Ultrafast melting and resolidification of gold particle irradiated by pico- to femtosecond lasers

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Ultrafast melting and resolidification of a submicron gold particle subject to pico- to femtosecond laser pulse are studied in this paper. The nonequilibrium heat transfer in the electrons and lattice is described using a two-temperature model, and the locations of the solid-liquid interface are determined using an interfacial tracking method. The interfacial velocity, as well as elevated melting temperature and depressed solidification temperature, is obtained by considering the interfacial energy balance and nucleation dynamics. The results showed that the maximum melting depth, peak interfacial temperature, and velocity increase with the decreasing particle size and pulse width or with the increasing laser fluence.

I. INTRODUCTION

Selective laser sintering (SLS) is a rapid manufacturing/ tooling technology that can build functional parts from powdered material via layer-by-layer sintering (for amorphous powder, such as polycarbonate) or melting (for crystalline powder, such as metal) by a directed laser beam. Fabrication of the metal part is very challenging because the temperature required to bind the metal powder particles is much higher than that to bind the amorphous powders particles. The single-component metallic parts can be fabricated by melting the skin of the powder particles and can join together the nonmelted solid cores by resolidifying the liquid layer. The SLS of a single-component metal powder via partial melting is very challenging due to the very small window of the processing parameters. Inappropriate combination of scanning velocity and laser beam intensity may result in nonmelting or complete melting of metal powder particles.

While cw lasers were exclusively used in the SLS by the early researchers, pulsed lasers with pulse width ranging from millisecond to nanosecond have been used to sinter metallic powders. Su et al. investigated the full dense laser sintering of tool steel powder using a pulsed Nd:YAG (yttrium aluminum garnet) laser of 0.5–20 ms and average power up to 550 W. Abe et al. studied the SLS of titanium powders using a YAG laser of 1–5 ms and an average power of 50 W. Morgan et al. performed SLS experiments of gas atomized 316 L stainless steel using a Nd:YAG laser, and the results showed that vaporization recoil force overcame the surface tension acting on the melt and improved the cohesion compared to cw laser. Fischer et al. suggested that for nanosecond laser sintering, only the skin of the powder particle is melted and the core of the particle remains at its initial temperature. The skin temperature was estimated using a one-dimensional (1D) conduction model of a single spherical particle surrounded by a continuum that represents the neighbor particles. In addition, the degree of partial melting and, ultimately, the local porosity of the final product can be easily controlled. Recently, the first author and his students carried out particle level modeling of solid-liquid and liquid-vapor phase changes during nanosecond laser sintering.

It has been considered as a drawback of SLS technique that the fabricated metal parts are always porous and require postprocessing to be fully densified. However, porosity is not always undesirable for some applications such as in wick structure of heat pipes, electrodes of fuel cells, aerospace, and bioengineering. Porous metallic materials, which consist of metal skeleton and beneficial pores, possess useful properties that the corresponding bulk materials do not have. Instead of avoiding the porosity of the product, it is possible to utilize and control the porosity of the final product by controlling the laser pulse width, repetition rate, laser intensity, and scanning velocity. While nanosecond laser can be used to sinter metal particles, application of pico- to femtosecond lasers in the sintering of metal powder particles may allow more accurate control of porosity of the sintered part via controlling the degree of partial melting.

Because the pulse duration of femtosecond lasers is shorter than the relaxation times of most materials, and they have extremely high irradiance, the phenomena associated with femtosecond laser interaction with matter are very different from those associated with longer pulses. Sintering of metallic submicron or nanoparticles with femtosecond lasers has not been investigated. During laser-metal particle interaction, the laser energy is first deposited into electrons on the metal particle surface. The excited electrons move into deeper parts of the metal with velocity close to the Fermi velocity (≈10⁶ m/s) by ballistic motion. Meanwhile, those hot electrons are diffused into deeper part of the electron gas at a speed (<<10⁴ m/s) much lower than that of the ballistic motion. The hot electrons also collide with lattice—referred to as electron-lattice coupling—and transfer energy to the lattice. Nonequilibrium between electrons and lattice can be described by the two-step heating process. A pioneered work by Anisimov et al. modeled heat conduction in the electrons using Fourier’s law (parabolic heat conduction in
electrons) but neglected the heat conduction in the lattice. Qiu and Tien\textsuperscript{17} rigorously derived another two-temperature model from the Boltzmann transport equation. They considered heat conduction in the electrons using a hyperbolic model and, again, neglected heat conduction in the lattice. Chen and Beraun\textsuperscript{18} considered heat conduction in the lattice using a hyperbolic model and proposed a dual-hyperbolic model. Their results showed that the inclusion of lattice conduction resulted in more accurate thermal response than the models without lattice conduction. When laser pulse width is on the order of 100 fs or longer, the hyperbolic effects on both electron and lattice conduction in pure metals can be neglected and the dual-parabolic two-temperature model is reasonably accurate.\textsuperscript{19,20}

Under high laser fluence and/or short pulse, the lattice temperature can exceed the normal melting point before melting takes place. In the resolidification stage, on the other hand, the liquid can be supercooled and the solid-liquid interface can be cooled below the melting point at room temperature. Once phase change is triggered in a superheated solid or undercooled liquid, both of which are thermodynamically metastable states, the solid-liquid interface can move at a very high velocity (on the order of 10 to 10\textsuperscript{3} m/s). The rapid phase change phenomena induced by an ultrashort pulse laser are controlled by nucleation dynamics at the interface rather than by interfacial energy balance.\textsuperscript{21}

Kuo and Qiu\textsuperscript{19} modeled picosecond laser melting of metal film using the dual-parabolic two-temperature model. Chowdhury and Xu\textsuperscript{20} studied the melting and evaporation of a gold film induced by a femtosecond laser. The energy equation for electrons was solved using a semi-implicit Crank–Nicholson scheme, while the energy equation and phase change for the lattice were solved using an explicit enthalpy model in Refs.\textsuperscript{19} and \textsuperscript{20}. While an explicit scheme is easy to apply, a very small time step is required to ensure numerical stability. The development of an implicit scheme for enthalpy model that can outperform the explicit scheme is numerically challenging.\textsuperscript{22} While the temperature transforming model\textsuperscript{23} and the enthalpy linearization model\textsuperscript{24} can be easily discretized using the implicit scheme, they cannot model superheat in solid in the melting stage and undercooling of liquid in the solidification stage because both models assume phase change occurs in a range of temperatures near the melting point at room temperature.

Zhang and Chen\textsuperscript{25} proposed an implicit, fixed grid interfacial tracking method to solve kinetics controlled rapid melting and resolidification during the ultrashort pulse laser interaction with a free-standing metal film. For the case with a simple electron thermal conductivity model and without the electron ballistic motion, the results agreed with those in Ref.\textsuperscript{19}. Zhang and Chen\textsuperscript{26} incorporated the nonlinear electron heat capacity obtained by Jiang and Tsai\textsuperscript{27,28} and a temperature-dependent electron-lattice coupling factor based on a phenomenological model\textsuperscript{29} into the interfacial tracking method. In this paper, ultrafast melting and resolidification of submicron particles irradiated by pico- to femtosecond lasers will be modeled using the interfacial tracking method and the effects of particle size, laser pulse width, and the fluence on ultrafast melting and solidification will be investigated.

II. PHYSICAL MODEL

Melting and resolidification of a submicron particle subjected to temporal Gaussian heat flux from a laser beam will be modeled. The diameter of the metal particle is much smaller than the diameter of the laser beam, which is in turn much smaller than the dimension of the sintered part. Since the laser radiation penetrates the powder bed over a distance of several powder-sphere diameters, it can be assumed that the multiple scattering of radiation leads to a nearly homogeneous distribution of the heat flux within the optically penetrated layer,\textsuperscript{9} which leads to an almost normal incidence of the radiation on the surfaces of the grains in the underlying layers. Figure 1 shows the physical model of melting and resolidification under consideration. A particle with a radius of \( r_p \) (diameter of \( d_p \)) and an initial temperature of \( T_i \) is subjected to a temporal Gaussian laser pulse with a full width at half maximum pulse width of \( t_p \) and fluence of \( J/(J/m^2) \) from the surface \( r=r_i \). Due to the symmetry of the particle, the model can be simplified to 1D in the \( r \)-direction. The origin of time is chosen as the time when the laser pulse is at its maximum.

The energy equations of the free electrons (\( e \)) and lattice (\( l \)) are, respectively,

\[
C_e \frac{\partial T_e}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left[ r^2 k_e \frac{\partial T_e}{\partial r} \right] - G(T_e - T_l) + S, \tag{1}
\]

\[
C_l \frac{\partial T_l}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left[ r^2 k_l \frac{\partial T_l}{\partial r} \right] + G(T_e - T_l), \tag{2}
\]

where \( C \) represents heat capacity, \( T \) is the temperature, \( k \) is the thermal conductivity, 105.1+0.2914 \( C \) is the temperature-dependent electron-lattice coupling factor, and \( S \) is the intensity of the internal heat source. The heat conduction in both electrons and lattice is assumed to be parabolic since the laser pulses considered are 100 fs or longer. Equation (2) is valid in both solid and liquid phases but not at the solid-liquid interface.

The heat capacity of electron is assumed to be proportional to the electron temperature in most existing works,\textsuperscript{12,17,19,25} i.e.,

\[
C_e = B_e T_e, \tag{3}
\]

where \( B_e \) is constant. Equation (3) is valid for \( T_e < 0.1 T_F \), where \( T_F \) is the Fermi temperature.\textsuperscript{30} Jiang and Tsai\textsuperscript{27,28} obtained electron heat capacity using the solid state model but its explicit form was not given. Chen et al.\textsuperscript{31} analyzed the
results of Ref. 27 and suggested that the electron heat capacity can be approximated as

\[
C_e = \begin{cases} \frac{B_e T_e}{T_P} & T_e < T_P/\pi^2 \\
2B_e T_P/3 + C_e'/3, & T_P/\pi^2 \leq T_e < 3T_P/\pi^2 \\
Nk_B + C_e'/3, & 3T_P/\pi^2 \leq T_e < T_F \\
3Nk_B/2, & T_e \geq T_F, \end{cases}
\tag{4}
\]

where

\[
C_e' = B_e T_P/\pi^2 + \frac{3Nk_B/2 - 2B_e T_P/\pi^2}{T_P - T_P/\pi^2} (T_e - T_P/\pi^2),
\tag{5}
\]

where \(N\) is the number density of atoms and \(k_B\) is the Boltzmann constant.

At nonequilibrium condition, thermal conductivity of the electrons depends on the temperatures of both electrons and lattice. For a wide range of electron temperatures ranging from room temperature to Fermi temperature, the electron thermal conductivity can be calculated by\(^{32}\)

\[
k_e = \chi \left( \frac{\partial^2 T_e}{\partial T_e^2} + 0.16 \right) \frac{4(\pi^2 + 0.44)}{(\pi^2 + 0.092)^{3/2}} \frac{\partial T_e}{\partial \theta_e},
\tag{6}
\]

where 37.72 + 0.0717 T_e − 1.721 × 10^−5 T_e^2 + 1.064 × 10^−9 T_e^3 and \(\theta_e = T_e/T_P\) are dimensionless electron and lattice temperatures. The two constants in Eq. (6) are \(\chi = 353\) W/m K and \(\eta = 0.16\) for gold.

Most of the ultrafast laser heating analyses have been carried out with a constant electron-phonon coupling factor \(G\). Due to the significant changes in the electron and lattice temperatures caused by high-power laser heating, \(G\) could be temperature dependent. The phenomenological temperature-dependent \(G\) proposed by Chen et al.\(^{29}\) is adopted in this work,

\[
G = G_{RT} \left[ \frac{A_e}{B_1} (T_e + T_l) + 1 \right]
\tag{7}
\]

where \(G_{RT}\) is the coupling factor at room temperature; \(A_e\) and \(B_1\) are materials constants for electron relaxation time. Since atoms in the liquid phase do not have long-range orders, electrons collide more frequently with liquid atoms than with the atoms in the solid crystals. The coupling factor for liquid phase is enlarged by 20% from the result obtained from Eq. (7) to account for the increased frequency of collisions.\(^{19}\)

The laser heat source term in Eq. (1) can be described by the following equation:

\[
S = 0.94 \frac{1 - R}{t_p(\delta + \delta_b)} \left[ 1 - e^{-r_p/(\delta + \delta_b)} \right] J \left[ \frac{r_o - r}{\delta + \delta_b} \right] 2.77 \left( \frac{t}{t_p} \right)^2,
\tag{8}
\]

where \(R\) is the reflectivity of the thin film, \(t_p\) is the laser pulse duration, \(J\) is the laser pulse fluence, \(\delta\) is the optical penetration depth, \(\delta_b\) is the ballistic range, and \(1 - e^{-r_p/(\delta + \delta_b)}\) is to correct the particle size effect. The ballistic range \(\delta_b\) is introduced because the ballistic motion and diffusion of the hot electrons spread the absorbed laser energy into much greater depth of electrons.\(^{15}\) While there is no general guideline on the value of the ballistic range, Wellershoff et al.\(^{33}\) studied the femtosecond laser interaction with gold film (without phase change) and obtained the best match between the predicted and measured electron and lattice temperatures with a ballistic range of \(\delta_b = 105\) nm.

The bulk thermal conductivity of metal measured at equilibrium \(k_{eq}\) is the sum of the electron thermal conductivity \(k_e\) and the lattice thermal conductivity \(k_l\). For gold, the former is 99% of \(k_{eq}\) while the latter only contributes to 1% of \(k_{eq}\). The lattice thermal conductivity is therefore taken as 1% of the thermal conductivity of bulk metal since the mechanism of heat conduction in metal is mainly by electrons, i.e.,

\[
k_l = 0.01 k_{eq}.
\tag{9}
\]

The energy balance at the solid-liquid interface is described by\(^{35}\)

\[
k_{ls} \frac{\partial T_{ls}}{\partial r} - k_{l,t} \frac{\partial T_{l,t}}{\partial r} = \rho_i h_m \frac{dr_i}{dt},
\tag{10}
\]

where \(T_{ls}\) and \(T_{l,t}\) are the solid and liquid lattice temperatures respectively, \(\rho\) is the mass density, \(h_m\) is the latent heat of fusion, and \(r_i\) is the radius of the solid-liquid interface. Alternatively, the location of the solid-liquid interface can also be measured from the surface of the particle as the thickness of the liquid layer.

The additional interfacial velocity due to the density change during the melting and resolidification has been considered.

The velocity of the interface \(u_s\) is dominated by nucleation dynamics, instead of energy balance [Eq. (10)]. For short-pulsed laser melting of gold, the velocity of the solid-liquid interface is described by\(^{19}\)

\[
u_s = \frac{V_0}{R_e} \left[ 1 - \exp \left( -\frac{h_m T_{l,t} - T_m}{R_e T_m} \right) \right],
\tag{11}
\]

where \(V_0\) is the maximum interface velocity, \(R_e\) is the gas constant for the metal, and \(\delta_b (\text{nm})\) is the interfacial temperature. The interfacial temperature \(T_{l,t}\) is higher than the melting point \(T_m\) during melting and lower than the melting point during solidification.

The time \(t=0\) is defined as the time when the peak of a laser pulse reaches the film surface. Therefore, the initial conditions of the problem are

\[
T_e(x, -2t_p) = T_i(x, -2t_p) = T_i.
\tag{12}
\]

The boundary conditions of the problem can be specified by assuming that the heat loss from the film surface can be neglected, i.e.,

\[
\left. \frac{\partial T_x}{\partial r} \right|_{r=r_o} = \left. \frac{\partial T_x}{\partial r} \right|_{r=r_o} = \left. \frac{\partial T_l}{\partial r} \right|_{r=r_o} = 0.
\tag{13}
\]
The volumetric enthalpy can be expressed as

\[ H_v = \int_0^{T_{fl}} C_{vl}(T) dT + f \rho \varepsilon h_m, \]  

where the first term is the enthalpy of solid phase at the interfacial temperature, and the second term is the latent heat due to partial melting. Substituting Eq. (15) into Eq. (14), one obtains

\[ C_{vl}(T_{fl}) \frac{dT_{fl}}{dt} + \rho \varepsilon h_m \frac{df}{dt} = \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 k_t \frac{\partial T_t}{\partial r} \right) + G(T_e - T_t). \]  

The liquid fraction \( f \) is related to the location of the solid-liquid interface by (see Fig. 2)

\[ \frac{df}{dt} = \frac{1}{(\Delta r)_p} \frac{ds}{dt} = \frac{u_s}{(\Delta r)_p}, \]  

where \((\Delta r)_p\) is the size of the control volume \( P \) and \( s \) is the thickness of the liquid layer.

Substituting Eq. (17) into Eq. (16) and integrating the resulting equation in the control volume that contains the solid-liquid interface, the interfacial velocity is obtained as

\[ u_s = \frac{1}{\rho \varepsilon h_m} \left[ 1 \frac{k_{lw} r_w^2}{(\Delta r)_w} (T_{lw} - T_{fl}) + \frac{k_{le} r_e^2}{(\Delta r)_e} (T_{le} - 0.5 f_P (\Delta r)_P) + G_P (T_{e,P} - T_{fl}) (\Delta r)_P \right] \frac{\Delta r}{(\Delta r)_P}, \]

where \( k_{lw} \) and \( k_{le} \) are the lattice thermal conductivities at

<table>
<thead>
<tr>
<th>Properties</th>
<th>Solid (s)</th>
<th>Liquid (l)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Coefficient for electron heat capacity, ( B_e ) (J/m³ K)</td>
<td>70⁰</td>
<td>1.2 \times 10^{-7} \text{b}</td>
</tr>
<tr>
<td>Material constant, ( A_e ) (K^{-2} s^{-1})</td>
<td>1.2 \times 10^{11} \text{b}</td>
<td>19.30 \times 10^{3}</td>
</tr>
<tr>
<td>Material constant, ( B_l ) (K^{-2} s^{-1})</td>
<td>5.05</td>
<td>5.17 \times 10^{4}</td>
</tr>
<tr>
<td>Electron-lattice coupling factor at room temperature, ( G_{ref} ) (W/m³ K)</td>
<td>2.2 \times 10^{16} \text{b}</td>
<td>320.973 - 0.01117 x - 2.747 x 10^{-3} T_f^2 - 4.048 x 10^{-3} T_f^3</td>
</tr>
<tr>
<td>Density, ( \rho ) (kg/m³)</td>
<td>19.30 \times 10^{3}</td>
<td>37.72 + 0.07117 x - 1.721 x 10^{-3} T_f^2 + 1.604 x 10^{-3} T_f^3 \text{c}</td>
</tr>
<tr>
<td>Specific heat, ( c_p ) (J/kg K)</td>
<td>105.1 + 0.29147 x - 8.713 \times 10^{-3} T_f^2 + 1.187 \times 10^{-5} T_f^3 - 7.051 x 10^{-3} T_f^4</td>
<td>163.205⁰</td>
</tr>
<tr>
<td>Thermal conductivity at equilibrium, ( k_{eq} ) (W/m K)</td>
<td>105⁰</td>
<td>20.6⁰</td>
</tr>
<tr>
<td>Reflectivity, ( R )</td>
<td>6.42 \times 10^{-3}</td>
<td>6.42 \times 10^{-3}</td>
</tr>
<tr>
<td>Optical penetration depth, ( \delta ) (nm)</td>
<td>20.6⁰</td>
<td>6.373 \times 10^{4}</td>
</tr>
<tr>
<td>Ballistic range, ( \delta_b ) (nm)</td>
<td>105⁰</td>
<td>1336</td>
</tr>
<tr>
<td>Melting point, ( T_m ) (K)</td>
<td>105⁰</td>
<td>1336</td>
</tr>
<tr>
<td>Fermi temperature, ( T_F ) (K)</td>
<td>6.42 \times 10^{4}</td>
<td>6.42 \times 10^{4}</td>
</tr>
<tr>
<td>Latent heat of fusion, ( h_m ) (J/kg)</td>
<td>6.373 \times 10^{4}</td>
<td>6.373 \times 10^{4}</td>
</tr>
<tr>
<td>Limit velocity, ( V_o ) (m/s)</td>
<td>1300⁰</td>
<td>1300⁰</td>
</tr>
<tr>
<td>Gas constant for gold, ( R_g ) (J/kg K)</td>
<td>42.21</td>
<td>42.21</td>
</tr>
</tbody>
</table>

\text{aReference 16.} \quad \text{bReference 29.} \quad \text{cReference 9.} \quad \text{dReference 20.} \quad \text{eReference 19.} \quad \text{fReference 34.}
faces $w$ and $e$ of control volume, respectively, and $T_{IL}^0$ is the solid-liquid interfacial temperature at the previous time step. The third and fourth terms in the bracket at the right-hand side of Eq. (18) represent the effects of the electron-lattice interaction and change of the interfacial temperature in the solid-liquid interfacial temperature. Equation (18) will be used together with Eq. (11) to determine the solid-liquid phase interfacial velocity and temperature. The interfacial location at each step can be determined once the converged interfacial velocity is obtained.

The numerical solution starts from time $t = -2t_p$, with the initial temperatures of electrons and lattice given by Eq. (12). Before onset of melting, the electron and lattice temperatures are obtained by solving discretized equations of Eqs. (1) and (2) simultaneously. Once the lattice temperature at the first control volume from the surface exceeds the melting point, the lattice temperature of the first control volume sets at the melting point and melting will be modeled. Since rapid melting and resolidification are controlled by nucleation dynamics, the interfacial temperature $T_{IL}$ is unknown and it is related to the interfacial velocity in Eq. (11). After melting is initiated, an iterative procedure is employed to solve for the interfacial temperature and the interfacial location at each time step.\textsuperscript{25,26}

IV. RESULTS AND DISCUSSIONS

Ultrafast laser melting and resolidification of pure gold submicron particles with different sizes are simulated first to investigate the size effect on the ultrafast phase change. The thermophysical and optical properties for gold employed in this study are shown in Table I. The thermophysical properties of the gold in Table I are for solid below melting point and liquid above melting point only. The properties for the

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**FIG. 3.** Effects of particle size on surface electron and lattice temperatures ($J=0.3$ J/cm$^2$ and $t_p=20$ ps).

**FIG. 4.** Effects of particle size on the interfacial locations, temperatures, and velocities ($J=0.3$ J/cm$^2$ and $t_p=20$ ps).
superheated solid and undercooled liquid are taken as those at the melting point because of lack of properties for these nonequilibrium states.

Figure 3 shows comparison of the electron and surface lattice temperatures for a 1 μm (diameter) gold particle under irradiation of a laser pulse with fluence of $J = 0.3 \text{ J/cm}^2$ and a pulse width of $t_p = 20 \text{ ps}$. The total grid number is 2502, and the time step is set at $\Delta t = 0.2 \text{ ps}$. One of the advantages of the present numerical algorithm is that it is unconditionally stable for any time step. The peak electron temperatures for the three particle sizes are 6336, 6415, and 6509 K occurring at $t = 4$, $6$, and 10 ps, respectively. The peak lattice temperatures at the surface of the particles with three different sizes are 1666, 1689, and 1724 K, respectively, and all occurred at $t = 26 \text{ ps}$. It is apparent that severe nonequilibrium between electron and lattice exists during the interaction between a picosecond laser and a particle. The peaks for electron temperatures are reached first, and the peaks for lattice are reached at a latter time; i.e., there is a delay between the response of electron and the lattice temperatures.

Figure 4 shows comparison of interfacial locations, temperatures, and velocities for gold particles with different sizes. The maximum melting depths for particles at three different diameters are 21.98 nm (occurring at $t = 292 \text{ ps}$), 25.31 nm (occurring at $t = 344 \text{ ps}$), and 31.89 nm (occurring at $t = 478 \text{ ps}$), respectively. Therefore, the maximum melting depth increases with decreasing particle size, and the time at which the maximum melting depth occurred delays as particle size decreases. The maximum interfacial temperatures in the melting stage for the three particles are 1609, 1625, and 1650 K, respectively. Thus, the maximum interfacial...
temperature increases with decreasing particle size; i.e., the departure of interfacial temperature from the melting point at room temperature increases with decreasing particle size. On the contrary, the minimum interfacial temperatures during solidification stages become closer to the melting point at room temperature as the particle size decreases. The maximum interfacial velocity for the three particle sizes are 227, 237, and 251 m/s, respectively. This trend is consistent with the increasing interfacial temperature with decreasing particle size.

Figure 5 shows the effects of laser pulse width on the electron and lattice temperatures at the particle surface for a 1 µm gold particle when the laser fluence is held at $J = 0.3 \text{ J/cm}^2$. The grid number is kept at 2502, while the time step varies from 1 fs to 2 ps depending on the pulse widths. For the three different pulse widths studied, the peak electron surface temperatures are 6355, 11391, and 16983 K, respectively. With the same laser fluence of $J = 0.3 \text{ J/cm}^2$, the total laser energy delivered to the gold particle is the same for all cases. However, the concentration of laser energy into short pulse duration results in dramatic increase in the electron temperature. The peak lattice temperatures at the surface for the three different pulse widths are 1666, 1836, and 1953 K, respectively. It can be seen that the surface lattice temperature is much lower than the electron temperature, especially for shorter pulse width. As the pulse width decreases, there is longer delay from the time at which peak electron temperature occurs to the time at which peak lattice temperature occurs. Figure 6 shows the interfacial locations, temperatures, and velocities for the three pulse widths studied. With decreasing pulse width, both the maximum melting
depth and duration of phase change increase. The maximum melting depths for the three pulse widths are 21.98, 32.51, and 38.99 nm, respectively. The maximum interfacial temperatures in the melting stage are 1609, 1749, and 1836 K, respectively, and the corresponding peak interfacial velocities are 227, 304, and 345 m/s. Thus, larger melting depth and higher interfacial velocity can be achieved by concentrating the same amount of laser energy into short pulse duration.

Figure 7 shows the electron and lattice temperatures at particle surface for different laser fluences, while the laser pulse width is kept at $t_p = 20$ ps. The peak electron temperatures at the particle surface for the three different laser fluences are 6355, 6761, and 7158 K, respectively. The time at which the electron temperature reaches the peak is 6 ps for all three cases. The peak lattice temperatures for the three cases are 1666, 1801, and 1946 K, respectively; all of them occur at the same time at $t=26$ ps. Figure 8 shows the effects of laser fluence on the interfacial locations, temperatures, and velocities. The maximum melting depths for the three laser fluences are 21.98, 34.93, and 50.10 nm, respectively. Both the time at which the peak melting depth occurs and the duration of phase change increase with increasing laser fluence. The peak interfacial temperature during melting stage for the three laser fluences are 1609, 1706, and 1810 K, respectively; the corresponding peak interfacial velocities are 227, 282, and 333 m/s.

Figure 9 shows the electron and lattice temperatures at the particle surface with different laser fluences, while the laser pulse width is kept at $t_p = 100$ fs. The three laser fluences used here are the same as those used in Figs. 7 and 8. The peak electron temperatures at the particle surface are
16 983, 17 445, and 18 479 K, respectively. The peak lattice temperatures for the three cases are, respectively, 1953, 2122, and 2295 K. Compared with the results shown in Fig. 7 that has the same laser fluences, it is apparent that the peak electron and lattice temperatures with \( t_p = 100 \) fs are much higher than those at \( t_p = 20 \) ps. Effects of laser fluence on the interfacial locations, temperatures, and velocity with \( t_p = 100 \) fs are shown in Fig. 10. The maximum melting depth for the three cases are 38.99, 53.64, and 69.03 nm, respectively, which are much higher than those for the case of \( t_p = 20 \) ps shown in Fig. 8(a). The peak interfacial temperatures during melting stage for the three cases are 1839, 1968, and 2080 K, respectively, and the corresponding interfacial velocities are 346, 396, and 432 m/s. Compared with the case of the same laser fluence but longer pulse width shown in Figs. 8(b) and 8(c), both interfacial temperature and velocity for femtosecond laser are significantly higher.

V. CONCLUSIONS

Melting and resolidification of a gold particle irradiated by pico- to femtosecond laser pulses are investigated numerically. The nonequilibrium heat transfer in electrons and lattice is described using a two-temperature model, and the locations of the solid-liquid interface are determined using an interfacial tracking method. The results showed that severe nonequilibrium between electron and lattice exists during the ultrafast melting and resolidification of a gold particle induced by a pico- to femtosecond laser heating. The peak for electron temperatures are reached first, and the peak for lattice are reached at a latter time; i.e., there is a delay between the response of electron and the lattice temperatures. The maximum melting depth increases with decreasing particle size, and the time at which the maximum melting depth occurred delays as particle size decreases. The maximum interfacial temperature increases with decreasing particle size; i.e., the departure of interfacial temperature from the melting point at room temperature increases with decreasing particle size. Under the same laser fluence of \( J = 0.3 \) J/cm², decreasing laser pulse width from 20 ps to 100 fs causes dramatic increase in the peak electron temperature at particle surface from 6355 to 16 983 K and increase in the peak lattice surface temperature of particles from 1666 to 1953 K. The maximum melting depth, peak interfacial temperature, and velocity increase with decreasing pulse width or with increasing laser fluence.

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