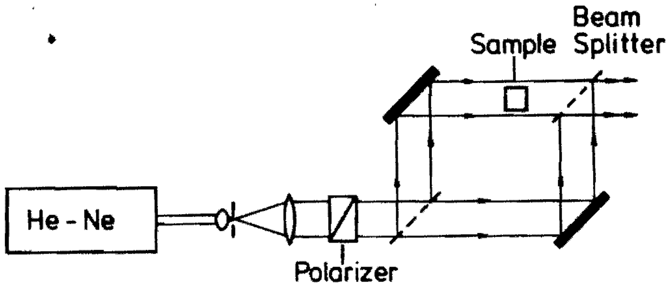

FIGURE 1 Mach–Zehnder interferometer used for the interferometric measurements.

For testing the described methods a Li-outdiffused layer was prepared by annealing a $LiNbO_3$ crystal in an oxygen atmosphere at 1100°C for 24 hours. Li outdiffusion causes an increase of the extraordinary refractive index in the surface layer of the crystal. This can be visualized by Mach–Zehnder interferometry (see Figure 3). The SRSHG results for this sample are shown in Figure 4, where contour lines for equal second harmonic intensity as a function of temperature and position in the crystal are plotted. Figure 4a shows the contour plot before annealing where a rather homogeneous phase matching temperature – which is equivalent for constant refractive indices – is found throughout the crystal. Figures 4b and 4c show the results for the annealed crystal measured with the focused beam method (4b) and the optical multichannel analyzer method (4c), respectively. An exponential-like decrease in the phase-matching temperature to the crystal surface is found indicating a corresponding increase in the extraordinary index of refraction. The reason for the smoother decrease in Figure 4c is a spatial mixture of different second harmonic waves in the case of the optical multichannel analyzer method. To check this assumption, a model calculation[8] was carried out simulating the wave propagation in an exponential index profile and detection by an optical array detector. The result of the calculation is shown

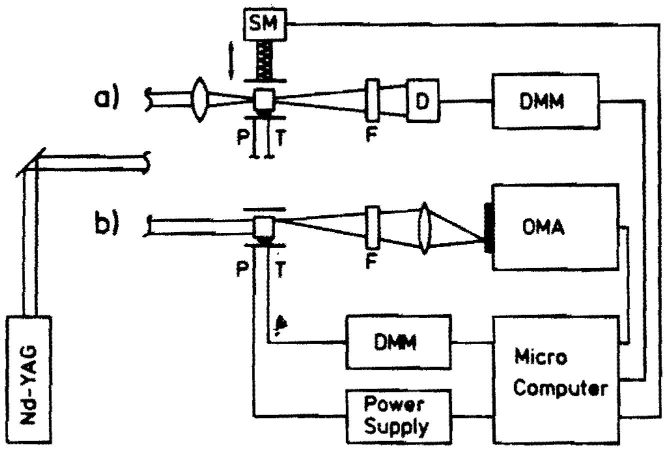

FIGURE 2 Experimental setups used for the SRSHG investigations: a) focused-beam method, b) detector-array method (SM: stepping motor, P: Peltier cooling or oven, T: thermocouple, F: optical filter for separating the generated harmonic light from the fundamental wave, D: photodiode, DMM: digital multimeter, OMA: optical multichannel analyzer). The Nd–YAG laser was driven in pulsed mode with a peak power of about 5 kW and a repetition rate of 1 kHz.

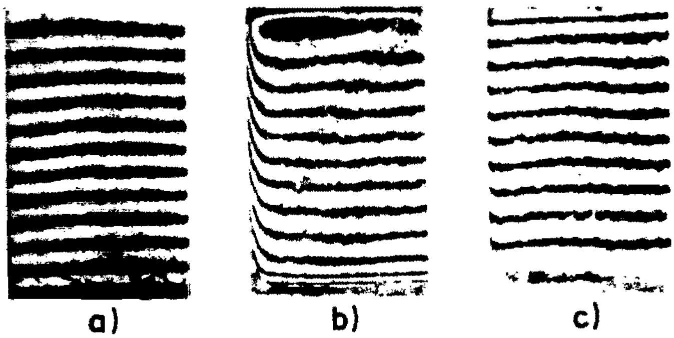

FIGURE 3 Interferometric view of the Li-outdiffused sample: a) before, b) extraordinary, and c) ordinary light polarization after outdiffusion.

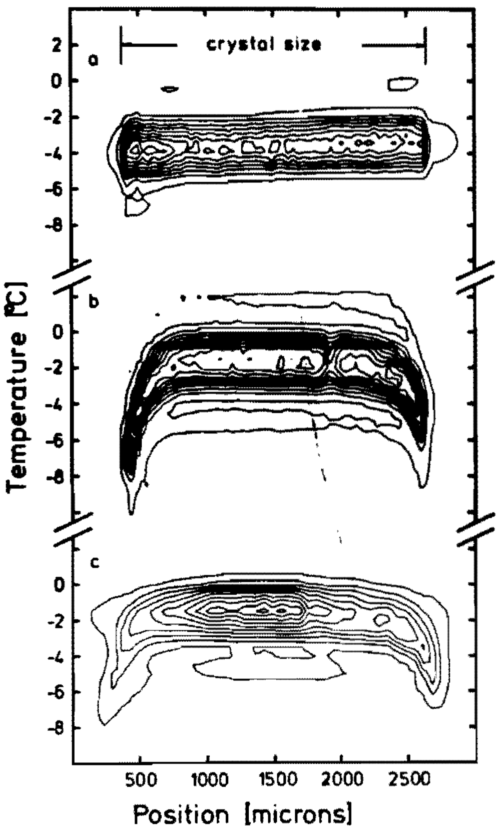

FIGURE 4 Second harmonic intensity contour plots for the Li-out-diffused sample, a) before outdiffusion, b) and c) after outdiffusion measured with focused-beam method (b) and detector-array method (c).

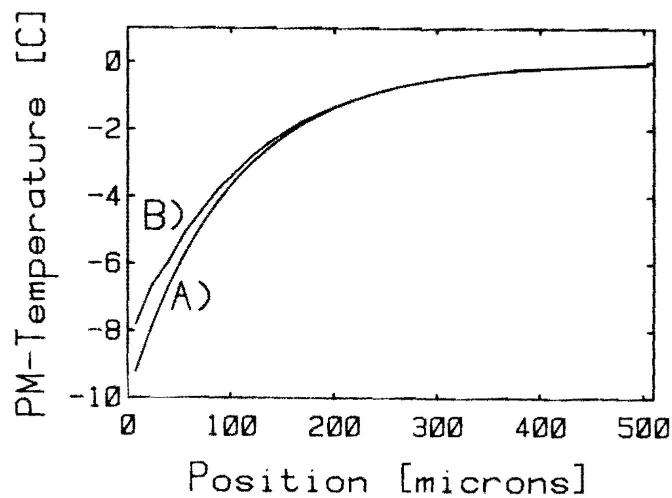

FIGURE 5 Model calculation for the measured phase-matching temperature in an exponential index profile: A) focused-beam method, B) detector-array method.

in Figure 5: A) phase matching temperature corresponding to the (assumed) real index profile (as measured by focused beam method), B) phase matching temperature as 'seen' by optical multichannel analyzer method. The calculation shows that large deviations occur between the two methods when large index gradients show up in the crystal. In spite of this disadvantage, for the measurements on Mg-doped crystals the optical multichannel analyzer method was applied because of its obvious multiplex advantage.

The magnesium-doped samples we investigated were grown by Grabmaier *et al.*[9] from congruent melt with Mg-concentrations (in melt) between 0 and 9%. In some of the crystals growth striations can be visually detected, the nature of which is still unclear. A typical crystal (6% Mg) with well visible striations is shown in Figure 6a. Mach–Zehnder measurements of this crystal (6b for extraordinary, 6c for ordinary index) show variations in the indices of refraction but do not give any information about the striation lines. SRSHG measurements of this crystal are plotted in Figure 7 as perspective view for better visualization and in Figure 8 as intensity contour plot. From these measurements large variations in the phase-matching temperature can be derived indicating refractive

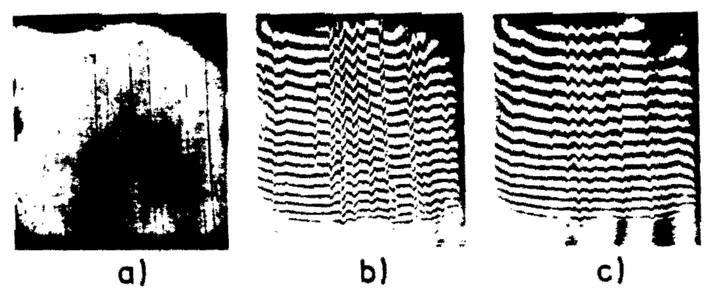

FIGURE 6 a) Mg-doped $LiNbO_3$ sample with striation lines and its interferometric views for extraordinary (b) and ordinary (c) polarized light.

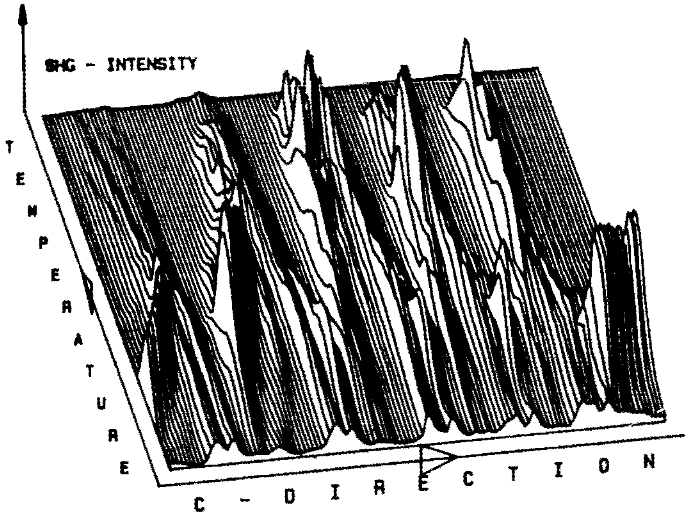

FIGURE 7 Perspective plot of the temperature and position dependence of the second-harmonic intensity in sample V104 grown from 6% Mg containing melt.

index variations and – at the border lines (or better: border planes) – relatively large steps in the index of refraction. From this fact we can interpret the striations as border planes between regions of different refractive indices which means regions of different Li/Nb ratio and/or different Mg content. All samples with visible striations showed similar results concerning the index inhomogeneities. The measured phase-matching temperatures of the different samples are plotted in Figure 9. They are in good agreement with results of Zhong Ji-guo *et al.*,[10] showing a maximum at 5.5% Mg content.

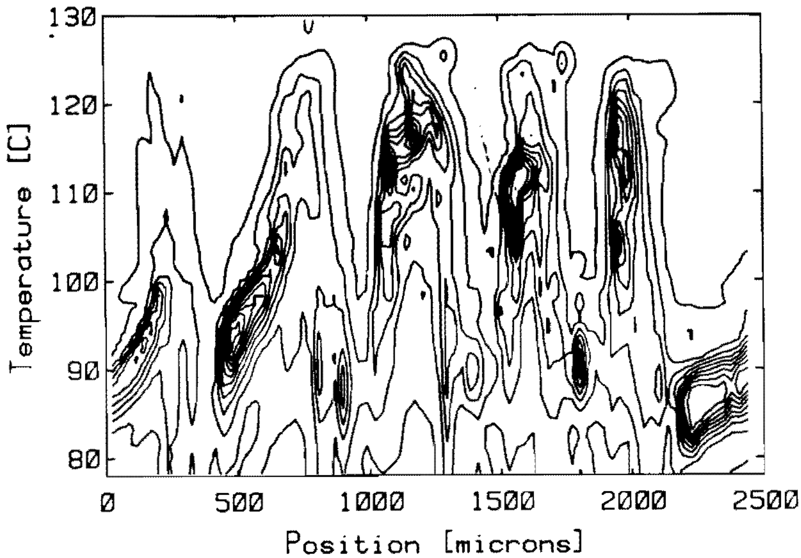

FIGURE 8 Intensity contour plot of Figure 7.

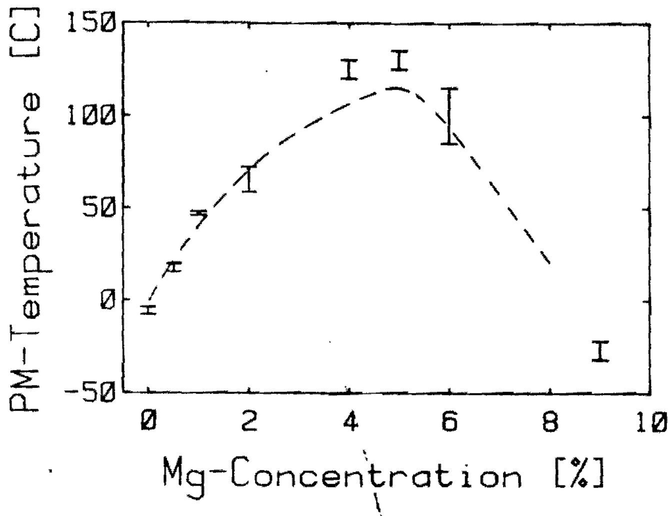

FIGURE 9 Phase matching temperatures of the investigated Mg-doped samples versus melt concentration (according to Ref. 9 the (mean) Mg content in the crystals is higher by a factor of 1.2). The dashed line represents the results of Ref. 10.

Spatially resolved second harmonic generation (SRSHG) has been proven to be a well-suited method for investigating index inhomogeneities in nonlinear crystals. A comparison with the interferometric method shows that by SRSHG additional information is gained. The method can be applied to all crystals exhibiting noncritical phase matching and tunable (for instance by temperature) phase-matching conditions.

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