**Simulation of the dynamics of heat accumulation during in-volume modification with ultrashort laser pulses**

Sebastian Nippgen\(^a\)*, Arnold Gillner\(^a,b\), Andreas Smolenko\(^c\)

\(^a\)RWTH Aachen University - Chair for Laser Technology LLT, Steinbachstraße 15, 52074 Aachen, Germany,

\(^b\)Fraunhofer-Institute for Lasertechnology ILT, Steinbachstraße 15, 52074 Aachen, Germany

\(^c\)RWTH Aachen University – MATHCCES Department of Mathematics, Schinkelstr. 2, 52062 Aachen, Germany

* Corresponding author. Tel.: +49-8906-470; fax: +49-8906-121. E-mail address: Sebastian.Nippgen@llt.rwth-aachen.de

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**Abstract**

Working with ultrashort pulsed (USP) laser systems with high repetition rates \( \geq 100 \) kHz, heat accumulation effects evolve and subsequently the absorption becomes linear. As a consequence, the process dynamics change significantly. Structuring glasses with those USP Lasers leads to melted lines that are used for waveguides, welding or selective laser-induced etching whose resulting qualities are strongly depending of the structuring results and given smoothness. We perform simulations during in-volume modification processes, by recalculating the absorption behavior in between the iteration steps of heat accumulation. The simulation results provide explanations for smoothness or non-smoothness of modifications comparable to experiments in glass.

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1. **Introduction**

High peak power of \( P_{\text{peak}} \geq 10^{13} \) W/cm\(^2\) enables the nonlinear absorption of laser radiation in dielectrics which are normally transparent to the used wavelength. Therefore, ultrashort pulsed (USP) laser systems are a key technology for generation of in-volume modifications, which can be used for marking of glasses, producing waveguides, fusion welding or even 3D-printing (Selective Laser induced Etching).

The used pulse repetition rates for manufacturing of in-volume modifications in the region of \( f \geq 100 \) kHz leads to heat accumulation. Subsequently, the absorption becomes linear and the process dynamics change significantly.

For simulating the material response to the used parameters of repetition rate \( f \), pulse duration \( \tau_p \), feeding speed \( v \) and pulse energy \( E_p \), a thermal conduction model given in [1] which assumes a fixed heat delivering source along the propagation axis \( z \) is adapted.

We are using two models for the simulation of the heat accumulation during the in-volume modification of glass. One model is used to calculate the absorption and therefore the coupled power density for each pulse during the simulation of irradiation. The other model is used for an iterative calculation of the coupling power density after reaching the static state of temperature distribution in each iteration step.

2. **Setup of simulation and experiments**

The simulation in this paper describes the heat accumulation within the moving coordinate system \( CS \) (Fig. 1a) of a tight focused laser beam localized at \( x, y, z = 0 \) with a constant feeding speed \( v \). \( CS' \) is the fixed coordinate system in relation to the described dielectric sample.
The field of temperature for a pulsed laser beam at a time \( t \) is be described by

\[
T(\vec{x}, t) - T_0 = \int_{t_0}^{t} \int_{\mathbb{R}^3} P_0(\vec{x}, t') G(\vec{x} - \vec{v}t - \vec{x}', t - t') d\vec{x}dt',
\]

where \( T_0 \) denotes the temperature of the sample before the irradiation process, \( P_0 \) the heat source of the pulsed laser beam and \( G \) the Green function of the heat conduction equation [2].

The heat source for a number of \( N \) pulses is given by [1]

\[
P_0(\vec{x}, t) = \sum_{i=1}^{N-1} \delta \left( t - \frac{2i}{f} \right) \frac{2\omega(x)}{\
\pi w(z)^2 f} e^{-\frac{r^2}{w(z)^2}}; 0 \leq z \leq l
\]

with \( r^2 = x^2 + y^2 \), \( w \) the Gaussian beam radius, \( \delta \) the delta distribution, \( l \) the length of region where the laser pulse will be absorbed and \( \omega(z) \) the power density.

The coupled power density \( \omega_{\gamma}(z) \) of an incoming laser pulse is calculated by

\[
\omega_{\gamma}(z) = P_0 \alpha(z) e^{-\frac{1}{2} a z'} dt'
\]

with \( P_0 \) the power of the laser pulse. As a good approximation the coupled power density is given by

\[
\omega(z) = az^m + b,
\]

where the parameters \( a, m \) and \( b \) are estimated from fitting to the heat effect zone of a cross section from the modification with a static temperature distribution model as described in [1]. This can be used to calculate the accumulated heat to a state of saturation (Fig.1b) in a first step of iteration.

The absorption coefficient \( \alpha \) of a laser pulse during the heat accumulation depends on the density \( n_e \) of free electrons in the conduction band. For a molecular single bonded dielectric the absorption coefficient is approximated in [3] and given by the following formula:

\[
\alpha(z) = \frac{e^2 \tau}{m_e \epsilon_0 c_z n} \left[ \frac{1}{1 + \left( \omega_{\gamma} \right)^2} \frac{\int_{-\lambda_0 T}^{\lambda_0 T} I(z,t) n_e(z,t) dt}{\int_{-\lambda_0 T}^{\lambda_0 T} I(z,t) dt} \right]
\]

with \( \tau \) the time between electron-heavy particle collisions, \( m_e \) the electron mass, \( \epsilon_0 \) the dielectric constant, \( c_z \) the speed of light in vacuum, \( n \) the refractive index of the sample and \( \omega_{\gamma} \) the angular laser frequency.

The free electron density is calculated during the absorption of an incoming laser pulse and during a state without absorbing a laser pulse, when the temperature \( T \) is mainly given by the free electrons density \( n_e \).

### 2.1. Electron density without laser pulse absorption

For \( f \tau_p \ll 1 \) the free electron density at temperature \( T \) can be described by the Saha-equation [4]

\[
n_e = \frac{1}{\lambda_T^4} \left( \frac{1 + 2 n_M \lambda_T^2 e^{\frac{E_g}{k_B T}} - 1}{e^{\frac{E_g}{k_B T}} - 1} \right)^2
\]

with \( \lambda_T \) the thermal de-Broglie wavelength, \( n_M \) the molecular density, \( E_g \) the band gap of the sample and \( k_B \) the Boltzmann constant.

### 2.2. Free electron rate equation during the absorption of a laser pulse

During the absorption of the laser pulse, the free electron density changes are a function of time given by the single rate equation [3]

\[
\partial_t n_e = \left( \frac{\partial n_e}{\partial t} \right)_{mp} + \eta_{casc} \left( \frac{\partial n_e}{\partial t} \right)_{casc}
\]

with contributions by multiphoton absorption (mp) and cascade ionization (case) with \( k \) the number of photons needed for raising an electron from valence to the conduction band and \( \eta_{casc} \) the cascade ionization rate. Because the Keldysh parameter of the used material in the simulation is \( \gamma_k \geq 0.52 \) the tunneling ionization will be neglected [6]. Since the temporal pulse shape of the simulated laser source is Gaussian and the electron density between two pulses decreases by a maximum of 20%, recombination and diffusion of the electrons are also neglected.

### 3. Models

#### 3.1. Pulse-to-pulse model

The simulation starts with the glass at room temperature \( T_0 \). The integer \( i = 1 \ldots N \) is used as a counter for the number of incoming laser pulses. The coupled power density distribution \( \omega_{\gamma}(z) \) is calculated for the temperature distribution in the moment before a subsequent laser pulse is absorbed by using the formula (2) for calculation of the free electron density. With the onset of the laser pulse absorption, the temporal evolution of the free electron density \( n_e \) is calculated by the rate equation (3).
3.2. Iterative model

In the iterative model the coupled power density distribution $\omega_t(z)$ is fixed for each iteration step counted by $s$. We choose equation (1) for the start of the simulation after fitting the parameters to experimental results.

The iterations are calculated until the temperature distribution in the glass due to heat accumulation will reach a static state (Fig.1b). With the given static temperature distribution, the free electron density $n_e$ is calculated by (2) and the temporal evolution of $n_e$ is calculated for the absorption of one pulse with (3). Then, $\alpha(z)$ and $\omega_t(z)$ are calculated for the given electron density $n_e$ and the heat accumulation simulation will start again until the temperature distribution converges or none converge can be seen.

4. Results

To sets of parameters we choose for the simulations are:

- S1 500 kHz, 20 mm/s and 1 µJ
- S2 1250 kHz, 50 mm/s and 1.2 µJ

for processing Borofloat®33. Moreover, we perform processing experiments for the same set of parameters to compare the simulation results to experimental modifications.

4.1. Pulse-to-pulse model

Due to high computational effort, the pulse-to-pulse simulation is limited to a maximum of 4000 laser pulses. The temperature distributions of both simulations tend to saturate up to 4000 laser pulses (Fig.2).

The temperature distribution of parameter set S1 exhibits a maximum located 30 µm above the focal spot at saturation. The distribution of parameter set S2 reveals two maxima located at 25 and 55 µm above the focal spot, respectively. Moreover, the maximum temperature is approximately 1000° higher compared to the parameter set S1. For both temperature distributions, the highest temperature is reached above the focus position.

4.2. Iterative model

Each heat accumulation simulation of an iteration step is performed with 8000 laser pulses using the described iterative model. The iteration 0 is calculated by the coupled density distribution (1) with the fitting method of [1].

4.3. Experimental results

In order to get a first approximation for the coupled power density (1) and a comparison between experimental results and simulation, Borofloat®33 has been processed with the two parameter sets S1 and S2 using an ultrashort pulsed laser system “Satsuma” with a central wavelength of $\lambda = 1030$ nm, beam quality $M^2 = 1.2$ and a pulse duration $\tau = 750$ fs. The laser radiation is focused with a microscopy objective with a numerical aperture of $NA = 0.4$. The spherical aberrations are compensated by the used objective.

The structuring of Borofloat®33 with parameter set S1 reveals a smooth structure, while the structuring with parameter set S2 shows a periodical modification (Fig.4).

Fig. 3. Simulation of the heat distribution along the propagation axis for the iterative model. (a) 500 kHz, 20 mm/s, 1 µJ (b) 1250 kHz, 50 mm/s, 1.2 µJ

After 25 iterations the simulated temperature distribution of parameter set S1 converges (Fig.3). The simulation for parameter set S2 has been stopped after 5 iterations because converge will not take place. The position of the maximum temperature for parameter set S1 is moving into the direction of laser beam source and is localized at approximately 30 µm above the focal spot.

The position of the maximum temperature of the simulated temperature distribution for the parameter set S2 is located towards the direction of the laser beam source and the maximum temperature decreases. At the third iteration a new temperature maximum evolves in the vicinity of the focal spot. During the following iterations, the temperature maximum above the focus position decreases and the maximum temperature at the focus position increases while it moves towards the laser beam source.

Fig. 4. Transmission light microscopy of structured Borofloat®33 in top view. (a) 500 kHz, 20 mm/s, 1 µJ. (b) 1250 kHz, 50 mm/s, 1.2 µJ.
5. Discussion

5.1. Pulse-to-pulse Model

The resulting temperature distributions of the pulse-to-pulse simulations reveal a difference between the parameter set $S1$ and the parameter set $S2$, which appears smooth $S1$ and disturbed $S2$ in the experiments. The simulation does not predict disturbed or periodic structures behavior as observed in the experiment (Fig.4). The simulation predicts the existence of two separated maxima of the temperature distribution, which are suggested to be two separated locations with significant higher absorption for subsequent laser pulses.

5.2. Iterative Model

With the simulation results of the iterative model a main reason for the difference between the resulting smoothness of structuring with parameter set $S1$ and the resulting periodic modifications $S2$ can be clarified. With beginning of iteration step 1 for parameter set $S2$ the following processes are observed (Fig.3):

- **Iteration 1**
  The main process for absorption in this step is cascade ionization. Temperature diffusion and absorption of laser radiation in the region above the focus leads to a movement of the maximum of temperature distribution towards the laser beam source. Most of the incoming laser pulse will be absorbed in this temperature maximum. Therefore, there is no heat input in the focus point and it cools down. Thus, there are no excited electrons and finally no absorption.

- **Iteration 2**
  The temperature in the upper region is decreasing due to the lower beam intensity and missing heat input in this region of the absorption volume. Subsequently, the free electron density and thus the absorption in this area are reduced and a larger proportion of laser radiation is transmitted.

- **Iteration 3**
  Since the upper head maximum is decreasing the intensity of transmitted laser pulses is increasing until multiphoton absorption appears at the focal spot. This raises the free electron density $n_e$ in the focus region. The absorption raises and a lower heat maximum appears.

- **Iteration 4**
  Cascade ionization and heat diffusion enlarges the absorption region at the focus leading to an increase of the local temperature. The upper temperature maximum is decreasing as described in iteration 2.

- **Iteration 5**
  The upper temperature maximum disappears in further iteration steps due to a missing heat input and the decrease of the free electron density $n_e$. The temperature in the lower temperature maximum therefore increases and cascade ionization becomes the main process. The lower heat maximum moves towards the beam source and absorbs most of the subsequent laser pulses. Therefore, the temperature in the region of the focal spot cools down until the process restarts with iteration 1.

The simulation results of the parameter set $S1$ reveals a similar behavior. Here, the temperature maximum moves towards the laser beam source as described for the simulation with parameter set $S2$ as well. The main difference is a separation in the temperature distribution between the temperature at the focal spot and the upper temperature maximum. The heat input in the upper temperature maximum is increased by a high free electrons density but the number of free electrons does not exceed a critical value. Therefore, subsequent pulses are partially transmitted.

6. Conclusion

The presented simulations are capable to reproduce qualitatively observed structural differences between smooth and disturbed modifications for processing glass with different sets of parameters. For a smooth modification a temperature distribution with one heat maximum that is linked to the focus themes to be necessary. The disturbances of the modifications occur as soon as a temperature maximum strives against the laser beam source and the resulting absorption weakens the intensity of the incoming laser pulses so tightly that the absorption in the focus area does not occur. In this case, the temperature maximum can no longer be supplied with heat, so that the temperature and the absorption become smaller. This leads to a separation from the upper to the lower temperature maximum, or to the focal spot. For an improvement of the simulations, the linear approximation of coupled power density $\omega_I(z)$ will be changed to a Gaussian distribution in our upcoming studies. Moreover, time-resolved experimental studies with a pump probe setup are planned for a further investigation of thresholds for the transitions from smooth to disturbed modifications and to get more insights into the transient local temperature distribution during the modification processing.

References