

Predictive coupled plasma and thermal modelling of femtosecond laser-glass interaction

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Abstract: Femtosecond laser processing enables high-precision micromachining of transparent materials through nonlinear absorption and localized energy deposition. However, predictive modelling of plasma generation and thermal accumulation in dielectric materials remains limited. In this work, a physics-based simulation framework is presented for femtosecond laser interaction with glass. Plasma-dependent Fresnel reflectance is incorporated to investigate plasma shielding during pulse propagation. The model is used to predict single-pulse ablation crater morphology. Experimental validation demonstrates agreement between simulated and measured crater depth and radius trends. This work provides a predictive framework for understanding ultrafast laser energy deposition and optimizing femtosecond laser processing of dielectric materials.

1. Introduction

Femtosecond laser processing has emerged as a powerful manufacturing technology for precision micromachining of transparent materials owing to its ability to deposit laser energy through nonlinear absorption with high spatial and temporal precision. Compared with conventional mechanical fabrication, femtosecond laser processing is a non-contact technique that is inherently not affected by tool wear while enabling flexible control of the interaction volume through the ability to focus inside the bulk. Because the laser pulse duration is much shorter than the thermal diffusion time, energy is deposited before significant amount of heat is dissipated into the bulk, resulting in reduced heat-affected zones and minimal collateral damage. These advantages have enabled a broad range of applications, including three-dimensional waveguide inscription for integrated photonics, laser welding of transparent materials, deterministic optical figuring and polishing, fabrication of freeform optical components, and microfluidic device manufacturing [1–8]. More recently, gigahertz burst-mode irradiation has demonstrated the potential to enhance material removal efficiency and improved surface quality by exploiting the incubation of plasma and temperature during gigahertz frequency bursts of pulses [9–12].

Despite the inherent benefits, the optimization of femtosecond laser processing remains time-consuming as it continues to rely heavily on experimental investigation. Ultrafast laser interaction with dielectric materials involves multiple strongly coupled physical processes spanning timescales from femtoseconds to microseconds. During irradiation, strong-field photoionization generates free electrons that accumulate to form a dense plasma. As the electron density increases, the optical properties of the material gradually evolve, affecting laser propagation, absorption, and reflection. Plasma formation modifies laser-energy coupling and may contribute to non-thermal material removal, while electron-lattice energy transfer governs the resulting thermal response, including softening and melting. These processes occur simultaneously and interact across multiple temporal and spatial scales, making predictive modeling valuable for understanding laser-matter interaction and optimizing processing conditions.

The theoretical description of ultrafast laser interaction with transparent materials has evolved substantially over the past several decades. A foundational description of strong-field photoionization was established by Keldysh, whose theory provides a unified treatment of multiphoton and tunneling ionization under intense laser irradiation [13]. Keldysh-based photoionization rates were subsequently incorporated into rate-equation models describing the evolution of the total free-electron density through photoionization, impact ionization, and electron recombination. The multiple-rate-equation (MRE) model was later developed to track the nonstationary energy distribution of electrons across discrete conduction-band energy levels, thereby describing laser-driven electron heating and the delayed transition to the avalanche-ionization regime [14]. These models provide a basis for predicting plasma generation in dielectric materials during femtosecond laser irradiation and have been widely applied to studies of laser-induced optical breakdown and material modification.

As the free-electron density increases, the generated plasma significantly influences laser-energy deposition by modifying the optical properties of the irradiated material. This behavior is commonly described using the Drude model [15,16], which relates the plasma density to the evolution of the complex dielectric permittivity and refractive index. The resulting changes in absorption and reflectance reshape the propagating laser pulse and can produce plasma shielding, which confines energy deposition near the surface and limits penetration into the material at high excitation levels. Coupling plasma-dependent optical properties and transient Fresnel reflectance to the laser-propagation model is therefore important for accurately predicting the spatial distribution of absorbed energy and the resulting material-removal morphology. Several studies combine these concepts to predict material removal; however, their approach remains limited to electron density prediction and does not incorporate the energy transfer to the lattice system while only demonstrating the crater morphology for a limited range of peak-laser fluences [16,17].

Following laser-energy deposition, energy transfer from the electrons to the lattice can be described using the two-temperature model (TTM) [18]. Comprehensive treatments [19] have summarized the fundamental mechanisms governing ultrafast energy deposition, plasma formation, heat transfer, and ablation in transparent materials. Jiang and Tsai [20] combined a plasma model with an improved two-temperature model to investigate single-pulse femtosecond ablation of fused silica. Their framework accounted for the time- and space-dependent optical properties of the generated plasma and was used to predict the ablation threshold, depth, and crater shape. However, their study did not provide a systematic direct validation of both crater depth and radius over a broad range of laser fluences.

While these studies have significantly advanced the understanding of individual physical mechanisms, predictive modeling of femtosecond laser processing remains challenging because laser propagation, plasma evolution, transient optical properties, and thermal transport are strongly coupled.

In this work, a predictive simulation framework is developed by coupling a multiple-rate-equation model with the two-temperature model to describe plasma generation and thermal accumulation during femtosecond laser interaction with glass. Plasma-dependent optical properties are calculated using the Drude model, and time-dependent Fresnel reflectance is coupled to the laser-propagation model to account for the continuous evolution of plasma shielding during irradiation. Building upon previous coupled plasma and two-temperature models, the present work incorporates MRE-based electron dynamics, continuously couples the transient optical response to laser-energy deposition, quantitatively evaluates predicted crater depths and radii against experimental measurements. The model is applied to single-pulse laser ablation, for which the predicted crater morphology (depth and radius) is validated over a range of laser fluences. By integrating the principal physical processes governing ultrafast laser-matter interaction and evaluating the resulting material-removal predictions

experimentally, this work provides a quantitative link between the underlying interaction physics and the resulting crater morphology.

2. Theoretical modelling

2.1 Equation framework

Nonlinear absorption at high peak intensities is governed by several mechanisms. Multiphoton ionization (MPI) occurs when two or more photons are simultaneously absorbed to excite an electron across the bandgap. MPI rate follows the Keldysh theory of photoionization [13]. At sufficiently high electron densities, these seed photoionized electrons can initiate avalanche ionization, where energetic electrons collide with bound electrons and promote them to the conduction band, resulting in exponential growth of the electron density. Recombination processes and collision losses act to reduce the electron density. The multiple rate equation (MRE) summarizes the rate of change of electron density $[\rho(r, z, t)]$ described by these mechanisms [4,14]:

$$\frac{\partial \rho(r, z, t)}{\partial t} = \eta_{photo} I^K(r, z, t) + \eta_{aval} I(r, z, t) \rho(r, z, t) - \eta_{rec} \rho^2(r, z, t) \quad (1)$$

Where η_{photo} , η_{aval} and η_{rec} are the rates of photoionization, avalanche ionization and recombination, respectively. K is the number of photons required for ionization [14]. The critical electron density serves as the criterion for material removal in this work. It is defined as the electron density at which the plasma frequency equals the incident laser frequency, marking the transition to a strongly plasma-dominated optical response associated with optical breakdown and material ablation:

$$\rho_c = \frac{\epsilon_0 m_e \omega^2}{e^2} \quad (2)$$

Where ϵ_0 is the permittivity of free space, m_e is the mass of an electron, ω is the incident laser frequency, and e is the charge of an electron. The simulation framework is built on the timescale of the femtosecond pulse propagating through the substrate. The pulse propagation and nonlinear energy absorption follows the principles laid out in predictive simulation for micro-bonding [4]. The intensity of a propagating laser pulse is modelled in this manner:

$$I(r, z, t) = \frac{2E_p}{\pi w_z^2 \tau_p} \times \sqrt{\frac{4 \ln 2}{\pi}} \times [1 - R(r, z, t)] \times \exp\left(-\frac{2r^2}{w_z^2}\right) \times \exp\left[-4 \ln 2 \left(\frac{t - 2\tau_p - n_0 z/c}{\tau_p}\right)^2\right] \times \exp\left[-\int_0^z \alpha(r, z', t) dz'\right] \quad (3)$$

Where E_p is the pulse energy, w_z is the Gaussian beam waist radius, τ_p is the full-width-half-max (FWHM) pulse duration (set to 210 fs), R is the reflectance, n_0 is the refractive index, c is the speed of light, and α is the absorption coefficient. The first two exponential terms are the Gaussian distribution of the pulse in space and time, respectively. The term $n_0 z/c$ is the time offset due to the speed of light to account for the translation of the pulse in space. The last exponential term is the decay of pulse intensity due to substrate absorption. The peak of the pulse arrives at the air-glass interface ($z = 0$) at time $t = 2\tau_p$. Laser-induced plasma generation modifies the material permittivity, and hence its reflectance, which can be described using the Drude model [15].

$$\epsilon^*(r, z, t) = \epsilon_r + i\epsilon_i = \epsilon - \frac{\rho(r, z, t)e^2}{\epsilon_0 m_e \omega} \cdot \frac{\omega \tau_D}{\omega \tau_D + 1i} \quad (4)$$

Where ϵ^* indicates the complex material permittivity upon excitation, ϵ is the unexcited material permittivity, and τ_D is the characteristic electron collision time. The complex refractive index can be described as:

$$n(r, z, t) = \sqrt{\epsilon^*(r, z, t)} = n_r(r, z, t) + in_i(r, z, t) \quad (5)$$

Where n_r and n_i are the real and imaginary terms of the refractive index, respectively. The Fresnel reflectance term follows as:

$$R(r, z, t) = \frac{[n_r(r, z, t) - n_0(r, z, t)]^2 + n_i^2(r, z, t)}{[n_r(r, z, t) + n_0(r, z, t)]^2 + n_i^2(r, z, t)} \quad (6)$$

The rate of energy absorption is described using the imaginary term of the complex refractive index and the incident laser intensity in this manner [16]:

$$\alpha(r, z, t) = \alpha_0 \left[\frac{2\omega n_i}{c} + \frac{E_g \eta_{aval} \rho(r, z, t)}{I(r, z, t)} \right] \quad (7)$$

Where α_0 is a linear scaling term to account for experimental mismatches in energy absorption and E_g is the bandgap energy. The first term describes the linear scaling of absorption depth with respect to the imaginary refractive index and the second is the nonlinear scaling of this absorption due to avalanche ionization. The transfer of energy from the electron to the lattice system is realized in the form of temperature change. The two-temperature model is a system of coupled electron and lattice temperature equations where the gain in electron temperature is fed by the laser absorption, and the loss is the energy transfer to the lattice. The lattice system has the same transfer term while both equations contain the classical heat conduction term described by the Laplacian of the temperature distribution. The two-temperature model is summarized in the following form [20,21]:

$$C_e \frac{\partial T_e(r, z, t)}{\partial t} = \nabla \cdot [k_e \nabla T_e(r, z, t)] - G[T_e(r, z, t) - T_l(r, z, t)] + S(r, z, t) \quad (8)$$

$$C_l \frac{\partial T_l(r, z, t)}{\partial t} = \nabla \cdot [k_l \nabla T_l(r, z, t)] + G[T_e(r, z, t) - T_l(r, z, t)] \quad (9)$$

where subscripts e and l refer to the electron and lattice systems, respectively, T is the temperature, k is the thermal conductivity, and C is the specific heat capacity. G is the transfer term between the electron and lattice systems and S is the source term to the temperature system inherited from the laser energy absorption. The derivatives are numerically solved in a forward-time-central-space (FTCS) finite differencing scheme. The calculation of the source term is only carried out in the timescale of the pulse, after which classical diffusion continues.

2.2 Pulse propagation and energy deposition

Nonlinear absorption is the primary mechanism for generating free electrons during femtosecond laser irradiation of glass, enabling plasma formation, optical breakdown, and subsequent ablation. Figure 1 demonstrates the effect of nonlinear absorption that accelerates the free-electron plasma generation and shields the bulk material beneath the initial surface layer. As fluence approaches the experimentally-determined ablation threshold of 4.3 J/cm² [8]

significant energy attenuation is observed due to induced nonlinear absorption near the surface of the substrate. This is shown with a snapshot of laser fluence distribution in the substrate during irradiation using two different scale range of fluence in Fig. 1. Figures 1(a) and 1(c) show this fluence snapshot during irradiation with 0.2 J/cm^2 peak-fluence pulse, while Figs. 1(b) and 1(d) show this snapshot during irradiation with 4.3 J/cm^2 peak-fluence pulse. For the latter case, as shown in Fig. 1(d) the majority of the pulse energy is absorbed within the first few hundred nanometers, providing insight for the resultant material removal depth of single pulse ablation craters discussed in Section 3. In contrast the lower peak-fluence pulse is seen to have minimal attenuation and continues to propagate through the material unperturbed, demonstrating that plasma shielding due nonlinear absorption for this case is minimal.

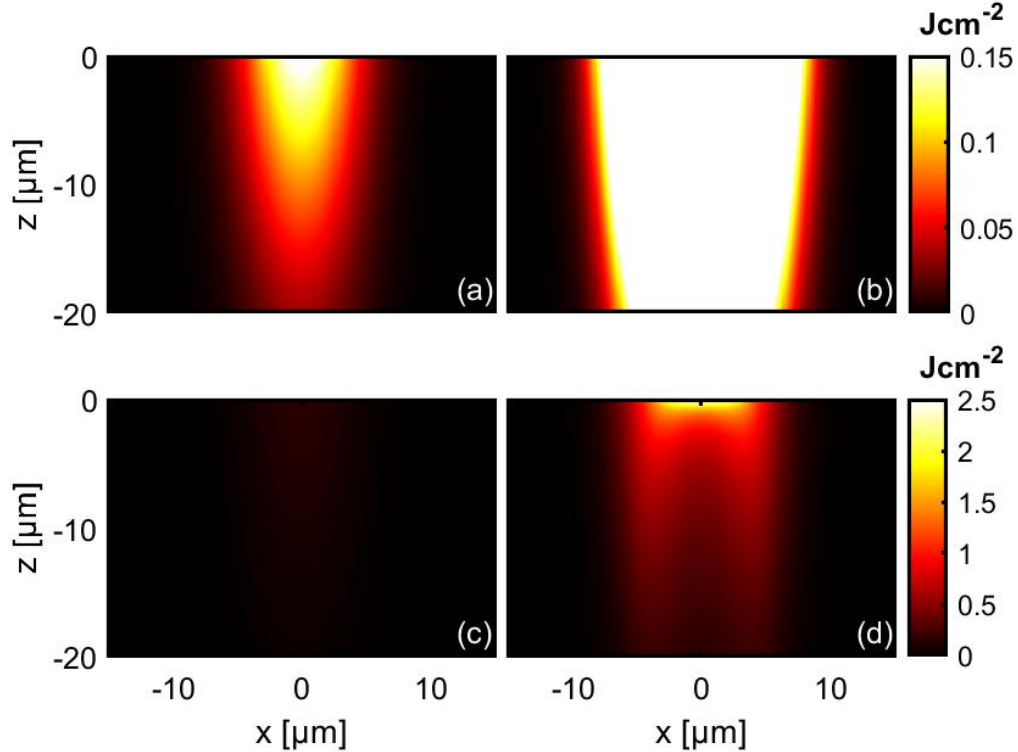


Figure 1. Snapshot of laser pulse fluence with incident peak fluence of (a) and (c) 0.2 J/cm^2 and (b) and (d) 4.3 J/cm^2 , demonstrating the effect of nonlinear absorption with increasing pulse energy.

The reflectance change with increasing electron density is crucial in controlling the magnitude of energy that radiates through the substrate. The Drude model and Fresnel equations (Eq. 4-6) are used to simulate the reflectance as the pulse propagates through the glass. Nonlinear absorption is determined with Eq. 7 and the MRE is subsequently solved to predict the electron density. Figure 2(a) shows the peak reflectance; Fig. 2(b) shows the peak absorption, and Fig. 2(c) shows the resultant peak electron density accumulation during the pulse for selected peak fluence.

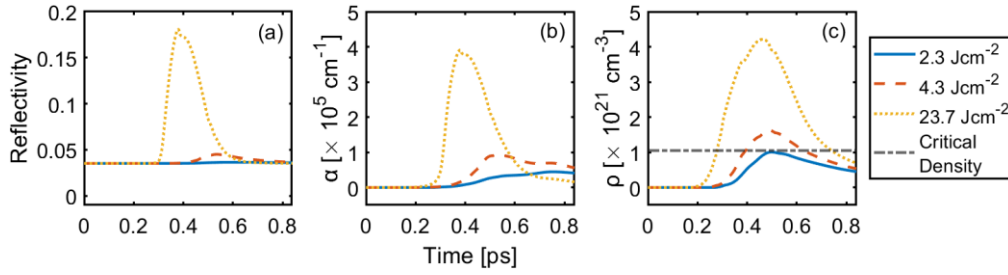


Figure 2. Peak (a) reflectance (b) absorption and (c) electron density during the propagation of the pulse for selected peak laser fluences.

The onset of reflectance change and increase in absorption follows the electron density exceeding the critical density threshold. Reflectance and absorption represent the two key mechanisms of plasma shielding that affects material removal efficiency. A finite delay exists between plasma generation and the onset of material removal, typically on the order of picoseconds [15]. Therefore, the shielding effect is present for the entire duration of a femtosecond pulse for fluences higher than the ablation threshold. Maximizing the material removal efficiency is therefore defined by a balance between energy lost due to reflection and absorbed due to an increase in absorption. As seen in Fig. 2, both mechanisms are accelerated in the time when the electron density has exceeded the critical density threshold.

3. Single-pulse ablation

3.1 Experimental setup

Single-pulse ablation experiments are carried out to compare crater morphology predictions using the simulation model. Figure 3 contains the system-level diagram of the femtosecond laser processing system. The source is a solid-state laser (Carbide, Light Conversion) with a tunable pulse-width (210 fs to 2 ps) at a central wavelength of 1030 nm. A motorized polarization-based attenuator (LS-Shape, LASEA) controls the pulse energy delivered onto the sample. The beam is directed onto the sample using a set of mirrors and focused using an antireflection coated aspheric lens. A three-axis linear stage system (Lxc 80F40, Jenny Science) is used to precisely position the sample under the focusing lens and a confocal imaging system is used to optimize the focusing on the top surface of the sample.

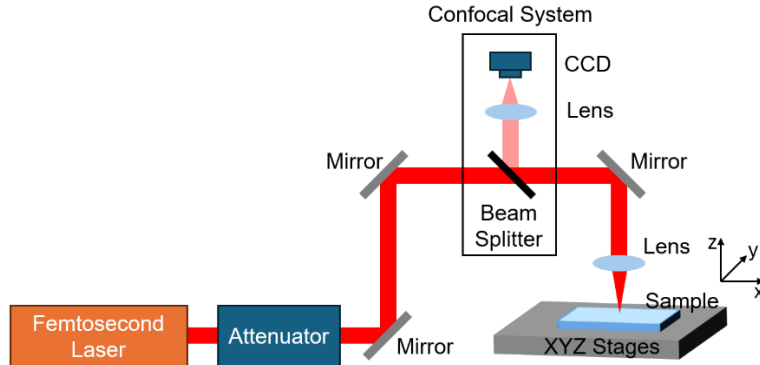


Figure 3. Layout of the femtosecond laser processing setup.

3.2 Single-pulse craters

The single-pulse regime provides the most direct comparison between experiment and simulation because cumulative multi-pulse effects such as incubation, heat accumulation, and pulse-to-pulse surface evolution are minimized. Single-pulse ablation experiments were

performed to validate the model-predicted material-removal crater morphology. The measured crater depth and radius were compared with the simulated material removal profile as a function of incident peak-fluence in Fig. 4(a) and Fig. 4(b), respectively. As shown in Fig. 4(a), below the ablation threshold, both experiment and simulation show minimal crater depth, indicating that the generated free-electron density and deposited energy remain insufficient to produce material breakdown and only soften and melt the material. Above the threshold, the crater depth increases with fluence as nonlinear absorption localizes energy deposition near the surface. As shown in Fig. 4(b), the depth trend begins to saturate with increasing peak-fluence, which is attributed to plasma shielding which limits further energy penetration deeper into the material. The offset between the measured and predicted radii is attributed to physical processes that are not explicitly included in the model, such as melt redistribution, redeposition, and stochastic material ejection. The agreement in crater-depth trends suggests that the critical-density criterion captures the dominant removal behavior under the single-pulse conditions studied here, while deviations in crater radius are expected because these edge-related processes are not explicitly modeled. These processes predominantly affect the outer edges of the crater and therefore its result is observed in the crater radius measurement. Overall, the comparison demonstrates that the model captures the dominant single-pulse ablation behavior and provides a basis for analyzing multi-pulse and burst-mode regimes.

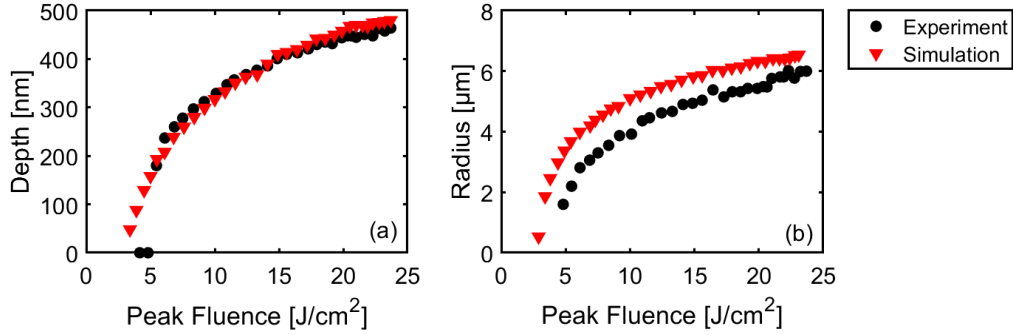


Figure 4. Crater (a) depth and (b) radius as a function of peak incident pulse fluence obtained using experiment and simulation.

Figure 5 shows a scanning electron microscope (SEM) image of a single-pulse modification at $2.3 \text{ J}/\text{cm}^2$ and $23.7 \text{ J}/\text{cm}^2$. As seen in Fig. 2, electron density accumulation at $2.3 \text{ J}/\text{cm}^2$ lies just below the critical density threshold, therefore only thermal modifications are observed. In contrast the region created using $23.7 \text{ J}/\text{cm}^2$ shows a clear material removal crater of depth 465 nm, where evidence of thermal softening is highly localized within the crater region.

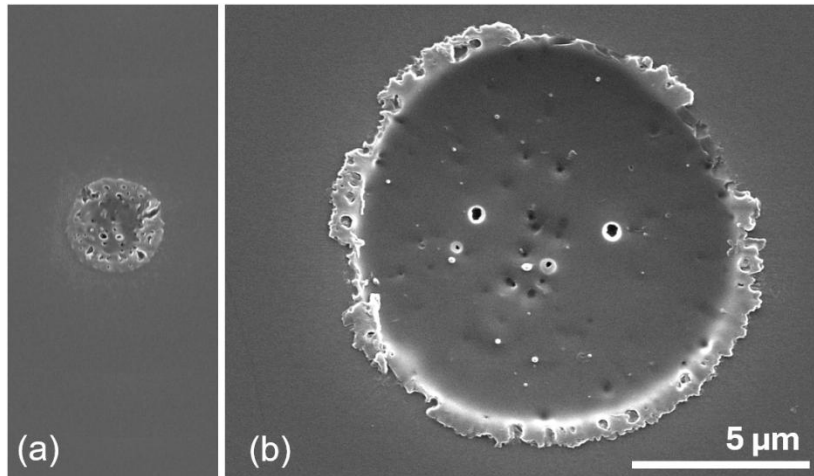


Figure 4. Scanning electron microscope images of single-pulse modifications created using (a) 2.3 J/cm^2 and (b) 23.7 J/cm^2 of peak laser fluence.

To investigate heat accumulation the two-temperature model was used to predict electron to lattice temperature transfer after the initial pulse energy deposition. Figure 6 shows the electron and lattice temperature evolution at the peak intensity location ($z = 0, r = 0$). For the incident pulse fluence of 2.3 J/cm^2 , equilibrium between the lattice and electron systems is achieved 150 ps after the delivery of the pulse. The electron temperature rises rapidly on the timescale of the pulse following the plasma density increase, while a comparatively slow decline is seen as the temperature is transferred to the lattice. The relatively long relaxation timescale compared to the femtosecond pulse duration indicates that energy deposition remains highly localized near the irradiated surface, consistent with the confined crater morphology shown in Fig. 7.

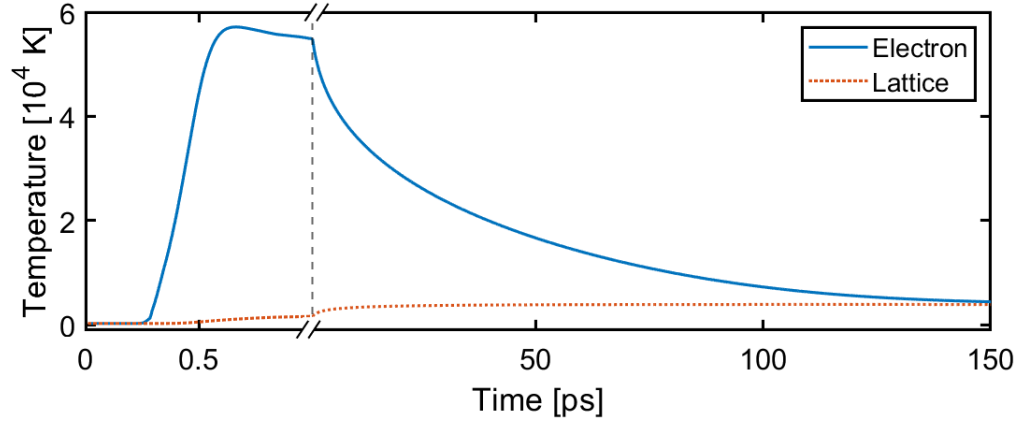


Figure 6. Lattice and electron temperature evolution at an input fluence of 2.3 J/cm^2 until equilibrium is reached.

Figure 7 displays the predicted temperature maps of the substrate after irradiation and temperature equilibrium of the TTM, with (a) 2.3 , (b) 4.3 and (c) 23.7 J/cm^2 peak pulse fluences. The result demonstrates the evolution of the lattice temperature and resultant material removal profile after laser irradiation. As energy deposition increases from Fig 7(a) to 7(c), the region exceeding the critical plasma density expands both laterally and in depth. Due to the minimal temperature dissipation at this timescale (as shown in Fig. 6) thermal accumulation remains localized near the crater, depicting the characteristic confined energy deposition using femtosecond lasers.

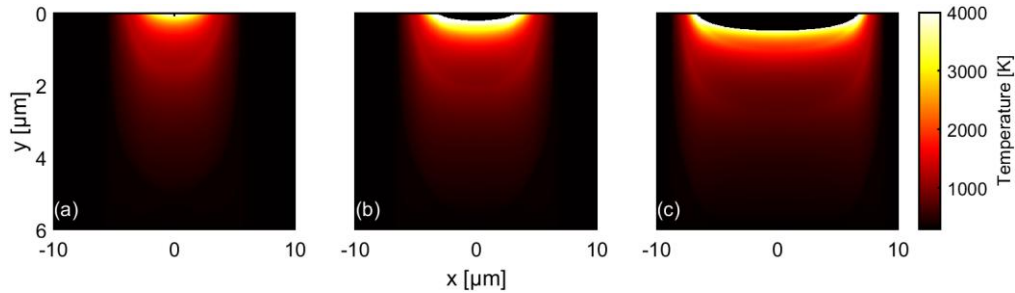


Figure 7. Lattice temperature after TTM equilibrium ($t = 150 \text{ ps}$) for an incident fluence of (a) 2.3 J/cm^2 , (b) 4.3 J/cm^2 , and (c) 23.7 J/cm^2 .

The volume of material above the softening temperature (1093 K) remains small and close to the crater region, within $2 \text{ }\mu\text{m}$ laterally and $5 \text{ }\mu\text{m}$ in depth from the boundaries of the crater. This indicates that any effect on the crater surroundings due to recasting and thermal softening will be minimal and highly localized.

4. Conclusion

A predictive modelling framework for femtosecond laser interaction with glass was developed by coupling the multiple rate equation for plasma generation with the two-temperature model for electron-lattice energy transfer. The model incorporates plasma-dependent Fresnel reflectance and nonlinear absorption to account for dynamic plasma shielding during pulse propagation and energy deposition. Experimental single-pulse ablation measurements demonstrated agreement with simulated crater morphology trends.

The simulations demonstrate that nonlinear absorption drives rapid free-electron plasma generation near the material surface, resulting in localized energy deposition, characteristic of femtosecond laser processing. With higher fluences, plasma shielding limits additional energy penetration into the substrate, producing saturation behavior in crater depth. The transition from sub-threshold thermal modification to material removal was accurately predicted through the evolution of electron density relative to the critical plasma density criterion.

The presented framework provides a physics-based approach for predicting ultrafast laser energy deposition and material response in dielectric substrates. These results contribute toward improved understanding and optimization of femtosecond laser micromachining of glass materials.

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Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

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