A study on the effect of the core-cladding interface on thermal poling in GeO$_2$-SiO$_2$ optical waveguides.

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Abstract: GeO$_2$-SiO$_2$ multilayer glass samples are thermally poled. The interface between the layers strongly affects the thickness of the second order nonlinearity as well as its overlap with the core of the waveguide.

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Silica is naturally an amorphous material with a macroscopic inversion symmetry that prevents second-order nonlinear effects. Different poling techniques have been devised in order to break the symmetry and thus induce a significant and permanent second-order optical nonlinearity, typically of the order of 1 pm/V. Thermal poling is probably the most popular and promising of these techniques, potentially allowing the fabrication of frequency converters and electro-optic modulators based on silica optical fibres and/or waveguides. In order to fully understand its underlying physical mechanism and optimise the nonlinearity, thermal poling of bulk fused silica has been thoroughly investigated and a deeper understanding has been achieved [1, 2]. However, when made into waveguides or optical fibres, the presence of the interface between GeO$_2$:SiO$_2$ core and SiO$_2$ cladding may affect the process and, to our knowledge, a systematic study of such effects has not been conducted yet. In early works on quasi-phase-matched (QPM) second-harmonic generation (SHG) to the blue in optical fibres (NA = 0.1), the occurrence of interactions between different order modes, hinting at a nonuniform nonlinearity profile across the core, was reported [3]. These interactions were eliminated by reducing the distance between core and anodic electrode to about 1-2 μm.

<table>
<thead>
<tr>
<th>Sample</th>
<th>poling T (°C)</th>
<th>Voltage (kV)</th>
<th>time (mins)</th>
<th>overlaid depth (μm)</th>
<th>L$_{\text{MF}}$ (μm)</th>
<th>L$_{\text{etch}}$ (μm)</th>
<th>L$_{\text{exp}}$ (μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
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<td>4</td>
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<td>30</td>
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<tr>
<td>B</td>
<td>310</td>
<td>4</td>
<td>30</td>
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<td>20</td>
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<td>35</td>
</tr>
<tr>
<td>C</td>
<td>310</td>
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<td>19</td>
<td>40</td>
</tr>
<tr>
<td>D</td>
<td>280</td>
<td>4</td>
<td>10</td>
<td>5</td>
<td>$\leq$ 18</td>
<td>4</td>
<td>30</td>
</tr>
<tr>
<td>E</td>
<td>280</td>
<td>4</td>
<td>60</td>
<td>5</td>
<td>$\leq$ 18</td>
<td>6</td>
<td>38</td>
</tr>
<tr>
<td>F</td>
<td>280</td>
<td>4</td>
<td>20</td>
<td>2.5</td>
<td>18</td>
<td>8</td>
<td>32</td>
</tr>
<tr>
<td>G</td>
<td>320</td>
<td>8</td>
<td>20</td>
<td>5</td>
<td>38</td>
<td>34</td>
<td>-</td>
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<tr>
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<td>20</td>
<td>5</td>
<td>26</td>
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<td>-</td>
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Table 1. Nonlinear thickness (L) in poled germano-silicate waveguides. The pure-silica overladding, the Ge-doped and the underlying pure-silica layer all have the same thickness. L$_{\text{MF}}$ and L$_{\text{etch}}$ refer to values obtained from Maker's fringe and etching measurements respectively. L$_{\text{exp}}$ refers to the values reported in [4] for bulk Hazell glass.

However, according to the results on thermal poling of bulk silica [4], the nonlinear region should have extended for about 20 μm under the anode, thus including the whole core and producing a uniform profile. More recently, studies on QPM SHG at around 1.5 μm found that, even after reducing the core-anode distance to the minimum loss-allowed value (6 μm for NA = 0.065), the efficient generation of second-harmonic in the LP$_{11}$ mode could not be eliminated [5], again hinting at a nonuniform nonlinearity profile. It is very likely that an interface problem occurs, which must be investigated to optimise the process for the fabrication of
integrated optical devices. In this paper we present the results of the experiments carried out on three-layer germano-silicate waveguides poled under different temperature and voltage conditions and then assessed using the Maker's fringe technique in order to measure the thickness and the value of the nonlinearity. The samples were fabricated by using modified chemical vapour deposition to deposit a three layer glass structure on a low grade silica substrate. The surface of the substrate was first etched using SF6 to remove any impurities from the glass surface and then fire polished. The three layer structure was fabricated by first depositing several buffer layers of silica followed by germanosilicate layers to form the waveguide region and finally another set of silica layers to form the overclad coating.

The sample's structure is shown in Fig. 1: a 2.5, 5 or 20 μm thick silica over-clad covers a 2.5, 5 or 20 μm, respectively, germano-silicate core. A third silica layer of the same thickness of the overcladding lies underneath and the whole structure is supported by a 200 μm thick low grade silica substrate. The NA of the waveguide is 0.1 corresponding to a GeO$_2$ content of about 5 mol%. The samples were poled in a vacuum jar in order to avoid electric breakdown and, more importantly, to reproduce the poling atmosphere used for optical fibres, which heavily affects the process [4]. The poled samples were assessed using a Maker's fringe technique: the pump was provided by a Q-switched mode-locked Nd:YAG laser operating at 1,064 μm, giving a coherence length for SHG in silica of about 24 μm. This technique allows one to obtain both value and thickness of the nonlinear region, providing the latter is at least 18 μm deep. The results are summarised in Table 1: in all instances a value of the nonlinearity (d$_{33}$) between 0.2-0.4 pm/V was obtained using a quartz sample as reference. The depth value obtained from the optical measurements for each sample $L_{exp}$ was cross-checked against the one obtained by HF etching of the glass $L_{etch}$ (reported in the seventh column). This was particularly important for the samples exhibiting a thickness of 18 μm, beyond the resolving power of the Maker's fringe setup. For all samples the agreement is very good. The last column in the same table, $L_{etch}$, reports the values of thickness of the nonlinear region as expected from reference [4]. The experimental results for samples D and E are illustrated in Fig. 2: in particular one can see that the two samples poled in standard conditions (280 °C, 4 kV), but for very different times give the same angle dependence, which can be fitted by a curve corresponding to a depth of 18 μm. Etching measurements show that $L \approx 5 \mu$m in both cases: the nonlinear layer is therefore confined within the overclad and does not exhibit the expected growth with poling time [4]. It is worth noting how, in the case of a 2.5 μm thick core, the nonlinearity reaches further and penetrates for about 8 μm, well beyond the core, although still much less than what expected for all silica samples. We subsequently tried to pole the samples under higher temperature and voltage, to increase the ions mobility and the electric field strength. The result of the Maker's fringe measurements regarding samples G and H, poled at 320 °C, 8 kV and 320 °C, 4 kV respectively, are shown in Fig. 3 and were confirmed by etching. The data are well fitted by a curve corresponding to a depth of 38 μm and 26 μm for sample G and H respectively. In both cases a value of the nonlinearity around 0.2 pm/V was also measured.

The reason for these results can be attributed to the GeO$_2$ core layer. The depth of the nonlinear layer for standard poiling conditions appears in fact to be limited by the core-overclad interface. The cations which are responsible for the formation of the depletion region during thermal poiling are significantly slowed down or trapped at the boundary and this limits the extent of the nonlinear layer. These findings are in
agreement with the recent results concerning thermal poling of PECVD Ge-Si-N waveguides [6, 7]; SIMS measurements show an accumulation of sodium and silver ions at both cladding-core interfaces. In that case, however, due to the much thinner layers the nonlinear region could penetrate further as we also obtained for sample F. In the case of optical fibres ion migration may still occur, although hindered, around the core. Nonetheless, the resulting distribution of charges around the core may cause screening effects thus producing a reduced and nonuniform electric field. This would explain the observation of higher order SHG interactions and the low values of the effective nonlinearity, 0.015 ps/V against the expected 0.06 ps/V from measurement in bulk, obtained to date. Poling under higher temperature and voltage effectively reduces the barrier encountered by the migrating ions at the core-clad interface, thus allowing the nonlinearity to extend much deeper into the sample. We believe these findings are of great relevance for the fabrication and optimisation of fibreised/integrated devices based on second-order optical nonlinearities.

References

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