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Temperature of spatially modulated surface of solid film heated by repetitive laser pulses

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Abstract
An analytical solution is obtained for the classical heat conduction problem for a solid film with a surface that is simply deformed and irradiated by repetitive laser pulses. Convective heat losses from the surface and the film–substrate interface are taken into account. The solution, evaluated for large times, shows that the surface deformation increases the average surface temperature. This increase is pronounced when the absorption length of the laser radiation is less than the film thickness (even in the case when the amplitude of surface deformation is much less than its wavelength). At any point on a deformed surface the temporal dependence of temperature is characterized by narrow spikes, which are repeated with the period of pulse repetition. The spatial dependence of surface temperature follows surface deformation.

1. Introduction

In this paper we study classical heat conduction in solid films heated by repetitive laser pulses. This problem is of practical importance for surface modification technologies, since laser–surface interaction may control the structure and properties of thin films and their surfaces. Strong laser radiation induces structural and morphological changes in matter which are responsible, for example, for the degradation of light emitting devices, laser damage of optical components and non-uniform melting of semiconductor surfaces [1]. Laser annealing and fast recrystallization may lead to special types of structures, and laser assisted thin film deposition processes have been developed [2]. Also, athermal driving of molecular surface diffusion by pulsed laser beams has been a subject of recent experiments [3–6]. Also, methods based on pulsed laser heating have been proposed for measuring thermal properties of materials [7].

There have been a number of theoretical studies of heat conduction under conditions of repetitive pulsed laser heating; see for instance [7–9] and references in the latter paper. These studies demonstrated that such a heating mode gives rise to the quasistationary regime, in which the surface temperature of a thick solid film oscillates about the mean value with the pulse repetition frequency. However, the irradiated surface was treated as planar and undeformed, e.g. the roughness was not accounted for. In a separate line of research, the growth and topography of laser-induced ripple structures was studied [10–13]. The form of spatial temperature profile on a rippled surface is of paramount importance for understanding the formation and development of ripples. These and related studies assume, sometimes implicitly, that the surface temperature is periodic with the period of the ripple. However, experimental validation of this basic premise proved very challenging [13] and, to our knowledge, the question is still open. It is the primary motivation for this paper. We also think that the results obtained may prove useful for problems of pattern formation on semiconductor surfaces; oscillatory driving of surface diffusion by pulsed laser beams has been proposed in [14] as a possible route to control pattern formation.

In this paper we examine the effect of simple, cosine-like surface deformation on surface temperature. By analytically solving the appropriate heat conduction problem, we obtain the

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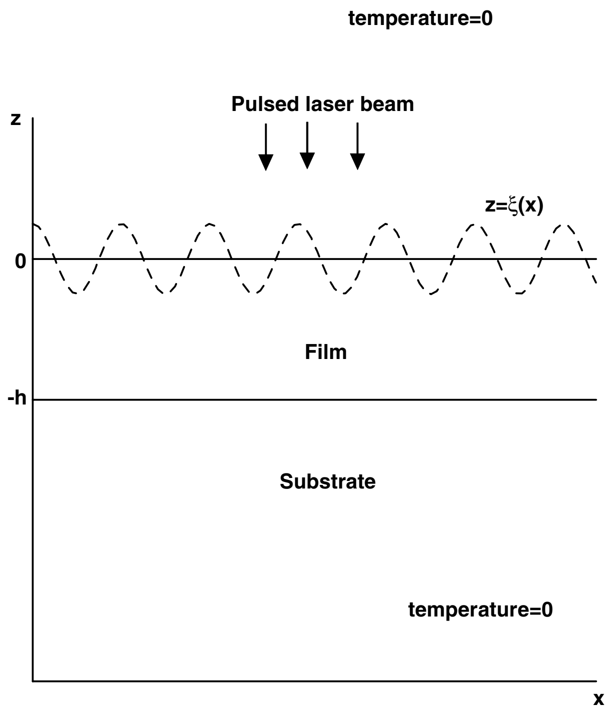

Figure 1. Problem geometry.

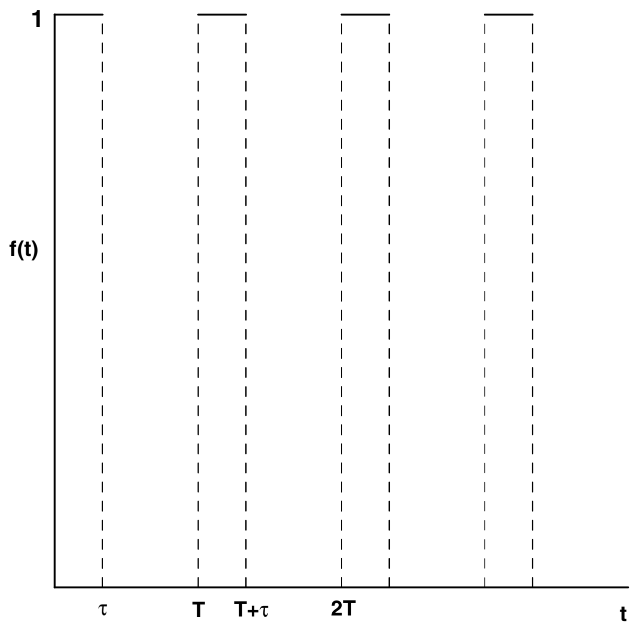

Figure 2. Laser power density profile.

temporal and spatial oscillatory features directly. In particular, we show that surface deformation by itself leads to increased surface temperature and that the spatial temperature profile follows surface deformation. We also discuss applications of our analysis to some rough and/or time-dependent surfaces.

2. Problem formulation and solution

We consider heat conduction in the horizontally infinite film of thickness h, deposited on a thick substrate (figure 1). The film–substrate interface is planar, while the shape of the laser-heated surface of the film is described by the equation $z = \xi(x) = \xi_0 \cos \kappa x$; here ξ_0 is the amplitude and κ the wavenumber. Without loss of generality, the initial (reference) temperature of the film, substrate and the media above the film is taken zero. We assume also that the substrate and the media have high heat capacities and thus their temperature stays zero throughout the film heating process; however, the convective heat losses from the surface and the film–substrate interface are non-negligible.

The Fourier problem of heat conduction is stated as

$$\frac{\partial U}{\partial t} = \chi \nabla^2 U + I \exp(\delta z) f(t), \tag{1}$$

$$U(z, x, 0) = 0, \tag{2}$$

$$z = \xi(x): \quad \nabla U \cdot \vec{n} + \beta_1 U = 0, \tag{3}$$

$$z = -h: \quad \frac{\partial U}{\partial z} - \beta_2 U = 0, \tag{4}$$

where $U(z, x, t)$ is the temperature, χ the thermal diffusivity, δ the absorption coefficient, β_1 and β_2 the heat exchange coefficients, ∇ the two-dimensional gradient operator, $\vec{n}$ the unit outward normal vector to the surface, finally, $I = q_0 \chi \delta (1 - r_f)/\lambda$ (where q_0 is peak laser power density, r_f the reflection coefficient and λ the thermal conductivity) and $f(t)$ is laser power density profile. For a pulsed laser, the approximation by repetitive step function is routinely used. Thus,

$$f(t) = \sum_{k=0}^{\infty} H(t - kT) H(kT + \tau - t), \tag{5}$$

where $H(t - c)$ is Heaviside function, τ the duration of the pulse and T the repetition interval (figure 2).

To nondimensionalize the problem, we take h for unit of length and T for unit of time. The nondimensional initial-boundary value problem (IBVP) is:

$$\frac{\partial U}{\partial t} = P_1 \nabla^2 U + P_2 \exp(\bar{h} z) \bar{f}(t), \tag{6}$$

$$U(z, x, 0) = 0, \tag{7}$$

$$z = \bar{\xi}(x): \quad \nabla U \cdot \vec{n} + B_1 U = 0, \tag{8}$$

$$z = -1: \quad \frac{\partial U}{\partial z} - B_2 U = 0, \tag{9}$$

$$\bar{f}(t) = \sum_{k=0}^{\infty} H(t - k) H(k + \tau_0 - t), \tag{10}$$

where U now stands for nondimensional temperature (defined as dimensional temperature times $\lambda/(hq_0)$), $P_1 = \chi T/h^2$, $P_2 = \chi T \delta (1 - r_f)/h$, $\bar{h} = h\delta$, $\tau_0 = \tau/T$. Also, $B_1 = h\beta_1$ and $B_2 = h\beta_2$ are Biot numbers and $\bar{\xi}(x) = \bar{\xi}_0 \cos \bar{\kappa} x$, $\bar{\xi}_0 = \xi_0/h$, $\bar{\kappa} = \kappa h$.

Next, we assume that surface deformation is small ($\xi_0 \ll 2\pi/\kappa$) and thus the corresponding perturbation of the temperature field is also small. We write $U(z, x, t) = u(z, t) + v(z, x, t)$, $|v(z, x, t)/u(z, t)| \ll 1$, $-1 \leqslant z \leqslant 0$, where $u(z, t)$ is the temperature field in the film with undeformed surface and $v(z, x, t)$ is the temperature field in the film with deformed surface. Now, the problem (6)–(10) splits in two IBVPs, namely:

Problem I.

$$\frac{\partial u}{\partial t} = P_1\frac{\partial^2 u}{\partial z^2} + P_2\exp(\bar{h}z)\bar{f}(t), \tag{11}$$

$$u(z,0) = 0, \tag{12}$$

$$z = 0: \quad \frac{\partial u}{\partial z} + B_1 u = 0, \tag{13}$$

$$z = -1: \quad \frac{\partial u}{\partial z} - B_2 u = 0; \tag{14}$$

Problem II.

$$\frac{\partial v}{\partial t} = P_1\nabla^2 v, \tag{15}$$

$$v(z,x,0) = 0, \tag{16}$$

$$z = 0: \frac{\partial v}{\partial z} + B_1 v = -\left(B_1\left[\frac{\partial u}{\partial z}\right]_{z=0} + \left[\frac{\partial^2 u}{\partial z^2}\right]_{z=0}\right)\bar{\xi}(x) \equiv g(x,t), \tag{17}$$

$$z = -1: \quad \frac{\partial v}{\partial z} - B_2 v = 0. \tag{18}$$

Notice that equation (15) is homogeneous, that the boundary condition in problem II is imposed on the surface $z = 0$ and that it uses the solution of problem I. Derivation of the boundary condition (17) is shown in the appendix A.

Solution of these problems can be found analytically using the method of separation of variables.

The general solution of problem I is given by

$$u(z,t) = u_H(z) + \sum_{n=0}^{\infty} Z_n(z)T_n(t). \tag{19}$$

The first term (solution of the homogeneous problem) vanishes due to the zero initial condition, equation (12), and the homogeneous boundary conditions (13) and (14).

The eigenfunctions of the homogeneous BVP are

$$Z_n(z) = (\lambda_n + B_1^2)^{-1/2}(\sqrt{\lambda_n}\cos\sqrt{\lambda_n}z - B_1\sin\sqrt{\lambda_n}z), \tag{20}$$

where $\lambda_n = \mu_n^2$ and μ_n is the nth nonnegative root of the characteristic equation

$$\cot\mu = \frac{1}{B_1+B_2}\left(\mu - \frac{B_1B_2}{\mu}\right), \tag{21}$$

and

$$T_n(t) = \frac{I_n}{\|Z_n\|^2}\exp(-q_n t)\int_0^t \bar{f}(t')\exp(q_n t')\,\mathrm{d}t', \tag{22}$$

where

$$q_n = P_1\lambda_n, \quad I_n = P_2\int_{-1}^0 \exp(\bar{h}z)Z_n(z)\mathrm{d}z, \qquad \|Z_n\|^2 = \int_{-1}^0 Z_n(z)^2\mathrm{d}z. \tag{23}$$

With $\bar{f}(t)$ given by equation (10), the integration in equation (22) gives

$$T_n(t) = \frac{I_n}{q_n\|Z_n\|^2}\sum_{k=0}^{\infty} H(t-k) - H(t-(k+\tau_0)) - \exp(q_n(k-t))[H(t-k) - \exp(q_n\tau_0)H(t-(k+\tau_0))]. \tag{24}$$

We also have, from equations (19) and (20),

$$\left[\frac{\partial u}{\partial z}\right]_{z=0} = -B_1\sum_{n=0}^{\infty}\frac{\sqrt{\lambda_n}}{\sqrt{\lambda_n + B_1^2}}T_n(t), \qquad \left[\frac{\partial^2 u}{\partial z^2}\right]_{z=0} = -\sum_{n=0}^{\infty}\frac{\lambda_n^{3/2}}{\sqrt{\lambda_n + B_1^2}}T_n(t). \tag{25}$$

The general solution of problem II is given by

$$v(z,x,t) = s(z,x,t) + w(z,x,t), \tag{26}$$

where

$$w(z,x,t) = \frac{1 + B_2(1+z)}{B_1 + B_2 + B_1B_2}g(x,t), \tag{27}$$

(with $g(x,t)$ is the right-hand side of the equation (17)) and $s(z,x,t)$ is the solution of problem III.

Problem III.

$$\frac{\partial s}{\partial t} = P_1\nabla^2 s + r(z,x,t), \tag{28}$$

$$s(z,x,0) = \phi(z,x), \tag{29}$$

$$z = 0: \quad \frac{\partial s}{\partial z} + B_1 s = 0, \tag{30}$$

$$z = -1: \quad \frac{\partial s}{\partial z} - B_2 s = 0. \tag{31}$$

In equations (28)–(31)

$$r(z,x,t) = -\frac{\partial w}{\partial t} + P_1\frac{\partial^2 w}{\partial x^2} = \frac{1+B_2(1+z)}{B_1+B_2+B_1B_2}\left[-\frac{\partial g}{\partial t} + P_1\frac{\partial^2 g}{\partial x^2}\right], \tag{32}$$

$$\phi(z,x) = -w(z,x,0) = -\frac{1+B_2(1+z)}{B_1+B_2+B_1B_2}g(x,0). \tag{33}$$

Since $u(z,0) = 0$ (problem I), $g(x,0) = 0$ and thus $\phi(z,x) \equiv 0$, the initial condition (29) is zero.

The solution of problem III is written as

$$s(z,x,t) = \int_{-1}^0\int_{-\infty}^{\infty}\int_0^t G(M,Q,t-t')r(\zeta,\eta,t')\mathrm{d}t'\mathrm{d}\eta\mathrm{d}\zeta, \tag{34}$$

where

$$G(M,Q,t) = \frac{1}{2\sqrt{\pi\chi t}}\exp\left(-\frac{(x-\eta)^2}{4\chi t}\right)G_1(z,\zeta,t) \tag{35}$$

is Green's function. In equation (35) $G_1(z,\zeta,t)$ is Green's function of the one-dimensional heat conduction equation with mixed boundary conditions at $z = -1$ and $z = 0$. This latter Green's function can be written as

$$G_1(z,\zeta,t) = \sum_{n=0}^{\infty}\exp(-q_n t)\frac{Z_n(z)Z_n(\zeta)}{\|Z_n\|^2}, \tag{36}$$

where $Z_n(z)$ are eigenfunctions of the corresponding Sturm–Liouville problem (equation (20)). Substitution of equations (32), (35) and (36) in equation (34) gives

$$s(z,x,t) = \sum_{n=0}^{\infty} C_n L_n(x,t) Z_n(z), \tag{37}$$

where

$$C_n = \frac{1}{\|Z_n\|^2} \int_{-1}^{0} \frac{1 + B_2(1+z)}{B_1 + B_2 + B_1 B_2} Z_n(\zeta)\mathrm{d}\zeta, \tag{38}$$

$$L_n(x,t) = \int_{-\infty}^{\infty}\int_0^t \frac{1}{2\sqrt{\pi\chi(t-t')}} \exp\left(-\frac{(x-\eta)^2}{4\chi(t-t')}\right) \times \exp(-q_n(t-t'))\left[-\frac{\partial g}{\partial t'} + P_1\frac{\partial^2 g}{\partial \eta^2}\right]\mathrm{d}t'\mathrm{d}\eta, \tag{39}$$

Finally, substitution of equation (24) in equations (25), equations (25) in $g(\eta, t')$, and then substitution of $g(\eta, t')$ in equation (39) and integration, gives

$$L_j(x,t) = \bar{\xi}_0 \cos(\bar{\kappa}x)(1 + P_1\bar{\kappa}^2)(S_1 + S_2), \quad j = 0, \ldots, \infty, \tag{40}$$

where

$$S_1 = -\sum_{n=0}^{\infty} \frac{\sqrt{\lambda_n}}{\sqrt{\lambda_n + B_1^2}}(B_1 + \lambda_n)\frac{I_n}{q_n\|Z_n\|^2} \times \sum_{k=0}^{\mathrm{int}(t-\tau_0)} \frac{q_n}{p_j - q_n}[\exp(q_n(k-t)) - \exp(p_j(k-t))] \times [H(t-k) - \exp(q_n\tau_0)H(t-(k+\tau_0))] + \exp(p_j(k-t))[\exp(q_n\tau_0) - \exp(p_j\tau_0)] \times H(t-(k+\tau_0)), \tag{41}$$

$$S_2 = -\sum_{n=0}^{\infty} \frac{\sqrt{\lambda_n}}{\sqrt{\lambda_n + B_1^2}}(B_1 + \lambda_n)\frac{I_n}{q_n\|Z_n\|^2} \times \sum_{k=0}^{\mathrm{int}(t-\tau_0)} \frac{1}{p_j}[1 - \exp(p_j(k-t))][H(t-k) - H(t-(k+\tau_0))] + \frac{1}{p_j - q_n}[\exp(q_n(k-t)) - \exp(p_j(k-t))][\exp(q_n\tau_0)H(t-(k+\tau_0)) - H(t-k)]. \tag{42}$$

In equations (41) and (42) $p_j = q_j + P_1\bar{\kappa}^2$ and 'int' stands for 'integer part'. Note that $L_n, T_n \to 0$ as $\tau_0 \to 0$ (laser is off), and thus in this limit the film temperature is unchanged, $U(z,x,t) = U(z,x,0) = 0$. Problem III is now solved, and so is problem II.

3. Results

Despite the solution being analytic, its numerical evaluation is difficult and time consuming. This is due to, primarily, the need for the quadruple loop in order to evaluate functions $L_j(x,t)$ (equation (40)) and the small time steps (see below). Moreover, the summations in equations (41) and (42) contain certain combinations of increasing and decreasing exponential functions, making the evaluation prone to roundoff errors. By experimenting with values of nondimensional parameters we discovered that the evaluation is numerically stable for values of τ_0 not to exceed 0.05 in most cases. It must be noted that we are interested, primarily, in the large-times limit; as will be seen, the film heats up significantly at large times even when the pulse duration is small compared with the repetition interval. Note that we are still restricted by relatively large values of τ (see below), for which it is known [15] that Fourier theory gives accurate predictions. The temperature is proportional to the amplitude of surface deformation, $\bar{\xi}_0$ (see equation (40)).

Table 1. Parameters for case I.

Parameter	Value	Units
h	0.1	cm
χ	0.227	$cm^2\ s^{-1}$ [9]
δ	10	cm^{-1}
r_f	0.5	—
β_1	0.1	cm^{-1}
β_2	10^{-3}	cm^{-1}
T	10^{-4}	s
τ	see text	s
P_1	2.27×10^{-3}	—
P_2	5.7×10^{-4}	—
$\bar{h}$	1	—
B_1	0.01	—
B_2	10^{-4}	—
τ_0	see text	—
$\bar{\xi}_0$	10^{-2}	—
$\bar{\kappa}$	2.0	—

Next, we describe two physically important situations corresponding to different absorption lengths. For each situation, we studied several truncations $\sum_{n=0}^{N} a_n$ of the infinite Fourier series. The temperature increased smoothly and insignificantly as N was increased through the interval [25 : 100]. Taking larger N tends to increase temperature values. All results correspond to $N = 50$. The length of the simulation domain in the x-direction equals two wavelengths of the surface modulation. The time step $\Delta t = 10^{-3}$; this is sufficient to accurately resolve the adjacent Heaviside functions $H(t-k)$ and $H(t-(k+\tau_0))$ (equations (41) and (42)) for values of τ_0 used. The characteristic equation (21) is solved by bisection.

The physical and nondimensional parameters for case I are summarized in table 1. In this case the entire film is transparent for laser pulses, since the absorption length $\delta^{-1} = h$.

Figure 3 shows the nondimensional temperature U (that is, one of the film with the deformed surface) along the film thickness for $\tau = 2.5 \times 10^{-6}$ s ($\tau_0 = 0.025$), as a function of time. As expected, at large times the entire film heats up very slowly. With cooling at the exposed surface and at the substrate–film interface, the maximum temperature occurs not at the exposed surface but in the interior of the film; this is in agreement with the result obtained by Blackwell using the Laplace transform method for the heat conduction in the semi-infinite, one-dimensional slab of material [16].

Figure 4 compares the time-dependences of surface temperatures u (that is, one of the film with undeformed surface) and U at the point ($z = 0, x = \pi$). The x-value is chosen arbitrarily; difference of time-dependences at x_1 and x_2 equals $A(\cos x_1 - \cos x_2)$, since at any given

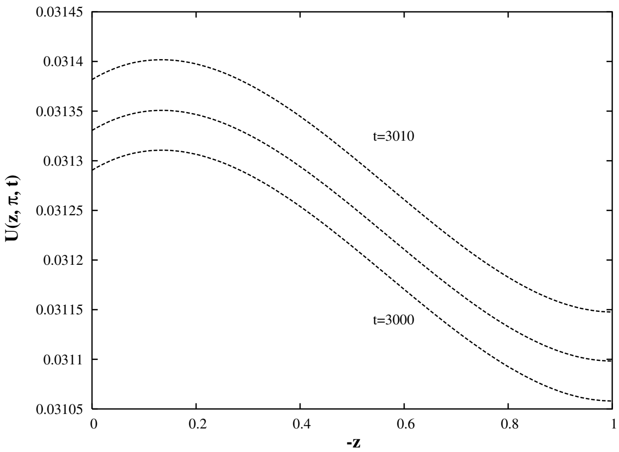

Figure 3. Case I. Bulk temperature profiles at large times.

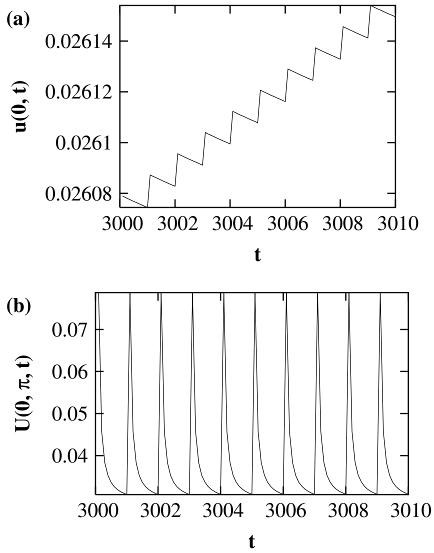

Figure 4. Case I. Surface temperatures versus time. (*a*) Surface is planar. (*b*) Surface is modulated.

time the spatial dependence of temperature is function $A(\cos x + 1)$ (where A is much less than amplitudes of spikes in figure 4(*b*)) due to surface deformation of the cosine form, see equation (40). On the surface the temperatures behave in a quasistationary manner, where the slow growth is modulated by fast oscillations due to laser pulses. The oscillation period equals the pulse repetition period. The surface cosine-like deformation results in increased temperatures. The oscillations are square-shaped when the surface is planar, but spiky when the surface is deformed. For even larger times one expects the slow growth component to vanish and only the oscillation component to remain.

We also performed computations using the value $\tau_0 = 0.05$. The average surface and bulk temperatures increased roughly two-fold compared with the case $\tau_0 = 0.025$, and so did the magnitude of the spike-like oscillations. Also, the spikes appeared more narrow.

Values of the physical parameters for case II are the same as for case I, except $\delta = 6.16 \times 10^5\,\mathrm{cm}^{-1}$ [9] and $\bar{\xi}_0 = 10^{-4}$. The former value results in nondimensional values $\bar{h} = 6.16 \times 10^4$ and $P_2 = 34.9$. The characteristic absorption

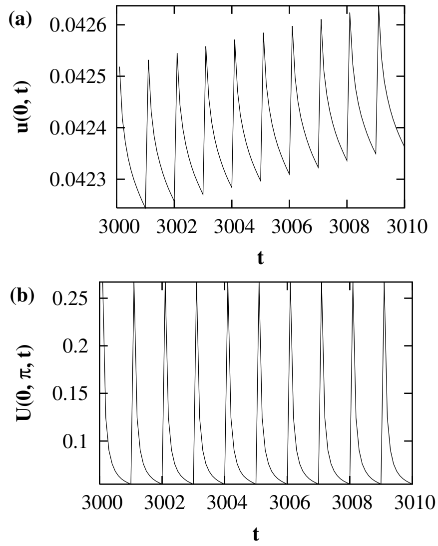

Figure 5. Same as figure 4, but for the case II parameters.

length of laser radiation $\delta^{-1} \ll h$; thus, the bulk of the film does not heat up and the temperature there stays zero.

Figure 5 compares the time-dependences of surface temperatures u and U at the point $(z = 0, x = \pi)$ for case II at $\tau_0 = 0.025$. One notices the same order of magnitude increase of average temperature for the surface deformation amplitude that is two orders less than that for case I. Also, in case I the average temperature increased roughly two-fold due to surface modulation, but in case II the increase is almost four-fold. Comparing figures 4(*b*) and 5(*b*), one can see that the amplitude of spikes is approximately six times larger for case II parameters.

4. Conclusions

We examined heat conduction in the solid film with simply deformed surface irradiated by repetitive laser pulses. We showed that the surface deformation increases the average surface temperature; the more so the less is the absorption length of laser radiation compared with the film thickness. The surface temperature oscillates in time, and at any point on a surface this dependence is characterized by periodically repeated narrow spikes. This period equals the repetition interval of laser pulses. The amplitude of spikes also increases with the decrease of the absorption length. The spatial profile of surface temperature is of same form as the surface deformation.

The periodically rough surface $z = \xi(x)$ can be represented by the truncated Fourier series in cosines. Using the principle of superposition, the solution of the heat conduction problem is then written as $U(z, x, t) = \sum_{i=0}^{N} U_i(z, x, t)$, where each term has a form determined by the wavenumber of the particular cosine function. Also, the temperature field on a surface that evolves on a much larger characteristic time scale than the time scale of a few laser pulses can be considered quasistationary. This is the case for surfaces that evolve by surface diffusion and evaporation–recondensation of atoms or molecules. The shape evolution of such surfaces is often self-similar [17], which could make them particularly convenient for studying the impact of pulsed laser heating on particle motion.

Appendix A. Derivation of the boundary condition (17)

Subscripts in this appendix denote differentiation.

First, write equation (8) in component form and then substitute $U = u + v$:

$$z = \bar{\xi}(x): \quad v_x n_x + u_z n_z + v_z n_z + B_1 u + B_1 v = 0. \qquad \text{(A.1)}$$

This equation can be approximated as follows (since surface deformation is small):

$$z = \bar{\xi}(x): \quad v_x [n_x]_{z=0} + u_z [n_z]_{z=0} + v_z [n_z]_{z=0} + B_1 u + B_1 v = 0. \qquad \text{(A.2)}$$

Then, linearizing u_z and u about $z = 0$, taking into account equation (13) and noticing that $[n_x]_{z=0} = 0$, $[n_z]_{z=0} = 1$, one arrives at equation (17).

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