Effect of the interference in overlapped double-pulse irradiation at the silicon surface

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Abstract
We studied the excitation process of silicon under an intense double pulse. We employed the three-temperature (electron, hole, and lattice) model (3TM) together with Maxwell’s equations. We solved Maxwell’s equations by the finite-difference time-domain approach. The lattice temperature and absorbed energy at the surface increase significantly when the two laser pulses overlap with constructive interference. On the other hand, destructive interference reduces the efficiency of laser excitation significantly. On an average, the overlapped double pulse increases the efficiency to about twice of the distinct two-pulse case.

Keywords: laser processing, three-temperature model, double pulse, FDTD

1 Introduction
In the recent years, we have been able to use intense laser pulses of duration ranging from femto- to pico-second time scales to investigate laser-matter interaction. In particular, laser processing of semiconductors using ultrashort pulses attracts great interest, due to its application in nano-structuring\cite{1,2}. Ultrafast laser pulse enables precise processing of the material, without causing damage in the surrounding area\cite{3}. There are two approaches to study the laser processing, single-shot and multi-shot.

The single-shot experiment is suitable to study excitation process during the laser pulse\cite{4–6}, while the multi-shot experiments can study how to control the efficiency of laser processing\cite{7,8}. In the multi-shot experiment, a pulse-train with distinct laser pulses is employed. Recently, enhancement of laser ablation and processing by an overlapped double-pulse irradiation has been reported\cite{9–11}.

When the two pulses are overlapped, the interference between them plays a crucial role as shown in Fig. 1. If the two pulses with same irradiance ($I_\text{r} \propto E^2$) overlap perfectly, constructive interference makes 4-times intense laser field ($4I_\text{r} \propto (2E)^2$), while the destructive interference makes a zero intensity region\cite{12}. In the case of non-collinear configuration (Fig. 1(a)), position-dependent intensity distribution occurs. The collinear configuration (Fig. 1(b)) makes constructive and destruction pulse after the second half mirror. Since the laser intensity at the target
depends on the relative phase of two pulses, overlapped double-pulse experiments have been done between 4fτ and 0 intensity, we should take into account the effect of interference.

Some theoretical approaches have been proposed as the extension of two-temperature-model (TTM) [13–15]. Recently, we have developed a new numerical model including the time-evolution of electron, hole, and lattice temperatures (3 temperature model=3TM), for the laser excitation process of the silicon [16]. One of the features of 3TM is the direct treatment of electromagnetic field (EM-field) with finite-difference time-domain (FDTD) method. In the previous work, we have reported that the 3TM-FDTD reproduces the pulse-duration dependence of the damage threshold of silicon not only qualitatively but also quantitatively.

This paper is organized as follows: Section 2 describes the analytical formulation of 3TM and numerical method for FDTD method, section 3 presents results of excitation in silicon for different delay time of the two pulses and laser intensity, and then finally, we would like to summarize the work in section 4.

2 Formulation

2.1 3TM

The time-evolution of electron and hole densities, \( n_e \) and \( n_h \) is described as:

\[
\frac{\partial n_{e(h)}(t)}{\partial t} = \frac{\alpha I}{2\hbar \omega_0} + \frac{\beta I^2}{2\hbar \omega_0} - \frac{\gamma_e n_e}{n_e + n_h} n_h - \frac{\gamma_h n_h}{n_e + n_h} n_e + \frac{1}{2}(\theta_e n_e + \theta_h n_h + \theta_e n_h + \theta_h n_e) + \nabla D_{e(h)} \cdot \vec{J}_{e(h)} + D_{e(h)} \nabla \cdot \vec{J}_{e(h)} - (\mu_e n_e + \mu_h n_h + \mu_e n_h + \mu_h n_e) \nabla \cdot \vec{F} + n_{e(h)} \nabla \cdot \vec{F}
\]  

(1)

where \( \omega_0 \) is the laser frequency and \( \alpha \) is the single photon absorption coefficient for transition from VB to CB [17]. \( \beta \) is the two-photon absorption coefficient for which we use the interpolation of the DFT calculation when \( 2\hbar \omega_0 > E_o \) [18], where \( E_o \) is the optical gap, and we employ the model described in Refs. [19, 20] for \( E_o \geq 2\hbar \omega_0 \geq E_g \). \( \gamma_{e(h)} \) is the Auger re-combination coefficient [15] and \( \theta_{e(h)} \) is the impact ionization coefficient [14]. Equation (1) also includes the effect of spatial charge distribution and the associated electric field, and \( J_{e(h)}, D_{e(h)} \) and \( \vec{F} \) are the charge current, diffusion coefficient

\[
J_{e(h)} = \nabla n_{e(h)} + \frac{m_{r,e(h)} n_{e(h)} H_{e(h)}^{-1/2}(\eta_{e(h)})}{k_B T_{e(h)}} \nabla E_g + \frac{n_{e(h)}}{T_{e(h)}} \left( 2H_{e(h)}^{-1/2}(\eta_{e(h)}) H_{0}(\eta_{e(h)}) - \frac{3}{2} \right) \nabla T_{e(h)}
\]

(2)

and the electric field induced by the electron–hole separation [15], respectively. Here \( \eta_{e(h)} \) is the reduced Fermi level:

\[
\eta_e = \frac{\phi_e - E_C}{k_B T_e}
\]

(4)

\[
\eta_h = \frac{E_V - \phi_h}{k_B T_h}
\]

(5)

and

\[
H_{\xi}^\xi(\eta) = \frac{F_{\xi}(\eta)}{F_{\xi}(\eta)}
\]

(6)

where \( F_{\xi}(\eta) \) is the Fermi integral given by

\[
F_{\xi}(\eta) = \frac{1}{\Gamma(\xi + 1)} \int_0^\infty dx \frac{x^\xi}{1 + \exp(x - \eta)}
\]

(7)

\( F_{\xi}(\eta) \) are used depending on whether \( \eta \) is positive [21] or negative [22].

The total dielectric function along with the effect of band structure re-normalization [23] is expressed by

\[
\epsilon(\omega) = 1 + \frac{n_0 - n_e}{n_0} \epsilon_L(\omega + \delta E_g / \hbar) + \epsilon_D(\omega)
\]

(8)

where \( n_0 \) is the density of valence electrons. It should be noted that \( n_e \) and \( n_h \) are nearly the same due to the effect of \( \vec{F} \) and can be approximated at \( n_e \). \( \epsilon_L(\omega) \) is the innate dielectric function, \( \delta E_g \) represents the band re-normalization by carrier density, and \( \epsilon_D(\omega) \) is the complex dielectric function calculated from Drude model. The temperature dependent optical parameters of silicon
are referred to from Ref.[17]. Since this temperature dependence in \( \epsilon_L(\omega) \) does not include band re-normalization, we shift the photon energy by \( \delta E_g \) (Tab. 2). \( \epsilon_{D} \) accounts for the effect of plasma in the excited system. Considering the electron and hole sub-systems,

\[
\epsilon_D(\omega) = -\frac{4\pi n_e e^2}{\omega_0^2} \left[ \frac{1}{m_{e,cd}^*} \left( 1 + i \frac{\omega}{\omega_0} \right) \right] \frac{1}{\omega_0^2} \left[ \frac{1}{m_{h,cd}^*} \left( 1 + i \frac{\omega}{\omega_0} \right) \right] \]
\]

where \( m_{e,h}^{*} \) is the effective mass for the conductivity [24] and \( \nu_e \) and \( \nu_h \) are the collision frequencies, describing the electron-hole (\( e-h \)), electron-phonon (\( e-ph \)) and hole-phonon (\( h-ph \)) collisions. The \( e-ph \) and \( h-ph \) collisions are assumed to have the same frequency which is dependent on lattice temperature [24]. Effect of electron-ion core collisions is also considered [25]. The electron temperature dependence of the electron-ion core collisions is fitted to the damping time data from Sato et al. [25] as \( \tau_{e-ion} = 0.98 + 0.2(k_BT)^{-3.5} \) fs. The collision frequencies for \( e-h \) interactions are calculated as per the model presented in Ref.[26]. The total one-photon absorption coefficient including free-carrier absorption is

\[
\alpha_f = \frac{2\pi}{\lambda} \Im(\epsilon(\omega)),
\]

where \( \lambda \) is the laser wavelength in vacuum.

Since 3TM considers three sub-systems, viz. electron, hole and lattice, their temperatures also evolve separately. The temperature evolution is expressed as:

\[
\frac{C_e \partial T_e}{\partial t} = m_{r,e}(\alpha_f I + \beta I^2)
+ E_g \gamma_n n_e n_h
- \frac{C_e}{\tau}(T_e - T_i) - \nabla \cdot \vec{w}_e
- \frac{\partial n_e}{\partial t} \left( m_{r,e} E_g + \frac{3}{2} k_B T_e H_{1/2}^{1/2} (\eta_e) \right)
- m_{r,e} n_e \left( \frac{\partial E_g}{\partial T_e} \frac{\partial T_e}{\partial t} + \frac{\partial E_g}{\partial n_e} \frac{\partial n_e}{\partial t} \right),
\]

\( C_h \frac{\partial T_h}{\partial t} = m_{r,h}(\alpha_f I + \beta I^2)
+ E_g \gamma_h n_h n_e
- \frac{C_h}{\tau}(T_h - T_i) - \nabla \cdot \vec{w}_h
- \frac{\partial n_h}{\partial t} \left( m_{r,h} E_g + \frac{3}{2} k_B T_h H_{1/2}^{1/2} (\eta_h) \right)
- m_{r,h} n_h \left( \frac{\partial E_g}{\partial T_h} \frac{\partial T_h}{\partial t} + \frac{\partial E_g}{\partial n_h} \frac{\partial n_h}{\partial t} \right),
\]

The third and fourth terms in Eq. (11) account for the loss of energy due to electron-lattice interaction and energy current. The last two terms on right hand side include the changes in carrier density and band gap energy. The heat capacities \( C_{e,h} \) are defined from the internal energy as

\[
C_{e,h} = \frac{\partial U_{e,h}}{\partial T_{e,h}} = \frac{\partial}{\partial T_{e,h}} n_{e,h}(m_{r,e,h} E_g + \frac{1}{2} k_B T_{e,h} H_{1/2}^{3/2} (\eta_{e,h}))
\]

\[
C_e = \frac{3}{2} n_e k_B H_{1/2}^{3/2} (\eta_e)
+ \frac{3}{2} n_e k_B H_{1/2}^{3/2} (\eta_e)
\]

\[
C_h = \frac{3}{2} n_h k_B H_{1/2}^{3/2} (\eta_h)
+ \frac{3}{2} n_h k_B H_{1/2}^{3/2} (\eta_h).
\]

\( \vec{w}_{e,h} \) are the energy currents given by

\[
\vec{w}_e = -\frac{1}{e} (2k_B T_e H_0^1 (\eta_e) + m_{r,e} E_g) \vec{j}_e - \kappa_e \nabla T_e,
\]

and

\[
\vec{w}_h = \frac{1}{e} (2k_B T_h H_0^1 (\eta_h) + m_{r,h} E_g) \vec{j}_h - \kappa_h \nabla T_h.
\]

Here

\[
\vec{j}_e = n_e e^0 H_{1/2}^0 (\eta_e) \{ m_{r,e} \nabla E_g + k_B \nabla T_e + 2k_B H_0^1 (\eta_e) \nabla T_e \}
\]

(18)
and
\[
\vec{j}_h = -n_h \mu_e H_{0/2}^0(\eta_h) \{ m_r h \nabla E_h \\
+ k_B \nabla \eta_h T_h + 2k_B H_{0/2}^0(\eta_h) \nabla T_h \} 
\] (19)
are the electrical current by the Zeeback effect and gradient of the quasi Fermi-levels of electron and hole.

2.2 FDTD

The propagation of the laser pulse is described by solving the Maxwell’s equations using FDTD method. Mur’s absorbing boundary condition is employed to prevent reflection from the boundary [27]. Another salient feature is that the electric field is considered to be complex for the calculation of laser intensity, to ensure a non-zero field at all points in time and space. Assuming a one-dimensional system, the electric field is:

\[
E(x, t) = E_1(x, t) e^{i\omega t} + E_2(x, t) e^{i(\omega t + \phi)} \tag{20}
\]

Here \(E_{1(2)}\) includes Gaussian envelope, and \(\phi\) is the relative phase between the two pulses. The electric field of the two pulses are expressed as

\[
E_1(x, t)|_{x = 0} = E_0 e^{-\frac{(t - T_{\text{peak}})^2}{T_d^2}}, \\
E_2(x, t)|_{x = 0} = E_0 e^{-\frac{(t - T_{\text{peak}} - T_d)^2}{T_d^2}}, \tag{21}
\]
where \(T = t_p/(4\sqrt{\ln 2})\), \(t_p\) being the FWHM pulse duration, \(T_{\text{peak}}\) is the time of peak intensity, and \(T_d\) is the delay time. In this work we assume only two replica pulses. We define the peak of the first pulse (\(T_{\text{peak}}\)) as 4\(T\) to ensure the negligible field intensity at \(t = 0\). The laser irradiance \(I_r(x, t)\) is defined by the absolute value of the electric field as

\[
I_r(x, t) = \frac{c}{8\pi} R[\sqrt{\epsilon}]|E(x, t)|^2. \tag{22}
\]

Evaluation of charge current induced by the laser field is a crucial part of the module. We calculate the current with and without excitation i.e., for photo-absorption and dielectric response. For dielectric response, \(j_0(x, t)\) is calculated as:

\[
j_0(x, t) = \chi_r(\omega) \frac{\partial P(x, t)}{\partial t} = -\chi_r(\omega) \frac{\partial^2 A(x, t)}{c^2 \partial t^2} \tag{23}
\]
where \(A(x, t)\) is the vector potential, \(P\) is the polarization and \(\chi_r\) is the real part of susceptibility \((\chi = (\epsilon - 1)/4\pi)\). The Maxwell’s equation thus, becomes

\[
\frac{1}{c^2} (1 + 4\pi \chi_r(\omega)) \frac{\partial^2 A(x, t)}{\partial t^2} - \frac{\partial^2 A(x, t)}{\partial x^2} = \frac{4\pi}{c} j(x, t) \tag{24}
\]
where

\[
j(x, t) = (\alpha_f(\omega) + \beta(\omega) I_r(x, t)) \frac{c R[\sqrt{\epsilon}]}{4\pi} E(x, t) \tag{25}
\]
is the current associated with photo-absorption.

3 Results and Discussions

The behavior of the EM-field with constructive and destructive interference is the important point of overlapped double pulse. Figure 2 shows the time-evolution of laser field at the Si surface. The irradiance of the single pulse is \(1 \times 10^{12}\) \(\text{W/cm}^2\), the frequency is \(1.55\) eV, and the pulse duration in FWHM is \(100\) fs. Since our approach includes reflection at the surface, the irradiance at the surface is decreased from the incident pulse.

The constructive interference makes a 4-times more intense pulse with \(T_d = 0\) fs, while the destructive interference makes net zero intensity. As \(T_D\) increases, the peak intensity of pulse approaches the intensity of a single pulse. With \(T_d = 200\) fs, we cannot distinguish the difference between \(\phi = 0\) and \(\pi\).

The constructive interference (\(\phi = 0\)) may be important because it increases the efficiency of laser processing. Figure 3 shows the time-evolution of electron, hole, and lattice temperatures at the surface together with the laser irradiance. We can see an abrupt increase of electron and hole temperatures with \(T_d \leq 100\) fs. Above
Table 1 List of symbols and notations used in the text

| Symbol | Description |
|--------|-------------|
| $n_e(h)$ | electron(hole) density |
| $n_0$ | Density of valence electrons |
| $\eta_e(h)$ | Reduced quasi Fermi-level |
| $\mu_e(h)$ | Electron(hole) mobility for Maxwell-Boltzmann distribution |
| $\mu_e(h)$ | Electron(hole) mobility |
| $\kappa_e(h)$ | $\frac{k_B^2 \sigma_e(h) T_e}{\epsilon_e^2} [6H_0^2(\eta_e(h)) - 4H_1^2(\eta_e(h))^2]$: Thermal conductivity |
| $\phi_e(h)$ | quasi Fermi-level |
| $T_e(h)$ | electron(hole) temperature |
| $T_l$ | Lattice temperature |
| $\phi_e(h)$ | Quasi Fermi-level |
| $E_C$ | Lower energy limit of Conduction band |
| $E_V$ | Upper energy limit of Valence band |
| $m_{r,e}(h)$ | $m_e(h)/m_h^*$: Ratio of the effective mass |
| $\sigma_e(h)$ | $-(+)en_e(h)\mu_e(h)H_1^0(\eta_e(h))$: Electrical conductivity |

Table 2 List of parameters

| Symbol | Description |
|--------|-------------|
| $\kappa_l$ | Lattice thermal conductivity [28] |
| $C_l$ | Lattice heat capacity [28] |
| $\tau$ | e-ph relaxation time [29–31] |
| $\tau_0$ | e-ph relaxation time constant [14, 32] |
| $m_e^*$ | Density-of-State effective mass of electron at 300 K [33, 34] |
| $m_h^*$ | Density-of-State effective mass of hole at 300 K [33, 34] |
| $m_{e,cd}^*$ | Effective mass of electron for conduction [24] |
| $m_{h,cd}^*$ | Effective mass of hole conduction [24] |
| $\mu_e^0$ | Electron mobility [34, 35] |
| $\mu_h^0$ | Hole mobility [34, 35] |
| $\gamma_e$ | Auger recombination coefficient [15, 36] |
| $\gamma_h$ | Auger recombination coefficient [15, 36] |
| $\theta_e(h)$ | Impact ionization coefficient [28] |
| $E_g$ | Band gap function |
| $\delta E_g$ | Band re-normalization |

| Value | |
|-------|---|
| $1585T_l^{-1.23}$ | W/(cm K) |
| $1.978 + 3.54 \times 10^{-4}T_l - 3.68T_l^{-2}$ | J/(cm³ K) |
| $\tau_0(1 + (n_e/(8 \times 10^{20}))^2)$ | 240 fs |
| $0.36m_e$ | |
| $0.81m_e$ | |
| $0.26m_e$ | |
| $0.37m_e$ | |
| $8.5 \times 10^{-3}$ | m²/Vs |
| $1.9 \times 10^{-3}$ | m²/Vs |
| $2.3 \times 10^{-31}$ | cm³/s |
| $7.8 \times 10^{-32}$ | cm³/s |
| $3.6 \times 10^{10} \exp(-1.5E_g/k_BT_e(h))s^{-1}$ | |
| $1.16 - 0.72 \times 10^{-4}T_l^2/(T_l + 1108) - 1.5 \times 10^{-8}n_e^{1/3}$ | eV [14] |
| $-1.5 \times 10^{-8}n_e^{1/3}$ | eV |
shows the incident intensity dependence of the 
absorption. In the case of same peak intensity, the 
higher peak intensity induces more efficient photo-
absorption. In the case of same peak intensity, the 
excitation by double pulse is efficient due to the 
$T_l$-dependence in $\alpha$.

Figure 5 shows the incident intensity dependence of (a) $T_l$ at 10 ps and (b) absorbed energy. The intensity-dependence is significant as $T_d$ decreases. At intensity of $1 \times 10^{11} \text{ W/cm}^2$, the excitation is negligible with all $T_d$, while the $T_l$ exceeds melting temperature (1687 K) with $5 \times 10^{11} \text{ W/cm}^2$, and the energy exceeds the bonding energy (2.3 eV/atom) between $7 \times 10^{11}$ and $1 \times 10^{12} \text{ W/cm}^2$ overlapped double-pulse.

4 Summary

In this work we apply the 3TM [16] for silicon to the double-pulse excitation process. We found that the constructive and destructive interference between the two overlapped pulses affect

$T_d = 200$ fs, there is a knee structure after the first pulse. In particular, with $T_d = 400$ fs, we can see a kink at the second pulse in the $T_l$. This "kink" structure in $T_l$ may occur with $T_d$ longer than the typical time scale of electron-phonon energy transfer ($\tau_0 = 240$ fs).

In some cases, the $\phi$ is random. The $\phi$-dependence of the $T_l$-dependence at 10 ps and absorbed energy are shown in Fig. 4. The $\phi$-dependence is significant in $T_d < 200$ fs. The thick-solid line indicates the average. On average, the interference effect increases the $T_l$ to about 1.53 times and absorbed energy to about 2.5 times with respect to distinct two-pulse case ($T_d = 500$ fs).

The $T_d$-dependence for $T_d > 200$ fs shows slow increase as $T_d$. This relatively weak $T_d$-dependence may due to the $T_l$-dependence of the excitation rate. The most intense $T_l$-dependence in our approach is the phonon-assisted one-photon absorption in $\alpha$[17]. The increase in $\alpha$ induces the increase of $T_l$ and energy absorption at the second pulse.

The two horizontal lines indicates the results with different laser parameters which gives same fluence as the case of two distinct pulses. The double-pulse excitation is more efficient than a longer pulse (200 fs), while it is less efficient than an intense pulse ($2 \times 10^{12} \text{ W/cm}^2$). Since our calculation includes the two photon absorption, higher peak intensity induces more efficient photo-absorption. In the case of same peak intensity, the excitation by double pulse is efficient due to the $T_l$-dependence in $\alpha$.

Figure 3 shows the irradiance at the silicon surface with (a) constructive and (b) destructive interference. The irradiance of the incident lasers are $1 \times 10^{12} \text{ W/cm}^2$. 

Fig. 2 Irradiance at the silicon surface with (a) constructive and (b) destructive interference. The irradiance of the incident lasers are $1 \times 10^{12} \text{ W/cm}^2$.

Fig. 3 Time-evolution of electron (red-solid), hole (green-dotted), and lattice (blue-dashed) temperatures with different delay time.
the excitation efficiency significantly. On the average, the overlapped double-pulse increases the efficiency about two times compared to the distinct double-pulse case. Our results indicates that we can enhance and control the excitation of silicon by the overlapped double pulse. Also, our result indicates that the excitation efficiency with two pulses increases depending on the time-delay due to the decrease of the band gap by the lattice temperature.

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