Photorefractivity of zirconium-doped lithium niobate crystals

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Reduction of lithium niobate photorefractivity is essential for optimizing all-optical communication networks.

Polarization-independent all-optical wavelength converters (AOWCs) enable dynamic signal routing, wavelength reuse, path protection, and restoration. These features will be of utmost importance in future generation, high bit-rate optical communication networks. An efficient AWOC—operating on a 100Gb/s phase-modulated polarization-multiplexed signal at 1550nm—was recently realized by exploiting the cascade of two nonlinear optical processes in a lithium niobate (LN) waveguide. This cascading technique for wavelength conversion is only effective if the LN crystals are prepared with alternating up and down domains (i.e., periodical poling) of appropriate periodicity. Currently, LN crystals offer the best performance in nonlinear devices, but their applicability is limited by the photorefractive effect, which is a change of refractive index under strong illumination. The standard growth process of LN produces lithium-deficient (i.e., congruent) crystals. The presence of defects in the congruent crystals gives rise to photorefractivity, which is detrimental to the efficiency of nonlinear interactions. To avoid this, one can operate the LN-based wavelength converters at temperatures above 100°C. Since this is impossible for most in-field applications, crystals with negligible photorefractivity must be developed. Here, we explore the use of zirconium-doped congruent LN crystals (Zr:LN) for AOWC and other optical devices.

Photorefractivity can be reduced by employing stoichiometric LN or magnesium ion (Mg²⁺)-doped LN. However, neither approach is fully satisfactory because it is often difficult to grow large crystals of high optical quality. Also, there are difficulties associated with the fabrication of periodically poled waveguides. Alternatively, the photorefractivity of congruent LN crystals can be strongly reduced by doping the crystal with tetravalent ions, such as hafnium (Hf⁴⁺) or zirconium (Zr⁴⁺). Importantly, the concentration required to obtain a substantial reduction of photorefractivity (i.e., the threshold concentration) for Zr⁴⁺ should be much lower than that of Mg²⁺ (5.5mol%) because of the increased charge. This is advantageous because lower dopant concentrations should simplify the growth of large, homogeneous crystals and also the fabrication of periodically poled waveguides.

We grew our crystals—using the Czochralski technique—from a melt composed of equal amounts of lithium and niobium oxides. We added zirconium dioxide (ZrO₂) as the dopant, in a variable range between 0 and 3mol%. With these samples in hand, we then examined their optical properties.

Figure 1. Birefringence variation (δΔn) induced on lithium niobate crystals, as a function of the zirconium (Zr) dioxide doping concentration, at pump-beam intensities of 300 (○), 600 (▲), 900 (●), and 1200 (■) W/cm².

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If we define $\Delta n_o$ and $\Delta n_e$ as the photoinduced changes of the ordinary and extraordinary index of refraction, respectively, the change in light-induced birefringence is $\delta \Delta n = \Delta n_e - \Delta n_o$. We measured this using a highly sensitive Sénarmont apparatus. We induced the photorefractive effect using a 532nm laser beam linearly polarized along the optical axis of the crystal and propagating perpendicular to it. We used a helium-neon laser probe beam as the output, operating at 632.8nm. Figure 1 shows the behavior of the induced birefringence as a function of $\text{ZrO}_2$ concentration. The plots clearly suggest a Zr$^{4+}$ threshold concentration of ~2mol% above which the photorefractive effect is strongly reduced.

We then investigated the photorefractive behavior at greater beam intensities using a qualitative approach to directly observe the distortion of the light transmitted by the crystal. We focused a 532nm laser beam using a convex lens onto a 4mm-thick crystal wafer placed in the focal plane. When the laser intensity exceeds a certain value, the transmitted light beam spot is smeared and elongated along the optical axis, with decreased intensity at the center. Figure 2 shows the transmitted light spots for six different crystals that were irradiated with a laser power density of 7kW/cm$^2$ over several minutes. We recorded the images in the far field, ~1m from the beam focus. The 3mol% Zr:LN crystal withstood a high-intensity beam without noticeable smearing, whereas the beam crossing the 2.3mol% sample showed some distortion. Importantly, this analysis emphasizes that the low-intensity behavior described in Figure 1 is insufficient for assessing suppression of photorefractivity.

In summary, we have demonstrated the dependence of doping concentration of Zr:LN crystals in both low- and high-intensity experiments. Photorefractivity can be essentially eliminated with Zr$^{4+}$ concentrations between 2.3 and 3mol%. Importantly, Zr:LN crystals are promising candidates for efficient cascaded wavelength converters operating at room temperature. Our future efforts will involve the fabrication of periodically poled Zr:LN substrates and waveguides.

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