Nonlinear refraction and absorption of Mg doped stoichiometric and congruent $LiNbO_3$

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The light induced change of refraction is studied in pure and Mg doped $LiNbO_3$ with congruent and stoichiometric compositions by the *Z*-scan method using all-lines visible argonion laser, up to MW/cm^2 intensity level. In Mg-doped congruent and stoichiometric crystals with Mg concentrations above threshold a positive change in the refractive index was found, in contrast to all other cases where beam fanning and negative change of the refractive index were observed. The beam distortion in the samples doped above threshold was related to thermal lensing, while below it to the photorefractive effects. It was also shown that for thermal lensing nonlinear absorption plays a dominant role. The *Z*-scan method was found to be an alternative technique to decide whether the Mg dopant level is above or below the photorefractive threshold. The damage resistance of the Mg doped samples above threshold was higher for the stoichiometric crystal than for the congruent one and increased with the amount of the built-in Mg concentration. [DOI: 10.1063/1.1635993]

I. INTRODUCTION

Lithium niobate, $LiNbO_3$ (LN) is one of the most frequently used nonlinear-optical materials, with various applications. Large crystals with congruent composition (Li/Nb ratio ~0.945) have been readily available in excellent optical quality for several decades. Congruent lithium niobate crystals (cLN) possess photorefractive (PR) properties, which is a drawback in many fields of optics, since for device applications long-term stability of the beam is required. The unwanted PR damage of LN may be suppressed by certain dopants (like Mg, Zn, or In) added in concentrations exceeding a characteristic threshold level.[1,2]

Nowadays, by the preparation of bulk stoichiometric crystals with Li/Nb ratio of ~0.98–1, LN has a renaissance. Improvements of the material properties have opened the field for further investigations and applications. In stoichiometric LN (sLN) crystals the electro-optic and nonlinear-optic effects are reported to be more marked and the coercive field to be significantly lower than in cLN.[3–5] However, undoped sLN crystals exhibit weak optical damage resistance when irradiated with a high-power laser beam.[6,7]

So far, various methods have been used to characterize the optical damage resistance of LN crystals. Almost all monitor the light induced change in the refractive index. One of the most suitable experimental tools for this purpose is the *Z*-scan method.[8] Here a focused (Gaussian) beam illuminates the nonlinear material and the sample is scanned along the propagation path around the focal plane. Information about the basic nonlinear characteristics of materials, namely the sign and magnitude of the nonlinear refraction (n_2) and the coefficient of nonlinear absorption (β), are deduced from the dependence of the far field on axis irradiance versus the position of the sample relative to the focal plane. Initially, this method was developed for "thin" nonlinear samples (for much shorter crystal length than the Rayleigh range of the focused laser beam). Later, it was completed for the case of thick samples.[9] The theoretical fitting formula of Ref. 9 contains n_2 and β as independent fitting parameters. For thin samples β can be determined by the open-aperture *Z*-scan method as well.[8] For samples having thickness with the same order of magnitude as the Rayleigh range, the fitting formulas of Ref. 8 can be used only as an approximation, since the variations in the beam size are not taken into account.

In recent articles,[10,11] we examined the light induced change of refraction in LN by the *Z*-scan method and compared 5.0 mol % Mg doped cLN and sLN crystals. We found that using an all-lines visible argon ion laser, up to MW/cm^2 intensity level, the stoichiometric crystals behave differently compared to congruent ones, namely the sLN sample shows positive (defocusing) and the cLN sample shows negative (focusing) change in the extraordinary refractive index. Such beam distortions can be the result of several effects. Besides photorefraction, the thermo-optical effect and at high beam intensity Kerr nonlinearity may lead to changes of the refractive index. While the light induced refractive index changes of Mg doped cLN crystal were related to the PR effect, in the

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