Heat accumulation during high repetition rate ultrafast laser interaction: Waveguide writing in borosilicate glass

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Abstract. During high repetition rate (>200 kHz) ultrafast laser waveguide writing, visible heat modified zones surrounding the formed waveguide occur as a result of heat accumulation. The radii of the heat-modified zones increase with the laser net fluence, and were found to correlate with the formation of low-loss and cylindrically symmetric optical waveguides. A numerical thermal model based on the finite difference method is applied here to account for cumulative heating and diffusion effects. The model successfully shows that heat propagation and accumulation accurately predict the radius of the ‘heat modified’ zones observed in borosilicate glass waveguides formed across a wide range of laser exposure conditions. Such modelling promises better control of thermal effects for optimizing the fabrication and performance of three-dimensional optical devices in transparent materials.

1. Introduction
Ultrafast laser machining of photonic devices has attracted much attention recently because of its unique capability of fabricating three dimensional (3-D) optical circuits inside transparent materials. Various components have been demonstrated including couplers [1, 2], ring resonators [3], and active waveguides [4, 5]. Waveguides are typically defined by locally modifying the refractive index of the substrate through nonlinear optical absorption in a small focal volume. In early work, amplified Ti:Sapphire systems were used because of their commercial availability and high per pulse energy. But the waveguide writing speed was generally slow (~100 μm/s) due to their low repetition rate (~1 kHz). Further, the waveguide modes were often asymmetric due to transverse writing with an asymmetric focused laser beam. On the other hand, high repetition rate (>200 kHz) ultrafast laser machining introduces heat accumulation during laser processing, which minimizes thermal cycling between pulses and enables crack-free laser micromachining [6,7]. A practical benefit of the higher repetition rate is higher writing speeds, for example, of waveguides (~10 mm/s)[8].

One manifestation of heat accumulation is the formation of a large-diameter modification zone around buried optical waveguides. This occurs when thermal diffusion cannot remove the absorbed laser energy before the arrival of the next laser pulse [6,8]. The radius of the heat modified zone is controllable, for example, increasing with an increase in either repetition rate or the incident laser fluence. Our motivation is further evidence that large cladding radius also correlates with the formation of low-loss and cylindrically symmetric optical waveguides, an important requirement in fabricating 2-D and 3-D optical circuits.
To better understand the cumulative heating and diffusion effects, we report here a numerical thermal model based on the finite difference (FD) method [9]. Known physical properties of borosilicate glass (AF45) were combined with measured values of ultrafast laser absorption to model the 3-D spatial and temporal evolution of temperature within the laser-focal volume and surrounding heat affected zone. The thermal model provides a minimum radius in which the glass is held above the annealing point over many laser pulses. This radius is shown to predict the measured diameter of the heat-modified waveguides formed under various laser exposure conditions. By understanding such accumulative heating effects, one can better control this new domain of high repetition rate ultrafast laser material interactions for a broad range of surface and 3-D bulk micromachining applications.

2. Heat accumulation modelling

Figure 1 shows an optical microscope image of a waveguide written in borosilicate glass (Schott, AF45) with 400-fs pulses and 1-MHz repetition rate from a high repetition rate, fiber laser (IMRA America, µJewel). The writing laser was incident normal to the image plane and scanned parallel to the surface. Further details of laser exposure conditions are given in [8]. The center core waveguide in figure 1 is 5-μm diameter, defined approximately by the laser focal volume. Surrounding this core is a much larger cladding of r = 10 μm radius caused by thermal effects.

Analytical thermal diffusion equations have been previously applied in [6] to model the heat flow inside the bulk glass during high repetition rate waveguide writing. Although the model fits well with experimental data at pulse numbers less than 10^3, it greatly deviates from the observations at pulse numbers greater than 10^4. Further, it did not provide detailed information about the temperature evolution and heat accumulation in between individual pulses. Here we present a FD thermal diffusion model [9] that provides 3-D temperature profiles in borosilicate glass with heating defined empirically by the laser conditions during waveguide writing. With this FD model, the expansion of waveguide cladding due to heat accumulation as well as the temperature fluctuation near the laser focal volume due to heat diffusion were revealed with user defined time interval. The 1-MHz laser pulses were treated as periodic heat sources appearing as delta functions in time since the timescale for electron heating and electron-phonon coupling (<1 ps) is much shorter than the thermal diffusion time (>0.1 μs) under consideration. The laser energy dissipation was treated approximated as a spherical Gaussian of the form:

\[ E(r) = E_0 \exp \left( -\frac{r^2}{w_0^2} \right) \]

where \( r \) is the radial distance and \( w_0 \) is the 1/e radius at the focused laser beam waist. The normalization energy density, \( E_0 \), was defined by matching the volume integral of equation 1 to the measured absorbed energy per pulse. Given this spherical heating source, only the radial component of the 3-D heat diffusion equation was numerically solved:

\[ \frac{\partial}{\partial r} \left( r^2 \frac{\partial T}{\partial r} \right) = \frac{r^2}{D} \frac{\partial T}{\partial t} \]

where \( T(r,t) \) is the temperature and \( D = 8 \times 10^{-3} \text{ cm}^2/\text{s} \) is the thermal diffusivity of AF45 borosilicate.

The temperature profile was modified each time a new laser heating pulse arrives, by an instantaneous temperature rise, \( \Delta T(r) = E(r)/c_{p}\rho \), where \( c_p = 0.43 \text{ J/g-K} \) and \( \rho = 2.72 \text{ g/cm}^3 \) are the specific heat capacity at constant pressure and the density of the glass, respectively. To simplify calculations, a static exposure was assumed with a maximum number of laser heating pulses, \( N_{\text{dwell}} \), specified by the product of repetition rate and laser dwell time (\( w_0 \) divided by scan speed). This approximation provided temperature profiles representative of the waveguide writing conditions, and validated by observation of nearly identical heat-modification radii formed by static and scanning exposures when applied at the same net fluence. In the model, room-temperature values of specific heat, density and thermal diffusivity were used. To achieve precise results and satisfy the convergence condition [9], the simulation grid was selected to be 200 nm and the iteration time interval to be 10 ns.
for 1-MHz repetition rate. Temperature profiles were calculated within a 120-μm radius from the laser focus.

3. Results and discussion

Figure 2 shows the simulated temperature evolution at position \( r = 3 \) μm from the origin as a function of pulse number for repetition rates of 0.1, 0.5 and 1 MHz. The absorbed laser energy used in the calculation was 201 nJ (70% of 275 nJ incidence), and the beam waist (1/e^2 of intensity) was \( w_0 = 1.0 \) μm. The horizontal dashed line is positioned at the 663°C annealing temperature of AF45 glass, which is used to define the thermal modification threshold. At 100-kHz repetition rate, the temperature at this position always drops below this annealing point prior to the arrival of the next pulse. The volume surrounding the focal volume undergoes significant temperature cycling and minimum heat accumulation. This temperature cycling is less evident at 500 kHz and 1 MHz. The temperature also builds up to much higher values due to shorter time intervals between successive pulses. This heat accumulation effect leads to thermal modification volumes much larger than the laser focal volume. This high temperature ‘annealing’ zone grows in size with increasing laser dwell time (slower scan velocity), and is most evident at highest repetition rate.

Following a similar approach reported in references [6] and [8], we calculated the maximum radius out to where an annealing temperature of 663°C was maintained during the laser dwell time. Results are plotted (solid lines) in figure 3 for three laser pulse energies as a function of incident net fluence, defined here by the product of maximum pulse number, \( N_{dwell} \), and single-pulse fluence. The laser repetition rate was 1 MHz, corresponding to the strong heat accumulation condition. For comparison, figure 3 also plots (data symbols) the maximum radius observed in heat-modified waveguides with an optical microscope (i.e. the large radius cladding seen in figure 1). The incident laser pulse energies of 200, 275 and 350 nJ were reduced to 70%, 73%, and 75%, respectively, corresponding to the glass absorption values measured by placing a thermopile power detector below the glass sample. These corrected absorbed energies were also used in the FD simulation (solid lines in figure 3).

There is very good agreement between the simulated and experimental data in figure 3 over the range of scan velocities (2 to 60 mm/s) and pulse energy (200 to 350 nJ) examined. Small discrepancies can be accounted for by light scattering losses from the laser-induced plasma and unaccounted variations in our model for the specific heat, density and thermal diffusivity of the glass which can change significantly at high temperature. Temperature dependent corrections are not available for AF45 borosilicate glass properties. Nevertheless, the close match of the FD modeling
and the experimental observations (figure 3) is strongly supporting evidence for the heat-accumulation effects and thermal nature underlying the formation of cladding zones in high-repetition-rate ultrashort-pulse laser writing of waveguides.

Figure 4 shows the simulated and measured annealing diameters for borosilicate waveguides written at repetition rates of 0.5, 1.0, and 2 MHz. For all the simulations and fabricated waveguides, the average laser power was fixed at 275 mW, with the absorbed pulse energies corrected by measured absorptions of 78%, 73%, and 47% for 0.5, 1, and 2 MHz, respectively. The low absorption efficiency for the 2 MHz exposures is expected as lower pulse energy and laser intensity reduces the nonlinear absorption. Note the significant variation and control over waveguide radius with only small changes in laser scan rate (net fluence) and repetition rate. In all cases, the laser spot size was $w_o = 1.0 \mu m$.

Theoretical predictions of the annealing radius agree well with the measured cladding radius for high repetition rates (1 MHz and 2 MHz), but deviate at low repetition rate (500 kHz). Here, an approximately 30% to 60% larger heating zone is observed, possibly owing to temperature-dependent changes in the material properties that are unaccounted in the model. Such effects will be more pronounced at lower repetition rate due the longer interval between laser pulses. Efforts are underway to improve the FD model for a wider range of laser exposure conditions and transparent materials. Also, further effort is required to connect the heat-modified zones with the laser-induced refractive index profiles that define the waveguide. Such modeling will help better understand the photosensitivity mechanisms underlying refractive index changes, improve control over mode sizes, and provide additional insights to improving waveguide losses (currently at ~0.2 to 1.0 dB/cm).

4. Conclusion
A finite-difference simulation was successfully applied to model thermal processes underlying high repetition rate, femtosecond-laser waveguide writing. The theoretical results agreed well with experimental observations of heat-modified cladding zones for various laser pulse energies, scanning speeds, and repetition rates. The model could be further improved by introducing temperature-dependant material properties and extending the model to a scanning heat source.

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