Microscale heat transfer in fusion welding of glass by ultra-short pulse laser using dual phase lag effects

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Abstract. The heat transfer in microscale has very different physical basis than macroscale where energy transport depends on collisions among energy carriers (electron and phonon), mean free path for the lattice (~ 10 – 100 nm) and mean free time between energy carriers. The heat transport is described on the basis of different types of energy carriers averaging over the grain scale in space and collisions between them in time scale. The physical bases of heat transfer are developed by phonon-electron interaction for metals and alloys and phonon scattering for insulators and dielectrics. The non-Fourier effects in heating become more and more predominant as the duration of heating pulse becomes extremely small that is comparable with mean free time of the energy carriers. The mean free time for electron – phonon and phonon-phonon interaction is of the order of 1 and 10 picoseconds, respectively. In the present study, the mathematical formulation of the problem is defined considering dual phase lag i.e. two relaxation times in heat transport assuming a volumetric heat generation for ultra-short pulse laser interaction with dielectrics. The relaxation times are estimated based on phonon scattering model. A three dimensional finite element model is developed to find transient temperature distribution using quadruple ellipsoidal heat source model. The analysis is performed for single and multiple pulses to generate the time temperature history at different location and at different instant of time. The simulated results are validated with experiments reported in independent literature. The effect of two relaxation times and pulse width on the temperature profile is studied through numerical simulation.

1. Introduction
Laser welding of metallic materials is well established process and used in many industries. With miniature of the components and devices, the use of controlled and regulated heat from the laser for material processing is ever increasing in nonmetals like ceramics and glasses. The structural modification inside the bulk of transparent material with the application of controlled heat has been used beneficially for the fabrication of photonic devices of transparent materials. However, the joining of glass is difficult because of their brittleness, transparency, fragileness and high glass transition temperature. The application in information and communication industry brings attention of laser welding on dielectric or semiconductor materials such as silica glass and silicon crystal [1 - 2]. Conventional methodology of joining transparent materials like glass using CO2 or Nd:YAG laser must utilize an intermediate layer to absorb laser energy at the interface corresponding to the wavelength of the lasers. Use of absorbing material may significantly alter the weld joint properties and characteristics [3]. However, this may not be a problem for metallic materials since it has high
absorptivity of laser with these wavelengths and the welding can be carried out without any absorbing medium.

The duration of pulse on-time in ultra-short pulse laser is in the order of femtosecond (~ 10^{-15} s) to picosecond (~ 10^{-12} s) that is different from long or short pulse laser (typically nanosecond-microsecond-millisecond). In long or short pulse laser, the pulse energy is dumped over a long period thus allows the development of temperature in equilibrium within the pulse duration i.e. it is assumed that the temperature develops instantaneously [4]. In ultra-short laser, there is a time lag between the application of heat flux and the development of temperature within the system [5]. Therefore, using ultra-short pulse laser in advanced material processing for micro-nano devices is becoming significant due to its capability of high precision control on the application of heat flux. The heat-affected zone (HAZ) is small enough to avoid splatter, cracks, voids and residual thermal stresses as compared to nano-to-millisecond pulse laser. The interaction of ultra-short pulse laser with transparent material has very different physical basis. The energy deposited by the laser pulse creates a melt volume near the focal position between transparent material due to nonlinear absorption such as multi-photon or tunnel absorption [6]. However, re-solidification of the molten pool leads to structural modification. Ultra-short pulses are shorter than the time needed for most of the energy to diffuse within the atomic lattice. Therefore, no heat is transferred to the surrounding material that eliminates any unwanted material change. Thus, the HAZ produced is smaller as compared to short or long pulse laser processing. The laser parameters like peak power and pulse repetition rate of the ultra-short pulse laser is significant to control the width and depth of heating as well as the changes of mode i.e. laser ablation to laser welding or vice versa [7].

Very few research groups have performed direct joining and welding of transparent materials (mainly glass) using ultra-short pulse laser of femtosecond to picosecond pulses [8 - 12]. Tamaki et al. [8 - 10] performed the joining of different types of glasses (fused silica and borosilicate glass) using either picosecond or femtosecond laser. With optimum combination of parameters, a good weld joint strength was achieved. Common materials such as fused silica, SiC, diamond, glass etc. fall under the category dielectric materials. They also dissipate very less energy, i.e. have low dielectric loss. In the present work, the transparent material like glass is considered under the category of dielectric material for ultra-short pulse laser processing.

In fusion welding of transparent material, the laser energy is absorbed by nonlinear process with multiphoton ionization followed by avalanche ionization and the absorbed laser energy in the free electrons is transferred to the lattice to provide the temperature field in bulk glass [2, 7]. Internal modification at high pulse repetition rates is due to heat accumulation effect. The use of femtosecond laser to locally alter the structure of bulk transparent material as well as joining of materials has gained significant attention in recent past. The optical devices are fabricated or joined by measuring the refractive index changes like binary data storage. However, the mechanism for producing a refractive-index change in bulk transparent material relies on cumulative heating of the material around the focal volume by absorption of high-repetition train of femtosecond laser pulses [12]. Therefore, fundamental understanding on the interaction of ultra-short pulse laser with transparent materials is significant even in joining of two components.

To analyze the heating process through ultra-short pulse laser, a mathematical model is developed using non-Fourier assumptions such as finite speed of thermal wave propagation. The pulse duration is such that any phase transformation occurs in non-equilibrium condition and finite time is required for attaining equilibrium strongly depends on laser-material interaction [13]. Hence, understanding of lag behaviour in heat transfer may minimize any unwanted material change, produce limited HAZ and minimize induced thermal stress using ultra-short pulse laser [14]. To account for the finite speed of thermal wave, Fourier equation is upgraded from parabolic to hyperbolic that analyse the transient phase of the ultra-short pulse laser heating process when laser pulse duration approaches to femtosecond or picosecond level and the mechanism of radiation absorption becomes important [15]. Two-temperature models have been widely used in the investigation of ultra-short laser heating on metal [16 - 17]. The thermal wave oscillation model treats the heat energy received as a wave and does the further analysis based on wave nature of thermal energy. However, the single or dual phase lags accounting non-zero-time relaxation with respect to heat flux and temperature gradient is
probably more representative of non-equilibrium system in continuum scale against the response of thermal disturbance.

In the present work, the heat transfer phenomena are analysed using dual-phase lag model by developing a three dimensional finite element model [18]. Very limited work has been conducted in terms of mathematical model for fusion welding of glass. However, literature lacks the computation of temperature distribution of fusion welding of glass using a distributed heat source. To represent the laser, a quadruple ellipsoidal heat source model is developed for the simulation of laser microwelding process. The weld pool shape and size for glass are validated with experimental data from literature [7]. The present analysis is significant because quasi-equilibrium distribution of temperature interpret the delay in reaching the peak temperature of the system even for a moving laser beam. Till date, there is no such finite element based numerical model exists to analyse non-Fourier heat conduction in the application of ultra-short pulse laser for welding and joining of glass. The fundamental understanding of the heat transfer process and quantitative measurement of different thermal zone are outcomes of the simulation process.

2. Numerical model
The finite element based numerical model is developed using dual phase lag non-Fourier heat conduction model. The volumetric heat generation-term is considered for the mathematical formulation. First, the governing equations are presented and the discretized form of the equations is derived over the solution domain. A quadruple ellipsoidal volumetric heat source with parabolic distribution of heat flux density along depth direction is proposed for temperature simulation. Finally, the computational aspects of the numerical model are described.

2.1. Governing equations
The Fourier law of heat conduction that reaches under equilibrium condition instantaneously is expressed as

\[
\dot{q} = -k \nabla T
\]  

(1)

where \( \dot{q} \) is the applied heat flux, \( \nabla T \) is the temperature gradient and \( k \) is the thermal conductivity of the medium. In dual phase lag model, the Fourier heat conduction is modified as

\[
\dot{q}(t + \tau_q) = -k \nabla T(t + \tau_T)
\]  

(2)

where \( \tau_q \) and \( \tau_T \) are two finite relaxation times corresponding to heat flux and temperature gradient. The unsteady state heat conduction with internal heat generation is written as

\[
k \nabla^2 T + \dot{Q} = \rho c \frac{\partial T}{\partial t}
\]  

(3)

First order expansion of eq. (2) and manipulating of all above equations, the non-Fourier heat conduction equation with double phase lag in three dimensional form is written as

\[
\dot{T} + \tau_q \dot{T} - \alpha (\nabla^2 T) - \tau_T \alpha (\nabla^2 T) = \frac{k}{\alpha} \dot{Q} + \tau_q \frac{k}{\alpha} \frac{\partial \dot{T}}{\partial t}
\]  

(4)

where thermal diffusivity \( \alpha = \frac{k}{\rho c} \). In laser welding, there is no internal heat generation. This term is accounted to incorporate the volumetric absorption of the laser energy. This volumetric heat flux is reasonable to assume that it is applied explicitly within the system without any time lag and the heat source does not vary with internal variable \( t \). Therefore, the last term of Eq. (4) can be neglected and the governing equation is simplified as

\[
\dot{T} + \tau_q \dot{T} - \alpha (\nabla^2 T) = \frac{k}{\alpha} \dot{Q}
\]  

(5)
Using Galerkin’s weighted residue technique and neglecting the boundary interaction, the Eq. (5) is discretized in three dimensional spaces as

\[ [K][\mathbf{T}] + [M][\mathbf{\dot{\mathbf{T}}}] + [C][\mathbf{\ddot{\mathbf{T}}}] = \{\mathbf{F}\} \]  \tag{6}

where different terms are expressed as

\[ [K] = \int_{\Omega} k \left[ [N_x][N_x] + [N_y][N_y] + [N_z][N_z] \right] d\Omega \]  \tag{7}

\[ [M] = \int_{\Omega} \tau \rho c[N][N] d\Omega \]  \tag{8}

\[ [C] = \int_{\Omega} \rho c[N][N] d\Omega + \int_{\Omega} \tau k \left[ [N_x][N_x] + [N_y][N_y] + [N_z][N_z] \right] d\Omega \]  \tag{9}

\[ \{F\} = \int_{\Omega} \left( \frac{\kappa}{\alpha} Q \right) (N) d\Omega \]  \tag{10}

The matrix \([K],[M]\) and \([C]\) can be calculated by numerical integration. Gauss quadrature method is used for numerical integration. The second order time discretization follows Newmark method [19].

2.2. Heat source representation

The typical geometry of the weld profile on glass indicates the surface only heat flux is not sufficient to represent laser energy absorption within the system. The profile indicates the double ellipsoidal shape of the cross-section. In addition, due to linear movement of the heat source the front and rear part of the source are non-symmetrical. Hence, quadruple heat source model to account non-symmetry energy distribution by combining four ellipsoids with C\(^1\) continuity (Figure 1) better represent the volumetric heat. Since the laser is focused on the middle of the specimen, it is quite convincing to apply laser energy over a volume rather than a surface. The volumetric energy better represents the keyhole or string of energy in laser welding process. It is also assumed that the volumetric heat is applied to the solution domain without any relaxation in time. Once the volumetric heat flux is applied to the solution domain, there is relaxation in the development of temperature gradient and heat flux in dual phase lag model. The distribution of energy may not follow exponential decay along the Z-axis like GTA welding process. The distribution is relatively uniform towards depth direction. Hence, a parabolic distribution along the depth direction is proposed instead of exponential decaying trend for double ellipsoidal heat source model. Figure 1 depicts the volumetric heat source consists of parts of four ellipsoidal.

![Figure 1](image-url)

**Figure 1.** (a) Fusion zone of glass [7] and (b) proposed quadruple heat source model.

The volumetric heat flux distribution is represented as
\[ q(x, y, z) = q_m e^{-(Ax^2 + By^2)(1 - Cz^2)} \tag{11} \]

where \( A, B \) and \( C \) are distribution parameters and \( q_m \) is the maximum intensity at the centre point ‘O’.

The semi-axes of first ellipsoid are \( a_1, b, c_1 \) in the \( x, y, z \) axes directions respectively. It is assumed that the heat density falls to 0.05\( q_m \) at the surface of the ellipsoid. Therefore, along \( x \)-axis;

\[ q(a_1, 0, 0) = q_m \times \exp\left(-Aa_1^2\right) = 0.05 \times q_m \Rightarrow \exp\left(-Aa_1^2\right) = 0.05 \Rightarrow A \approx \frac{3}{a_1^2} \tag{12} \]

Similarly, \( B \approx \frac{3}{b^2} \); However, flux density is exactly zero at a distance \( c \) along \( Z \)-axis. Therefore, \( C = \frac{1}{c_1^2} \). The volumetric heat energy for the first ellipsoid is estimated as

\[ Q_1 = \int_0^\infty \int_0^\infty \int_0^{c_1} q(x, y, z)dxdydz = q_m \frac{\pi a_1 b c_1}{9} \tag{13} \]

The maximum heat flux intensity at the origin, in general, is defined by

\[ q_m = \frac{9\times Q}{4\pi abc} \tag{14} \]

The temperature gradient at the front and rear are different and as well as non-symmetric profile along the \( Z \)-direction. The fractions in heat deposited are accounted in the general expression of heat flux density distribution in eq. (11) and is expressed as

\[ q_{f1}(x, y, z, t) = \frac{9\times Q\times N_{f1}}{4\pi a_1 b c_1} \times e^{\left[\frac{3x^2}{a_1^4} + \frac{3y^2}{b^4}\right]} \left(1 - \frac{z^2}{c_1^2}\right) \tag{15} \]

The total amount of energy is estimated from distribution (by integrating),

\[ Q = \frac{1}{4} N_{f1}Q + \frac{1}{4} N_{f2}Q + \frac{1}{4} N_{r1}Q + \frac{1}{4} N_{r2}Q \tag{16} \]

where \( N_i \) and \( N_r \) are the fractions of heat deposited in the front and the rear sections, respectively such that

\[ N_{f1} + N_{r1} + N_{f2} + N_{r2} = 4 \tag{17} \]

Therefore, the fraction due to non-symmetrical heat deposited in the front section of the ellipsoid is expressed as

\[ N_{f1} = \frac{4a_1 c_1}{a_1 c_1 + a_2 c_1 + a_1 c_2 + a_2 c_2} \tag{18} \]

Total heat input (\( Q \)) is also estimated from the effective heat energy transferred to the workpiece from any heat source. Therefore, it is mathematically expressed as

\[ \beta Q = \eta P \Rightarrow Q = \frac{\eta P}{\beta} \tag{19} \]

where, \( P \) is the laser power and the constant term \( \alpha \) is introduced to account the over-estimation of heat energy due to discretized geometry (\( \beta \sim 1.1 \)). The terms \( \eta \) is the efficiency of heat source when it interact with the materials. Therefore, the value of ‘\( Q \)’ here is used to estimate the heat flux density
distribution at Eq. (15). It is convenient to introduce a fixed coordinate system \((\chi', y, z)\) on the workpiece such that
\[
x = \chi' + v(\tau - t)
\]  
(20)

when the heat source moves along the \(\chi'\)-axis, and \(v\), and \(\tau\) depict the welding speed, and a lag factor. The lag factor \(\tau\) is used to define the position of heat source at time \(t = 0\) on fixed coordinate system \((\chi', y, z)\). Therefore, the heat density distribution inside the front part of ellipsoid for material 1 within the fixed coordinate system is expressed as:

\[
q_{f1}(\chi', y, z, t) = \frac{9Q \times N_{f1}}{4 \pi a_1 b c_1} \times e^{-\frac{(3x'^2 + v(\tau - t))^2}{a_1^2}} \times e^{-\frac{3y^2}{b^2}} \left(1 - \frac{z^2}{c_1^2}\right)
\]  
(21)

2.3. Computational aspects

A finite element code using FORTRAN language is developed to simulate the temperature distribution for ultra-short pulse laser welding of glass. The linear system of equation depicted by eq. (10) is solved with second order temporal discretization. The ‘Lis’ i.e. library of iterative solver is implemented in the finite element code [20]. A volumetric heat generation term is also introduced resembling the laser welding process of glass. The most significant issue of the computational part is the choice of time step because total computational time for the desired solution geometry depends on the time step chosen. The cycle time of 50 kHz pulse laser is 20 μs with a pulse width of only 10 ps. Therefore, initial time step is considered as 1 ps which corresponds to \(2 \times 10^6\) times steps for single pulse. It accounts huge computational time. Hence, an adaptive time increment scheme is designed to increase the time step in the cooling zone that reduces the computation time exponentially based on the temperature change between two successive iterations.

In the present work, the dual phase lag model is considered for first order expansion of \(\tau_q\) and \(\tau_T\) from Eq. 2 that indicates the temperature profile is always stable (i.e. not wavy in nature). It is noteworthy that \(\tau_q < \tau_T\) (one order less or greater) is followed here to have obvious effect of relaxation times. In particular, the physical significance of non-zero \(\tau_q\) and \(\tau_T\) are interpreted as thermal inertia and microstructural interaction i.e. delay in establishing heat flux and delay in establishing temperature gradient across the body, respectively. In general, the relaxation times for insulators and dielectric films are represented as [15]

\[
\tau_T = \frac{9\tau N}{5} ; \tau_q = \tau_R
\]  
(22)

Where \(\tau_N\) and \(\tau_R\) are the relaxation times measuring the normal and umklapp (resistive) process in phonon scattering. However, these parameters are also temperature dependent. Therefore, it is extremely difficult to define the unified value of relaxation parameters. In the present analysis, \(\tau_q\) is considered of the order of 1 ps and \(\tau_T\) is 10 ps. The simplest correlation to define the relaxation parameter is \(\tau_q = \frac{a}{c^2}\) where \(a\) is the thermal diffusivity and \(c\) is the speed of thermal wave in that medium.

Figure 2a shows typical 3D temperature distribution over the solution domain of 0.1 mm x 0.1 mm x 0.05 mm at pulse frequency of 200 kHz. The pulse energy is 4.98 μJ released in the duration of 10 ps. The material property used in the present investigation is depicted in Table 1. The temperature profile is non-symmetric in nature due to moving laser along Y –direction. The colour code depicted in Fig. 2a indicates that the molten region corresponds to the isothermal line of 1323 K [7]. The red zone resembles the modification of inner structure due to highly focused laser beam that produce the electron cloud [21]. The fusion zone is confined into a small area that is typical characteristic of ultrashort pulse laser. The heat conductivity is several times less than metal as well as thermal diffusivity is almost one order less than metal. Figure 2b shows the few fixed points on the substrate material over which the temperature is recorded as a function of time.
Table 1. Material properties used in the present simulation.

| Property           | Value       |
|--------------------|-------------|
| Density            | $2.51 \times 10^3$ kg/m$^3$ |
| Specific heat      | 820 J/kgK   |
| Thermal conductivity | 0.96 W/mK |
| Absorptivity       | 0.80 – 0.85 |

Figure 2. (a) 3D temperature distribution using quadruple heat source model; (b) Location on the workpiece to measure temperature.

3. Results and discussion
The experimental data for the analysis is considered from independent literature [Ref. 7]. Ultra-short pulse laser (wavelength 1064 nm and $M^2 = 1.1$) with pulse duration of 10 ps is used for heating of borosilicate glass with a laser scanning speed of 20 mm/s [7]. The pulse repetition rate (or frequency) varies from 50 kHz to 500 kHz and corresponding pulse energy of 10.3 μJ to 2.46 μJ [7]. Figure 3a shows the temperature distribution of a single pulse at frequency of 50 kHz. Although the pulse duration is 10 ps, the peak temperature reaches after 50 ns due to lagging responses by differential effect of relaxation times. With increase in pulse frequency (i.e. decrease in pulse energy), although the peak temperature decreases, the delay in reaching peak temperature increases (Fig. 3b). Since the pulse energy decrease with increase in frequency, the peak temperature achieved decreases even for a constant pulse width. Since there is a difference in reaching peak temperature, it is concluded that the thermal inertia effect is significant at high frequency or high pulse repetition rate. The influence of pulse energy supply on temperature distribution is depicted in Fig. 3c at a frequency of 500 kHz. Temperature increases in successive pulses due to accumulation of heat by pulse energy. The peak temperature of 1200 K reaches at about 10 pulses. However, the increment is quite high as compared to metallic material due to difference in thermo-physical properties. Heat dissipation is restricted by the relaxation of heat transport with such picosecond level pulse duration and even at high pulse repetition rate. The variation in temperature profile is more obvious at a fixed location 1 as compared to location 2. The effect of on-off mode of pulse energy in a cycle time diminishes away from centre of applied heat flux.
Figure 3. Simulation of time-temperature profile for (a) single pulse, (b) single pulse for different pulse frequencies and (c) multiple pulses.

Figure 4 depicts the comparison between computed weld zone and corresponding experimentally measured macrographs [7]. At low pulse frequency and high pulse energy of 10.3 µJ, the molten zone produced is confined into a small area as compared to high frequency and low pulse energy. In the former case, the rate of heat accumulation is probably high with the combined effect of high pulse energy and low pulse frequency. In metals, the ablation mechanism is active with these process condition whereas high pulse frequency and low pulse energy tends to form comparatively larger molten zone with less peak temperature. This mode is suitable for welding or joining of materials. Overall, the heat-affected zone is limited and the material experiences rapid melting and solidification over a narrow space. The computed temperature profile agrees well with the experimentally measured macrographs as observed in Fig. 4.

Figure 4. Comparison between experimental [7] and computed macrographs at pulse frequency of (a) 50 kHz and (b) 200 kHz.

Figure 5 depicts the temperature profile on a transverse plane at a fixed location. Since the pulse frequency and pulse energy are different all these three cases, the temperature profile are not similar even for similar location and similar welding speed. The molten zone is more for the least pulse frequency and is less for the highest pulse frequency. This indicates that the developed numerical model is capable to consider the effect of pulse in laser welding.

Figure 6 depicts the time-temperature profile at different pulse frequency. Over a fixed time span, the peak temperature is very high for 50 kHz pulse frequency whereas the system reaches moderate maximum temperature at the frequency of 500 kHz. The low pulse frequency and high pulse energy creates the condition of material ablation with the maximum difference in the temperature. The variability of temperature within one pulse is the least for the case of highest frequency.
To investigate the influence of relaxation times in the simulation of temperature profile, the reference welding condition is considered as 200 kHz pulse frequency and 4.98 μJ pulse energy and the temperature are recorded at location 1. It is obvious from the figure that there are not many significant changes in the temperature profile even with changes of relaxation times several folds. Thus, the influence of relaxation times is less sensitive on temperature distribution for glass as compared to metal films [18].

The influence of pulse width is shown in Fig. 8 considering the reference condition of 200 kHz pulse frequency and 4.98 μJ pulse energy. With the increase in pulse width of ten times as compared to the present case, there is considerable increase in temperature. The longer pulse generates higher temperature and the difference between maximum to minimum temperature in a pulse increases. This phenomenon is due to thermal inertia that occurs in almost every pulse heating process. When the pulse width is too low, the temperature profile is lower side i.e. below melting point temperature even after 100 μs. Hence, the selection of optimum pulse width for a specific system is necessary to find the feasible solution of welding than ablation of materials.
4. Conclusions
The experimental measurement of all the parameters during ultra-fast laser processing is always expensive. Hence, the demand of a reliable process model using phase lag effects is ever increasing. Although a finite element based numerical model is developed to estimate three dimensional temperature distributions during welding of glass, the optimization of the process parameters and incorporation of temperature dependent material properties are still challenging task to enhance the reliability of the numerical process model. This can be considered as future direction of work for extensive use of the developed model. The significant findings from the present work are described below.

• The pulse energy is incorporated through volumetric heat generation term of the non-Fourier heat conduction equation. The quadruple heat source model along with parabolic distribution is proposed to incorporate non-symmetric temperature distribution.
• By interacting of the ultra-short pulse laser, the temperature generated at the laser focal point is considerably high. This is reduced by increasing the pulse repetition rate and by decreasing pulse energy.
• The delay in reaching peak temperature is of the order of ns after application of 10 ps pulse laser.
• The magnitude of relaxation times is less sensitive with the temperature profile than a metallic material.
• There is significant impact of pulse width during welding of glass using ultra-short pulse laser. Selection of optimum pulse width is helpful for the feasibility of welding.
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