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Spatially-Controlled Second Harmonic Generation in Lithium Niobate Silicate and Borosilicate Glasses

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Abstract: This work investigates the role of B₂O₃ addition into a silicate glass matrix on the orientational dependency of second harmonic generation (SHG), induced by LiNbO₃ crystals after femtosecond laser irradiation.

1. Introduction

Femtosecond (fs) laser direct writing (FLDW) is a powerful and versatile tool to enable a large variety of optical components to be fabricated in transparent materials such as glasses (e.g., Bragg gratings, waveguides, graded index lenses, birefringent optics) [1,2]. By controlling the fs-laser parameters (polarization, pulse energy, repetition rate, scanning speed) it becomes possible to induce controlled modifications inside the glass, hence local properties. As an example to this approach, LiNbO₃ crystals, having properties such as piezoelectricity and ferroelectricity, were induced in a 33%Li₂O–33%Nb₂O₅–34%SiO₂ (molar percentage) glass matrix (LNS) [3]. It was shown that the orientation of the polar axis of the LiNbO₃ nanocrystals can be dictated by controlling the writing laser polarization. Because of axial symmetry, the second order susceptibility tensor (χ(2)) of LiNbO₃ crystal is such that its nonlinear coefficient value along the polar axis (d₃3 = 34.4 pm/V) is much greater than the nonlinear coefficients values along the axes perpendicular to it (d₁₁ = 5.95 pm/V, d₂₂ = 3.07 pm/V). Therefore, a angular modulation of χ(2) properties, such as second harmonic generation (SHG), occurs provided the crystals can be oriented in a non-random fashion.

In this context, the aforementioned work is extended and the role of the glass matrix on the crystal orientability is investigated herein. Furthermore, the partial substitution of SiO₂ with B₂O₃ is of interest since introduction of B₂O₃ lowers glass-making temperature. Consequently, addition of B₂O₃ is expected to modify the kinetics of LiNbO₃ crystallization and the thresholds of the different regimes found for LNS glass. For this purpose, anisotropy of the SHG property is investigated in both static (writing speed = 0 µm/s) and scanning (writing speed ≠ 0µm/s) fs-laser irradiation conditions.

2. Experimental procedure

Four glasses, with compositions given in Table 1, are fabricated using conventional melt-quenching technique, and are shaped into plates of ~1 mm thickness upon cooling. Both top and bottom surfaces of each sample are then polished until optical roughness is reached.

Table 1. Glass labels and batch compositions (molar percentage)

<table>
<thead>
<tr>
<th>Label</th>
<th>Glass batch composition (molar percentage)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LNS34</td>
<td>33%Li₂O–33%Nb₂O₅–34%SiO₂</td>
</tr>
<tr>
<td>LNS27B7</td>
<td>33%Li₂O–33%Nb₂O₅–27%SiO₂–7%B₂O₃</td>
</tr>
<tr>
<td>LNS20B14</td>
<td>33%Li₂O–33%Nb₂O₅–20%SiO₂–14%B₂O₃</td>
</tr>
<tr>
<td>LNS13B21</td>
<td>33%Li₂O–33%Nb₂O₅–13%SiO₂–21%B₂O₃</td>
</tr>
</tbody>
</table>

Each glass sample is thus irradiated using a femtosecond laser (λ = 1030 nm, 300 fs, NA = 0.6) focused shifted by 350 µm below the surface. A first set of experiments consists of static irradiations to investigate the incubation time until SHG is detected online, through the observation of an intense green light characteristic of SHG signal at λSHG=λ/2=515 nm. In this experiment, laser parameters are varied, including pulse energy (from 0.25 to 2.5 µJ, step of 0.25 µJ) and laser repetition rate (100 kHz, 200 kHz, 340 kHz, and 510 kHz). A second set of experiments is to write lines (fixed frequency of 200 kHz and writing speed of 1 µm/s) using different pulse energies (0.25 µJ to 2.0 µJ, step of 0.25 µJ) and for three different laser polarization configurations: parallel (||), at 45°, and perpendicular (⊥) to the laser writing direction. Following this experiment, the angular dependency (total of 180°) of the SHG intensity for each irradiated line is measured according to the azimuthal angle (θ) of the polarization of the probing beam.

3. Results and discussion
Fig. 1a and 1b display the incubation time (in seconds), under static irradiation, to observe intense green light emission, characteristic of SHG. The comparison of the results for two glass samples, LNS34 and LNS20B14, illustrates well the fact that an addition of B_2O_3 typically promotes fast crystallization, especially at low repetition rates. These data are consistent with differential scanning calorimetry (DSC) measurements, to be discussed at the conference, showing that borosilicate samples (LNSB), relative to the silicate (LNS) one, tend to crystallize at lower temperatures.

In Fig. 1c and 1d are reported the normalized SHG intensity, measured for both LNS34 and LNS20B14 samples, in different writing configurations (i.e. //, at 45°, and ⊥) with respect to the writing direction (axis Y in Fig. 1c or 1d), at a fixed pulse energy value of 0.5 µJ. In both samples, the SHG intensity is strongly dependent on the probing polarization azimuthal angle (θ), and its maximum is found perpendicular to the writing polarization direction. This is consistent with findings in Refs. [3,4], where the polar axis (associated with maximum SHG intensity) is aligned perpendicular to the laser polarization (at low pulse energy). At higher pulse energies, the situation is more complex, and the maximum of SHG signal depends on both writing polarization conditions and glass matrix composition. These results will be discussed at the conference.

4. Conclusions

Lithium niobate silicate and borosilicate glasses are fabricated, and are subsequently irradiated using a femtosecond laser to induce non-centrosymmetric nano-crystals. SHG measurements are performed in order to detect crystallization and the anisotropy in the SHG response reveals a crystal preferential orientation induced by the irradiation. It is shown that substitution of SiO_2 with B_2O_3 preserves tunability of SHG. The borosilicate glass family is advantageous due to its lower melting temperature. Details with respect to application possibilities will be discussed at the conference.

5. References