Waveguide structures in heavy metal oxide glass written with femtosecond laser pulses above the critical self-focusing threshold


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We study the feasibility of femtosecond laser writing of optical waveguides in bulk 35PbO·35Bi2O3·15Ga2O3·15GeO2 glass, motivated by the extended transparency interval of heavy metal oxide glasses in the mid-infrared regime. Its large linear and nonlinear refractive indices cause critical self-focusing to occur even at low laser energies, leading to filamentary propagation and material damage. However, the viscosity of the laser-damaged region shows a considerable increase in the refractive index, which we attribute to a collateral, stress-induced densification due to the high pressures generated in the focal region. These regions of increased refractive index are strongly birefringent and sufficiently large to support efficient light propagation in transversally written structures. Optical waveguides with a refractive index increase ≥10⁻³ and minimal mode ellipticity have been obtained. © 2005 American Institute of Physics. [DOI: 10.1063/1.1888032]

Heavy metal oxide (HMO) glasses are formed by compound oxides with a heavy metal ion content (typically Pb or Bi) above 50 cat. %. Having phonon energies that are considerably lower than those of silicate or borate glasses, the transparency interval of HMO glasses extends into the mid-infrared (λ<7–8 μm),¹,² which makes them ideally suited for transmission applications in this spectral region. HMO glasses also show high densities, low glass transition temperatures, and excellent chemical and physical stability. Despite these exceptional properties, no effort has been made yet to produce waveguide structures or other photonic elements inside HMO glasses by means of focused femtosecond (fs) laser pulses. This lack of studies is likely related to the difficulty in achieving controlled nonlinear laser structuring in HMO glasses because of their small band gaps (Eg=2–3 eV), and large linear (n₂=2) and nonlinear (n₂=10⁻¹⁹–10⁻¹⁸ m²/W) refractive indices, compared to those of fused silica (Eg=7.5 eV, n₂=1.453, and n₂=2.4×10⁻²⁰ m²/W). The largest difficulty is imposed by the elevated n₂ values that lead to strong self-focusing,³ thus generating spatial energy distributions inside the glass that are hardly controllable.³,⁶ Optical waveguides showing a high refractive index increase have recently been demonstrated in chalcogenide glasses, which have optical properties that are similar to those of HMO glasses.⁷ However, in that work the optical depth confinement was imposed by the sample, being a 1.6 μm thin film deposited onto Si/SiO2 wafers, and the written waveguides were located at the sample surface.

The aim of this work is to study the feasibility of direct waveguide writing in bulk HMO glasses using fs laser pulses. For this study we have selected a “model” HMO glass with the composition 35PbO·35Bi2O3·15Ga2O3·15GeO2. The glass material was ad hoc synthesized from a mixture of the corresponding oxides melted in a Pt crucible at 900 °C for 30 min. After pouring the melt into preheated brass molds, the solid glass ingots were annealed for 15 min at a temperature close to the glass transition temperature Tg (~400 °C) and cooled down to room temperature at 2 °C/min. They were subsequently cut in blocks with parallel faces (1 cm × 1 cm × 1 cm) and polished. The band gap and the refractive index were determined using spectroscopic ellipsometry measurements, yielding Eg=2.0 eV and n₂=2.238 (at 800 nm). The nonlinear refractive index at 800 nm was determined to be n₂=5×10⁻¹⁹ m²/W, using Z-scan measurements.⁸ The transparency interval of this glass was measured to be 570 nm–6.1 μm.

Waveguiding structures were produced inside these glass blocks using a Ti:Al₂O₃ fs oscillator/amplifier system operating at 800 nm, a repetition rate of 1 kHz and a pulse duration of 100 fs. After attenuation, the irradiation beam was focused below the sample surface, using a long working distance (17 mm) microscope objective (NA=0.42). Using an XYZ translation stage, the sample was moved during irradiation either parallel to the incident beam (Z axis) or perpendicular to it (Y axis), producing longitudinal or transversal structures, respectively. For waveguide characterization, a cw laser beam was coupled into the previously written waveguides using an objective lens (NA=0.35). To improve the coupling efficiency, the sample surfaces were abraded and polished after the writing process. At the exit plane of the waveguides, an objective lens (NA=0.85) followed by a tube lens and a CCD camera was installed in order to record near-field images of the guided light distribution. When a diffuser is inserted before the first objective lens, transillumination images can be acquired in order to inspect the exit plane of the waveguide for laser-induced visible modifications.

Figure 1 shows the exit plane (X-Y) of longitudinal waveguides written at a constant speed of 100 μm/s. For a pulse energy of 1.8 μJ (measured at the sample site), a small dark region can be observed in the transillumination image [Fig. 1(a)]. A comparison with the corresponding near-field

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image [Fig. 1(b)] shows that weak light guiding occurs within a very small region (diameter $\approx 2 \mu m$). Interestingly, the guiding region is not located within the visibly modified region but next to it. This can be seen more clearly when both images are superimposed [Fig. 1(c)], confirming that light guiding occurs, and therefore that a positive refractive index change is induced, outside the visibly modified region. Also for structures written at higher pulse energies, light guiding is observed outside the visibly modified region [Fig. 1(d)], the latter becoming more contrasted. We have found that for all pulse energies above the material modification threshold (0.9 $\mu J$), the guiding regions are located in the vicinity of the damaged region.

Somewhat similar structures have been observed in phosphate glass IOG-1 after fs laser writing experiments during which the sample was moved at a small angle with respect to the incident beam as well as in transversal structures written in crystalline $\alpha$-quartz. Both works attributed the resulting refractive index modification to shock-wave propagation/fast quenching in the vicinity of the laser affected zone. Our results are consistent with this hypothesis, indicating a stress-induced increase of the refractive index in the off-center region. Two guiding regions on opposite sides of the laser-exposed region were reported in Ref. 9 and they have been attributed to an angled incidence of the writing laser, whereas no guiding was observed for normal incidence. In contrast, we do observe light guiding at normal incidence, although very weak and only on one side of the laser-exposed region. We attribute this difference to a larger radial extension of the stress-induced region of increased refractive index in the HMO glass. The deviation from an annular symmetry of the guiding region with respect to the central laser-exposed region is most likely related to a slight asymmetry in the spatial beam profile and correspondingly in the focal spot profile. We have checked that the position of the guiding region is not influenced by the polarization of the writing laser. While, in the case of the phosphate glass, fluorescent color centers were formed in the nonguiding laser-damaged region, we have found no significant differences in the fluorescence and Raman spectra (excited at 514.5 nm) between the nonirradiated, the damaged and the guiding regions of the HMO glass.

It is worth noting that the size of the damaged region in Fig. 1(a) is of the order of the irradiation wavelength, in spite of the enlarged focal size inside the glass due to refraction and spherical aberrations. This observation suggests that critical self-focusing plays a crucial role in the material modification. The presence of critical self-focusing in the HMO glass even at low energies, just above the structural modification threshold, can be appreciated also in the transversal waveguiding structures. Figure 2(a) corresponds to a transillumination image superimposed to a near-field image of the exit plane (X-Z) of a transversal waveguide written at a pulse energy of 1.8 $\mu J$ and a speed of 60 $\mu m/s$. The damaged region consists of a narrow filament of constant width that extends over a length of 65 $\mu m$, much longer than the depth of focus (14 $\mu m$). The length of the filament corresponds thus to the “distributed focus segment” associated to the different slices of the pulse that have a power above the critical self-focusing threshold power $P_{cr}$. According to the moving focus model described in Ref. 4. At the highest pulse energy used (7.2 $\mu J$), the induced filament has a length of about 100 $\mu m$ [partially shown in Fig. 2(b)] with several waists, most likely caused by the competition between self-focusing and plasma-defocusing. Similar structures have been described in fused silica upon filamentary propagation and damage at powers above $P_{cr}$.

As for the light guiding properties of the transversal structures, we have found that they guide light very efficiently, considerably better than the longitudinal ones. The location of the guiding regions, adjacent to the filament, can be appreciated in Figs. 2(a) and 2(b). For low pulse energies [1.8 $\mu J$, Fig. 2(a)], guiding is observed near the filament onset. For higher energies [7.2 $\mu J$, Fig. 2(b)] guiding occurs at both sides of the filament, $\approx 5 \mu m$ after the filament onset. The fields of view of Figs. 2(a) and 2(b) were selected to have a common filament onset, marked by a dotted line, although the absolute position of the filament onset shifts towards the surface with increasing energy. We conclude that a guiding region can be generated at different positions along the filament by changing the pulse energy and that these positions correspond to local intensity maxima of filamentary propagation.
beam propagation above $P_{cr}$. As for the mode profiles, the inset of Fig. 2 shows intensity cross sections extracted from the guiding images alone. Whereas a single Gaussian mode with a diameter of 4.6 μm is observed at low pulse energy, a double waveguide is formed at high energies as can be appreciated by the small dip in the center of the curve. Although the images shown in Fig. 2 were obtained by coupling 633 nm light into the waveguides, we observed comparable propagation modes for IR light coupling (800–980 nm). Moreover, comparable light guiding efficiency was observed for vertically and horizontally polarized light. The propagation losses were determined to be ≈0.7 dB/cm, taking into account Fresnel losses and mode mismatch. The refractive index increase $\Delta n$ associated to the transversal waveguides was determined from far-field measurements of the guided light intensity distribution as well as by determining the admittance angle of the light coupled into the waveguides. Both methods yield consistent values of $\Delta n = 5 \times 10^{-3}$, and little influence of the writing pulse energy on this value was observed.

In order to explore the origin of this considerable refractive index increase in the vicinity of the laser damaged region, we have analyzed the polarization properties of the written structures. For this purpose, the transillumination setup was modified by adding a polarizer before the sample, set at $-45^\circ$ with respect to the polarization of the writing laser beam, and an analyzer after the sample, set at $+45^\circ$. Figure 2(c) shows the resulting birefringence map of the transversal waveguide written at 1.8 μJ, corresponding to the region shown in Fig. 2(a). It is evident that strong birefringence is generated only in the very same region that shows light guiding in Fig. 2(a). This spatial coincidence of light guiding and birefringent region in this sample can also be appreciated in the corresponding intensity cross section (inset of Fig. 2). This result further supports the hypothesis of a stress-induced, positive, nonisotropic refractive index change in the vicinity of the region where the self-focused beam surpasses a certain power density threshold. This behavior is different from the one found in fused silica, in which birefringence is only observed within damaged regions exposed to excessive laser energy, which show high propagation losses that can be partially reduced upon thermal annealing.

In fused silica, high-quality optical waveguides are generated below the power for critical self-focusing $P_{cr}$ but above the irradiance required for optical breakdown $I_{ob}$, which is of the order of $10^{13}$ W/cm$^2$ for most dielectrics. This “window” for waveguide writing is relatively narrow for fused silica and is likely to be even narrower for HMO glass. Assuming that $I_{ob}$ in the HMO glass is comparable to that of other dielectrics, the relation $I_{ob} = P_{cr} / (\pi (d/2)^2)$ allows an estimation of the maximum spot size $d$ acceptable to induce optical breakdown without critical self-focusing to occur. Using the expression given by Marburger for $P_{cr} = (3.77 \lambda^4) / (8 \pi n_0 n_2 g_2)$, a spot size of 560 nm is obtained, much smaller than the actual spot size inside a HMO glass attainable. This implies that critical self-focusing occurs before reaching the optical breakdown threshold in these glasses and that waveguide writing can therefore only be achieved indirectly, outside the region of filamentary propagation, through stress-induced densification of adjacent regions. In fact, Marburger’s expression, derived for a cw beam, predicts such a behavior for any glass with high values of $n_0$ and $n_2$, since $P_{cr}$ is inversely proportional to the product $n_0 \times n_2$. Conveniently, the expression points out a way to potentially compensate this decrease of $P_{cr}$, namely by increasing the irradiation wavelength $\lambda$. So far, we have performed measurements at 1550 nm without observing any significant change, indicating that much longer wavelengths have to be used to avoid critical self-focusing to occur.

We can thus conclude that the most important consequence of the large linear and nonlinear refractive indices of HMO glass is that critical self-focusing occurs even at the transformation threshold. As a result, the laser energy is redistributed and absorbed along a narrow filament with a diameter of the order of the irradiation wavelength. The so-induced ultrafast temperature increase (~10$^8$ K within a few ps) at constant volume can lead to local pressures of hundreds of GPa, sufficient to substantially compress the material nearby and/or generate large residual stresses. This stress field leads to a permanent nonsotropic increase of the refractive index exceeding 10$^{-3}$ in the vicinity of the laser damaged zone of transversally written structures in the HMO glass studied, with a spatial extension large enough to support efficient waveguiding with low mode ellipticity. In contrast, only weak waveguiding is observed along longitudinal structures, possibly due to the different stress field associated to the different writing geometry.

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