Uncertainty Analysis of Melting and Resolidification of Gold Film Irradiated by Nano- to Femtosecond Lasers Using Stochastic Method

A sample-based stochastic model is presented to investigate the effects of uncertainties of various input parameters, including laser fluence, laser pulse duration, thermal conductivity constants for electron, and electron–lattice coupling factor, on solid–liquid phase change of gold film under nano- to femtosecond laser irradiation. Rapid melting and resolidification of a free-standing gold film subject to nano- to femtosecond laser are simulated using a two-temperature model incorporated with the interfacial tracking method. The interfacial velocity and temperature are obtained by solving the energy equation in terms of volumetric enthalpy for control volume (CV). The convergence of variance (COV) is used to characterize the variability of the input parameters, and the interquartile range (IQR) is used to calculate the uncertainty of the output parameters. The IQR analysis shows that the laser fluence and the electron–lattice coupling factor have the strongest influences on the interfacial location, velocity, and temperatures. [DOI: 10.1115/1.4032962]

Keywords: phase change, melting, resolidification, uncertainty, sample-based stochastic model

1 Introduction

At micro- and nanoscales, ultrafast laser material processing is a very important part in fabrication of some devices. Conventional theories established on the macroscopic level, such as heat diffusion assuming Fourier’s law, are not applicable for the microscopic condition because they describe macroscopic behavior averaged over many grains [1]. For ultrashort laser pulses, the laser intensities can be high as \(10^{12}\) W/m\(^2\) or even higher up to \(10^{21}\) W/m\(^2\). During the laser interaction with materials, those electrons in the range of laser penetration of a metal material absorb the energy from the laser light and move with the velocity of ballistic motion. The hot electrons diffuse their thermal energy into the deeper part of the electron gas at a speed much slower than that of the ballistic motion. Due to the electron–lattice coupling, heat transfer to the lattice also occurs and a nonequilibrium thermal condition exists [2]. The non-equilibrium of electrons and the lattice are often described by two-temperature models by neglecting heat diffusion in the lattice [3,4]. The accurate thermal response is only possible when the lattice conduction is taken into account in the physical model, particularly in the cases with phase change. Chen and Beraun [5] proposed a dual hyperbolic model which considered the heat conduction in the lattice.

In the physical process, melting in the lattice could take place for laser heating at high fluence. When the lattice is cooled, the liquid turns to solid via resolidification. The solid will be superheated in the melting stage, and the liquid will be undercooled in the resolidification stage. When the phase change occurs in a superheated solid or in an undercooled liquid, the solid–liquid interface can move at a very high velocity. Kuo and Qiu [6] investigated picoseconds laser melting of metal films using the dual-parabolic two-temperature model. Chowdhury and Xu [7] modeled melting and evaporation of gold film induced by a femtosecond laser. During the melting stage, the solid is superheated to above the normal melting temperature. During resolidification, the liquid is undercooled by conduction and the solid–liquid interface temperature can be below melting point. The solid–liquid interface can move at a high velocity which implies that the phase change is controlled by the nucleation dynamics, rather than energy balance [8].

In the melting and resolidification model of metal under picosecond/femtosecond laser heating, the energy equation for electrons was solved using a semi-implicit scheme, while the energy and phase change equations for lattice were solved using an explicit enthalpy model [6,7]. The explicit scheme is easier to implement numerically than the implicit scheme for the enthalpy model [9].

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characteristics of the output results. Among them, the laser fluence and pulse duration may fluctuate during the process. Moreover, the thermophysical properties of electrons and the lattice are not accurately determined at high temperatures. For example, the electron phonon coupling factor can increase, decrease, or exhibit nonmonotonic changes with increasing electron temperature [15]. These parametric uncertainties may influence the characteristics of the phase change processes (melting and resolidification) which will affect the predictions of interfacial location, temperature, and velocity and also the electron temperature. In the selective laser sintering, the fluence and width of laser pulses and the size of metal powder particles may influence the characteristics of the final product [16–20]. Therefore, study of parametric uncertainty is vital in simulation of the phase change of metal particles under nano- to femtosecond laser heating.

Sample-based stochastic model has been proposed to analyze the effects of the uncertainty of the parameters in order to integrate the parametric uncertainty distribution. Stochastic models possess some inherent randomness where the same set of parameter values and initial condition will lead to ensemble of different outputs. The stochastic model was applied on the nonisothermal filling process to investigate the effect of the uncertainty of parameters [21]. An improved simulation stochastic model was used in the Aspen (a chemical process simulator) process simulator by Diwekar and Rubin [22]. The applications of the stochastic model in optical fiber drawing process [23,24], thermosetting-matrix composite fabrication [25], microresonator [26], and proton change membrane (PEM) fuel cells [27] were found in the open literature. The sample-based stochastic model was applied to study the phase change of metal particle under uncertainty of particle size, laser properties, and initial temperature to investigate the influences of the output parameters in the solid–liquid–vapor phase change of metal under nanosecond laser heating [28]. COV was used to characterize the variability of the input parameters where the IQR was used to measure the uncertainty of the output parameters.

In this paper, the sample-based stochastic model will be applied to study the melting and resolidification of gold film irradiated by nano to femtosecond laser under certain electron-phonon coupling factor, laser fluence, laser pulse width, and constants for electron thermal conductivity to reveal the different influences of those parameters in the interfacial location, interfacial velocity, and interfacial and electron temperatures.

2 Physical Model
A gold film with a thickness $L$ and an initial temperature $T_i$ is subjected to a laser pulse with an full-width half-maximum pulse width $t_p$ and fluence $J$ from the left hand surface. The energy equations of the free electrons and the lattice are

$$\frac{C_e}{C_f} \frac{\partial T_e}{\partial t} = \frac{\partial}{\partial x} \left( k_e \frac{\partial T_e}{\partial x} \right) - G(T_e - T_i) + S$$

(1)

$$\frac{C_l}{C_f} \frac{\partial T_l}{\partial t} = \frac{\partial}{\partial x} \left( k_l \frac{\partial T_l}{\partial x} \right) + G(T_e - T_l)$$

(2)

where $C$ represents heat capacity, $k$ is the thermal conductivity, $G$ is the electron–lattice coupling factor, and $T$ is the temperature. The heat capacity of electrons expressed as below is only valid for $T_e < 0.1 T_F$ with $T_F$ denoting Fermi temperature

$$C_e = B_e T_e$$

(3)

where $B_e$ is a constant. According to Chen et al. [29], the electron heat capacity can be approximated by the following relationship:

$$C_e = \begin{cases} B_e T_e & T_e < \frac{T_F}{\pi^2} \\ \frac{2B_e T_e}{3} + \frac{C_e}{3} & \frac{T_F}{\pi^2} \leq T_e < \frac{3 T_F}{\pi^2} \\ \frac{N k_B}{3} + \frac{C_e}{3} & \frac{3 T_F}{\pi^2} \leq T_e < T_F \\ \frac{3 N k_B}{2} & T_e \geq T_F \end{cases}$$

(4)
where

\[
C'_e = B_e \frac{T_F}{\pi^2} + \frac{3NkB_eT_F}{2} \left( \frac{T_e}{T_F} - \frac{T_F}{\pi^2} \right)
\]

(5)

The bulk thermal conductivity of metal at equilibrium can be represented as

\[
k_{eq} = k_e + k_i
\]

(6)

Fig. 2  Stochastic convergence analysis of mean value of the input parameters (a) \(G_{RT}\), (b) \(\lambda\), (c) \(\eta\), (d) \(J\), and (e) \(t_p\)
Fig. 3  Stochastic convergence analysis of standard deviation of the input parameters (a) $G_{RT}$, (b) $\lambda$, (c) $\eta$, (d) $J$, and (e) $t_p$
At the nonequilibrium condition, the thermal conductivity of electrons depends on both electron and lattice temperatures. For a wide range of electron temperature ranging from room temperature, the thermal conductivity of electron can be measured as follows [30]:

$$k_e = \lambda \left( \frac{0.16}{(\theta_e^2 + 0.092)^{1/2}} \right) \theta_e$$  \hspace{1cm} (7)

where $\theta_e = T_e/T_F$ and $\theta_l = T_l/T_F$ are dimensionless temperature parameters and $\lambda$ and $\eta$ are the two constants for the thermal conductivity of electrons. In general, the values of those two constants for gold are $\lambda = 353$ W/mK and $\eta = 0.16$. For the low electron temperature limit ($\theta_e \ll 1$), the electron thermal conductivity can be expressed as

$$k_e = k_{eq} \left( \frac{T_e}{T_F} \right)$$  \hspace{1cm} (8)

Under high energy laser heating, the electron and lattice temperatures change significantly which results in a temperature-dependent coupling factor in the ultrafast laser heating. Chen et al. [14] proposed a relationship between electron and lattice temperatures for the coupling factor as follows:

$$G = G_{RT} \left[ \frac{A_e}{B_l} (T_e + T_l) + 1 \right]$$  \hspace{1cm} (9)

where $A_e$ and $B_l$ are two material constants for the electron relaxation time; $G_{RT}$ is the room temperature coupling factor.

The heat source term in Eq. (1) can be represented as

$$S' = 0.94 \frac{1 - R}{t_p(\delta + \delta_b)[1 - e^{-t_p/(\delta + \delta_b)}]} J \exp \left( \frac{x}{\delta + \delta_b} - 2.77 \left( \frac{t_p}{t_p} \right)^2 \right)$$  \hspace{1cm} (10)

where $R$ is reflectivity of the film, $J$ is the laser fluence, $\delta$ is the optical penetration depth, and $\delta_b$ is the ballistic range. At equilibrium, the bulk thermal conductivity of metal is measured as the summation of the electron thermal conductivity ($k_e$) and lattice thermal ($k_l$) conductivity. Free electrons are dominated in the heat conduction as the conduction mechanism is defined by the diffusion of free electron. So, for gold, the lattice and electron thermal

Fig. 4  Stochastic convergence analysis of mean value of the output parameters (a) $s$, (b) $u_n$, (c) $T_{lp}$ and (d) $T_e$
conductivities are taken as 1% and 99% of the bulk thermal conductivity, respectively [31].

The energy balance at the solid–liquid interface in the system is given as

\[ k_l \frac{\partial T_l}{\partial x} - k_s \frac{\partial T_s}{\partial x} = \rho_l h_m u_s \]

(11)

where \( k_l \) is the mass density of liquid, \( h_m \) is the latent heat of fusion and \( u_s \) is the solid–liquid interfacial velocity. For a metal under superheating, the velocity of solid–liquid interface is expressed as follows [6]:

\[
  u_s = V_0 \left[ 1 - \exp \left( - \frac{h_m}{R_g T_m} \frac{T_{II} - T_m}{T_{II}} \right) \right]
\]

(12)

where \( V_0 \) is the maximum interfacial velocity, \( T_{II} \) is the interfacial temperature, and \( R_g \) is the gas constant. The interfacial temperature could be higher than the normal melting temperature during melting and lower during solidification. The boundary conditions are given as

\[
  \left. \frac{\partial T_e}{\partial x} \right|_{x=0} = \left. \frac{\partial T_e}{\partial x} \right|_{x=L} = \left. \frac{\partial T_l}{\partial x} \right|_{x=0} = \left. \frac{\partial T_l}{\partial x} \right|_{x=L} = 0
\]

(13)

The initial temperature conditions are

\[
  T_e(x, -2t_p) = T_l(x, -2t_p) = T_i
\]

(14)

The total computational domain is discretized with nonuniform grids. The implicit finite-difference equations are solved in each of the CV and time step. The numerical solution starts from time \(-2t_p\). During the solving process, the lattice temperature is set as interfacial temperature for that CV that contains solid–liquid interface location. The energy equation in terms of enthalpy form is applied and solved for the solid liquid interface CV. The relationship of interfacial temperature and liquid fraction can be written as

\[
  C_{II} \frac{\partial T_{II}}{\partial t} + \rho_l h_m \frac{\partial f}{\partial t} = \frac{\partial}{\partial x} \left( k \frac{\partial T_l}{\partial x} \right) + G(T_e - T_l)
\]

(15)
where $T_{ij}$ is the interfacial temperature, $C_{is}$ is the heat capacity at solid–liquid interface, and $f$ is the liquid fraction in the system. The liquid fraction is related to the location of the solid–liquid interface [11]. Before onset of melting, Eqs. (1) and (2) are solved simultaneously to obtain electron and lattice temperatures until the lattice temperature exceeds the melting point. Once it exceeds, the lattice temperature is set as the melting temperature and phase change will be considered in the system. After melting starts, an

Fig. 6  Typical distributions of the input parameters (a) $G_R$, (b) $\lambda$, (c) $\eta$, (d) $J$, and (e) $t_p$.
iterative procedure is applied to find the interfacial temperature and the interfacial location at each time step [10].

3 Stochastic Modeling of Uncertainty

Stochastic modeling is a process where the variability of the output parameters is evaluated based on the different combination of the input parameters [28]. In this paper, a sample-based stochastic model is used to study the melting and resolidification of the gold film under uncertain laser fluence, pulse width, coupling factor, and thermal conductivity of electrons to show the effects of the output parameters such as interfacial location, interfacial temperature, interfacial velocity and electron temperature. Figure 1 shows the detailed procedure of stochastic modeling.

In the stochastic modeling process, the first need is to quantify the degree to which the input parameters vary, and then to determine the appropriate number of combination of the input parameters to use with a stochastic convergence analysis. After determining the number of combination of input parameters, one needs to calculate the uncertainties of the input parameters through the deterministic physical model that was previously established. Eventually, the variability of the output parameters is quantified based on the uncertainty of input parameters. The coupling factors at room temperature between electron and lattice ($G_{RT}$), laser fluence ($J$), electron thermal conductivity constants ($\lambda$ and $\eta$), and laser pulse duration ($t_p$) are the input parameters whose uncertainties are going to be investigated. Due to the unavailability of experimental distribution of those uncertain parameters, it is acceptable to assume that all the input parameters follow Gaussian distributions of uncertainty [23]. The Gaussian distribution is defined by a mean value ($\mu$) and a standard deviation ($\sigma$), where the mean value is expressed by the nominal value of uncertainty parameters and the standard deviation represents the uncertainty of the input parameters. The coefficient of variance (COV) is an important parameter which represents the degree of uncertainty of the input parameters. The COV is defined as

$$\text{COV} = \frac{\sigma}{\mu}$$

After determining the distributions of the input parameters, a commonly used sampling method called Monte Carlo sampling (MCS) is used to obtain the combination of the input parameters. According to the MCS, input parameters are randomly selected from their prescribed Gaussian distributions and combined them together as one sample. Due to the high dependency on the number of the samples of input parameters on the variability of the output parameters, the exact number of samples of input parameters is determined carefully. In the stochastic convergence

![Fig. 7 Typical distributions of the output parameters (a) $s$, (b) $u_s$, (c) $T_{I,h}$ and (d) $T_e$](image-url)
process, when the number of the sample increases the mean value and the standard deviation of input parameters converge to the nominal mean value and standard deviation of the Gaussian distribution. The mean value and standard deviation of the output parameters will also converge within a certain tolerance. After selecting the required number of samples for each input parameter, the physical model of melting and resolidification of gold film is solved. The effects of the input parameters variability on the output parameters uncertainty are evaluated by obtaining probability distribution of output parameters. The output parameters in this paper include interfacial location \( s \), interfacial temperature \( T_{I,I} \), interfacial velocity \( u_s \), and electron temperature \( T_e \). The probability distribution is calculated from the resulting set of the output parameters. The IQR is a measurement of variability, based on dividing a data set into quartiles. It is defined as the difference between the 25th percentile and the 75th percentile

\[
IQR = P75 - P25
\]  

4 Results and Discussion

The thermophysical and optical properties of pure gold film are [11]: \( B_s = 70 \text{ J/m}^2\text{K} \), \( A_s = 1.2 \times 10^7 \text{ K}^{-2}\text{s}^{-1} \) and \( B_I = 1.23 \times 10^{11} \text{ K}^{-1}\text{s}^{-1} \), \( G_{RT} = 2.2 \times 10^{16} \text{ W/m}^2\text{K} \) (solid) and \( 2.6 \times 10^{16} \text{ W/m}^2\text{K} \) (liquid), \( \rho = 19.30 \times 10^3 \text{ kg/m}^3 \) (solid) and \( 17.28 \times 10^3 \text{ kg/m}^3 \) (liquid) reflectivity, \( R = 0.6 \), \( \delta = 20.6 \text{ nm} \), \( \delta_n = 105 \text{ nm} \), \( T_m = 1336 \text{ K} \), \( T_F = 6.42 \times 10^4 \text{ K} \), \( h_m = 6.373 \times 10^3 \text{ J/kg} \), and \( V_0 = 1300 \text{ m/s} \). The sample-based stochastic model provides the output parameter distributions with respect to the uncertain input parameter distributions. A large number of input samples is required to get the real distribution of the output parameters. Due to the difficulty in prohibitively intensive computation, it is important to find a minimum number of input samples \( N \) with which steady necessary output distributions can be generated.

To find the required number of \( N \), we assume the nominal mean values of \( G_{RT}, \lambda, \eta, J, \) and \( t_p \) are \( 2.2 \times 10^{16} \text{ W/m}^2\text{K}, \) 353 W/mK, 0.16, 0.3 J/cm², and 20 ps, respectively. The coefficient of variance (COV) of each input parameter is set to be 0.02. Figure 2 represents the stochastic convergence analysis of the mean value of the input parameters \( G_{RT}, \lambda, \eta, J, \) and \( t_p \). It is shown from this figure that when the number of samples is small, the mean values of the input parameters fluctuate significantly. For the value \( N = 200 \), the mean values of the input parameters oscillate in a smaller range, suggesting that a total of 200 samples should be sufficient for steady nominal mean values of input parameters. Figure 3 represents the stochastic convergence analysis of standard deviation of the five input parameters. It is shown that
although the mean values of input parameters converges for 200 samples, the standard deviation still fluctuate. The reason behind this is that the deviation is a higher order moment which allows converging slower than the mean value. From Fig. 3, it may conclude that the minimum number of the input samples is 300. After determining the minimum number of input samples, the stochastic convergence analysis for the mean value and standard deviation of the output parameters are obtained, as shown in Figs. 4 and 5. It can be seen that when the number of the samples is beyond 300, the mean values of all the output parameters fluctuate in a smaller range (2.5%). Therefore, the minimum number of samples $N = 400$ is selected and used to calculate the results.

Figure 6 shows the typical distributions of the input parameters with the nominal mean values of $G_{RT}$, $\lambda$, $\eta$, $J$, and $t_p$ being $2.2 \times 10^{16}$ W/m$^2$K, 353 W/mK, 0.16, 0.3 J/cm$^2$, and 20 ps, respectively, and the COV of each parameter being 0.02. Figure 7 gives the typical distribution of the output parameters $s$, $u_s$, $T_{I,I}$, and $T_e$. In the histograms, the distributions of the output parameters are no longer Gaussian due to the nonlinear effect in the solid liquid interface.

The IQRs of the output parameters $s$, $u_s$, $T_{I,I}$, and $T_e$ as functions of COV of the input parameters $G_{RT}$, $\lambda$, $\eta$, $J$, and $t_p$ are shown in Fig. 8. When the COV of the one input parameter increases from 0.01 to 0.03 and the COVs of the other input parameters are kept constant at 0.01, the effect of that input parameter can be manifested. The IQRs of interfacial location, interfacial velocity, interfacial temperature, and electron temperature significantly increase from 1.5 nm to 4.5 nm, 8.4 m/s to 23 m/s, 298 K to 790 K, and 38 K to 110 K, respectively, when the COV of $J$ increases from 0.01 to 0.03. The IQR of interfacial location, interfacial velocity, interfacial temperature, and electron temperature significantly increases from 1.5 nm to 2.6 nm, 8.4 m/s to 16 m/s, 298 K to 550 K, and 38 K to 79 K, respectively, with the change of COV of $G_{RT}$ from 0.01 to 0.03. On the contrary, the COV of the thermal conductivity constants is relatively less impact to the interfacial location.

The IQR analysis of the interfacial velocity ($u_s$) shows that the laser influence $J$ is also most influential among the five input parameters. With the increment of COV of $J$ from 0.01 to 0.03, the IQR of $u_s$ increases from 8.8 m/s to 23.1 m/s. As shown in Fig. 8, the order of influence of the COV of the five input parameters on the output parameters are $J$, $G_{RT}$, $t_p$, $\lambda$, and $\eta$. Figure 9 represents the IQRs of $s$, $u_s$, $T_{I,I}$, and $T_e$ for different laser influences with different COVs. As previously described, the COV of $J$ varies from 0.01 to 0.03 while the COVs of other parameters remain the same. It can be seen from Fig. 9 that for each laser influence the COV of $J$ significantly affects the IQR of the output parameters. With the increase in the input value of COV from 0.01 to 0.03, the IQR of $s$, $u_s$, $T_{I,I}$, and $T_e$ show significant increase. For example, the IQR of $s$ increases from 1.575 nm to 5.25 nm, 10 m/s to 23 m/s, 398 K to 790 K, and 44 K to 115 K, respectively, for $J = 0.4 \, \text{J/cm}^2$. That means, the larger the COV is,
the more the IQR increases. Figure 10 represents the IQRs of $s$, $u$, $T_{l,i}$, and $T_e$ at different electron–lattice coupling factors ($G_{RT}$) with different COVs. Three values of $G_{RT}$, $2.1 \times 10^{16}$, $2.2 \times 10^{16}$, and $2.3 \times 10^{16}$ W/cm$^2$K are considered with the COV ranging from 0.01 to 0.03, and the COVs of the other parameter remain the same. It is shown in Fig. 10 that for each $G_{RT}$, its COV significantly affects in the IQR of the output parameters. With the increase in the input value of COV from 0.01 to 0.03, the IQR of interfacial location, interfacial velocity, temperature, and electron temperature (K) increase from 1.58 nm to 2.85 nm, 8.5 m/s to 18 m/s, 305 K to 540 K, and 37 to 79 K, respectively, with $G_{RT} = 2.3 \times 10^{16}$ W/m$^2$K. The IQRs of $s$, $u$, $T_{l,i}$, and $T_e$ increase as the COV increases. The reason is that with the increase in the electron-phonon coupling factor, the hot electron heated up faster the metal lattice, leading to a more severe superheating process. Figures 9 and 10 indicate that the interfacial location, velocity, temperature, and electron temperature greatly depends on the energy of laser and phonon-electron coupling factor.

5 Conclusion

The sample-based stochastic model was applied to analyze the influence of parametric uncertainty on melting and resolidification of gold film subjected to nano- to femtosecond laser irradiation. This approach produces reasonable results with minimum number of combination of the input parameters to use with a stochastic convergence analysis. Rapid solid–liquid phase change was modeled using a two-temperature model with an interfacial tracking method. Temperature dependent electron heat capacity, thermal conductivity, and electron–lattice coupling factor were considered. The uncertainties of laser pulse fluence, pulse duration, electron–lattice coupling factor, and electron thermal conductivity on the results of solid–liquid interface temperature, interfacial velocity and location, and electron temperature were studied. The results show that the mean value and the standard deviation of laser influence and electron–lattice coupling factor have dominant effects on rapid phase change.

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Nomenclature

$C_p$ = heat capacity (J/kg K)
$f$ = liquid factor
\[ G = \text{electron–lattice coupling factor (W/m}^3\text{K)} \]
\[ G_{RT} = \text{electron–lattice coupling factor at room temperature (W/m}^3\text{K)} \]
\[ h_{\text{lu}} = \text{latent heat of fusion (J/kg)} \]
\[ J = \text{laser influence (J/cm}^2\text{)} \]
\[ K = \text{thermal conductivity (W/mK)} \]
\[ P_{25} = \text{25th percentile} \]
\[ P_{75} = \text{75th percentile} \]
\[ R = \text{reflectivity} \]
\[ R_g = \text{gas constant (J/kg K)} \]
\[ S = \text{laser source term} \]
\[ t_p = \text{laser pulse (s)} \]
\[ T_e = \text{electron temperature (K)} \]
\[ T_{ll} = \text{interfacial lattice temperature (K)} \]
\[ u_s = \text{interfacial velocity (m/s)} \]
\[ \delta = \text{optical penetration depth (m)} \]
\[ \delta_b = \text{ballistic range (nm)} \]
\[ \eta = \text{thermal conductivity constant} \]
\[ \lambda = \text{thermal conductivity constant (W/mK)} \]
\[ \rho = \text{density (kg/m}^3\text{)} \]

**Subscripts**

- \( e \): electron
- \( l \): liquid
- \( m \): melting
- \( s \): solid

**References**


