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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 $35\text{PbO} \cdot 35\text{Bi}_2\text{O}_3 \cdot 15\text{Ga}_2\text{O}_3 \cdot 15\text{GeO}_2$ 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 vicinity 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 $\geq 10^{-3}$ 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 ($\lambda \leq 7\text{--}8\text{ }\mu\text{m}$),^{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 elements³ 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 ($E_g \approx 2\text{--}3\text{ eV}$), and large linear ($n_0 \approx 2$) and nonlinear ($n_2 \approx 10^{-19}\text{--}10^{-18}\text{ m}^2/\text{W}$) refractive indices, compared to those of fused silica ($E_g = 7.5\text{ eV}$, $n_0 = 1.453$, and $n_2 = 2.4 \times 10^{-20}\text{ m}^2/\text{W}$). The largest difficulty is imposed by the elevated n_2 values that lead to strong self-focusing,⁴ thus generating spatial energy distributions inside the glass that are hardly controllable.^{5,6} 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 $1.6\text{ }\mu\text{m}$ 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 $35\text{PbO} \cdot 35\text{Bi}_2\text{O}_3 \cdot 15\text{Ga}_2\text{O}_3 \cdot 15\text{GeO}_2$. The glass material was ad hoc synthesized from a mixture of the corresponding oxides melted in a Pt crucible at $900\text{ }^\circ\text{C}$ 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 T_g ($\sim 400\text{ }^\circ\text{C}$) and cooled down to room temperature at $2\text{ }^\circ\text{C}/\text{min}$. They were subsequently cut in blocks with parallel faces ($1\text{ cm} \times 1\text{ cm} \times 1\text{ cm}$) and polished. The band gap and the refractive index were determined using spectroscopic ellipsometry measurements, yielding $E_g = 2.0\text{ eV}$ and $n_0 = 2.238$ (at 800 nm). The nonlinear refractive index at 800 nm was determined to be $n_2 \approx 5 \times 10^{-19}\text{ m}^2/\text{W}$, using Z-scan measurements.⁸ The transparency interval of this glass was measured to be $570\text{ nm--}6.1\text{ }\mu\text{m}$.

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 ($\text{NA} = 0.42$). 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 ($\text{NA} = 0.35$). 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 ($\text{NA} = 0.85$) 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 $100\text{ }\mu\text{m/s}$. For a pulse energy of $1.8\text{ }\mu\text{J}$ (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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FIG. 1. Images of the exit plane of longitudinal waveguides written in HMO glass by moving the sample along the axis of the incident fs laser (z axis). Illumination and light coupling at 633 nm was performed along the z axis. (a), (b), and (c) show the same waveguide structure written at $1.8 \mu\text{J}$ as (a) a trans-illumination image; (b) a coupled near-field image; and (c) the superposition of both images; (d) corresponds to the superposition of a trans-illumination and a coupled near-field image of a structure written at $5.4 \mu\text{J}$.

image [Fig. 1(b)] shows that weak light guiding occurs within a very small region (diameter $\approx 2 \mu\text{m}$). 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 ($0.9 \mu\text{J}$), 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 beam⁹ as well as in transversal structures written in crystalline α -quartz.¹⁰ 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,⁹ 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

FIG. 2. Images of the exit plane of transversal waveguides written in HMO glass by displacing the sample along the y axis. The writing laser was incident from the left-hand side (z axis). Illumination and light coupling at 633 nm was performed along the y axis: (a) and (b) show the superposition of trans-illumination image and a coupled near-field image of waveguides written at 1.8 and $7.2 \mu\text{J}$, respectively; (c) represents the birefringence map (obtained with crossed polarizers) of exactly the same area as shown in (a). The dark channels in (a) and (b) correspond to damaged zones through critical self-focusing and filamentary propagation and the common onset is indicated by a vertical dotted line. The inset represents intensity cross sections along the x axis through the center of the corresponding coupled near-field distributions of (a) and (b) and the birefringence map (c).

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 $1.8 \mu\text{J}$ and a speed of $60 \mu\text{m/s}$. The damaged region consists of a narrow filament of constant width that extends over a length of $65 \mu\text{m}$, much longer than the depth of focus ($14 \mu\text{m}$). 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 ($7.2 \mu\text{J}$), the induced filament has a length of about $100 \mu\text{m}$ [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} .¹¹

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 [$1.8 \mu\text{J}$, Fig. 2(a)], guiding is observed near the filament onset. For higher energies [$7.2 \mu\text{J}$, Fig. 2(b)] guiding occurs at both sides of the filament, $\sim 5 \mu\text{m}$ 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

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 Δn associated to the transversal waveguides was determined from far-field measurements of the guided light intensity distribution¹² as well as by determining the admittance angle of the light coupled into the waveguides.¹³ Both methods yield consistent values of $\Delta n \approx 5 \times 10^{-3}$, 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° 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,¹¹ which show high propagation losses that can be partially reduced upon thermal annealing.¹⁴

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/cm² 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.⁵ Using the expression given by Marburger⁴ for $P_{cr} = (3.77 \lambda^2) / (8\pi n_0 n_2)$, 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 λ . 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 ($\sim 10^6$ K within a few ps) at constant volume can lead to local pressures of hundreds of GPa,¹⁵ sufficient to substantially compress the material nearby and/or generate large residual stresses. This stress field leads to a permanent nonisotropic increase of the refractive index $\geq 10^{-3}$ 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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