Ultrafast pulse train micromachining

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ABSTRACT

A new micromachining technique using user-defined trains of amplified femtosecond laser pulses is described. In this method, a 2-fold Michelson interferometer is used to split each output pulse of an amplified femtosecond laser system operating at 1 kHz into four different pulses at desired separations ranging from 1 ps to 1 ns. These quadruple pulses are then focused on metal, semiconductor and dielectric samples and the material removal characteristics are noted. The experimental results show that there is a distinct effect of the pulse separation on the machining characteristics. It is observed that, in some cases, use of the quadruple pulses separated by 1 ns provides better material removal than the original pulses separated by 1 ms. The femtosecond laser-material interaction is also modeled for the case of metal samples using the two-temperature model. Numerical simulations that were carried out show that irradiation with quadruple pulses lead to a reduction in the predicted melting threshold fluence, which agrees with the experimental observation.

Keywords: femtosecond, ultrafast, micromachining, pulse-train, ablation

1. INTRODUCTION

In recent years, the availability of reliable solid-state ultrafast laser sources has led to a great deal of research in material processing using femtosecond lasers. Both theoretical and experimental results have been widely reported and several review articles are to be found in the literature.\textsuperscript{1, 2} The interest in using ultrafast lasers for machining is primarily due to the fact that the extremely short time-scales of these laser pulses have interesting effects on the energy transfer to the sample and subsequent material transformation. Specifically, it has been seen that femtosecond lasers can machine a wide variety of materials and that very clean and small structures can be machined.

A significant amount of research has been done in identifying the energy transfer mechanisms during femtosecond irradiation of metals and three distinct regimes have been identified.\textsuperscript{3} The first regime is that of ballistic electron transport during which hot electrons excited by the laser beam move into the metal at very high velocities close to the Fermi velocity. This regime is highly non-equilibrium and the electron temperature is not well-defined during this initial period. Thermal equilibrium among the electrons is reached within the electron thermalization time and the second regime starts. Now the electron density of states (DOS) is represented by the Fermi-Dirac distribution and the electron temperature can be defined. This second regime is characterized by non-equilibrium between the electrons at a higher temperature and the lattice at a lower temperature. The temperature gradient causes the hot electrons to diffuse into the bulk and heat transfer is mainly due to the diffusion of the electrons. In the third and final regime, the lattice and the electrons reach thermal equilibrium and normal thermal diffusion carries the energy into the bulk. From the heat transfer viewpoint, the non-equilibrium between the electrons and the lattice can be described by considering the electrons and the lattice as two separate systems. This two-step approach to heat conduction wherein the laser energy is first absorbed by the electrons and subsequently transferred to the lattice was first reported by Anisimov et al.\textsuperscript{4} Qiu and Tien\textsuperscript{5} solved the Boltzmann equation to rigorously establish a hyperbolic two-step (HTS) radiation heating model for femtosecond laser heat transfer in metals. It was also shown that for cases where the heating time was long compared with the electron relaxation time, the hyperbolic effect could be neglected and the model could be simplified to a parabolic two-step (PTS) model. Both the HTS and PTS models were solved numerically to predict the heating of single and multi-layer thin metal films and the predictions were found to agree well with experiments.\textsuperscript{6}

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On the experimental front, there have been numerous reports in the literature on the use of femtosecond lasers for machining of different materials like metals,\textsuperscript{7} dielectrics,\textsuperscript{1,8,9} and also biological materials.\textsuperscript{10,11,12} Apart from material removal, femtosecond lasers have also been used for changing the structural properties of transparent materials by focusing into the bulk to create waveguides.\textsuperscript{13,14} Manufacture of medically important devices like stents in metals and polymers has also been reported.\textsuperscript{15} Several pump-probe experiments aimed at understanding the mechanism of femtosecond laser ablation have also been carried out.

Recently some experiments have been carried out using pulse-trains of high repetition rate and interesting machining effects have been reported. Herman et al.\textsuperscript{16} used a train of 430 pulses of 1.2 ps duration for machining fused silica samples and found that the surface microcracking or swelling that occurred with single pulse machining could be eliminated. It was observed that the heating effects associated with the high repetition rate may have improved the ductility of the surrounding glass. Subsequently, Lapczyna et al.\textsuperscript{17} used a similar pulse train to drill holes in aluminum foil and reported clean through-holes. Double pulse machining of silicon was also reported recently wherein it was shown that ablation was enhanced at a delay of 10 ps between the two pulses.\textsuperscript{18} As such, machining with a pulse train instead of a single pulse could possibly offer certain advantages, combining the positive aspects of both short and long-pulse interactions. In the present work, machining with a train of four pulses obtained by splitting a single pulse in a Michelson interferometer is described. Section 2 of this paper shows numerical modeling work based on the two-temperature theory that predicts the melting threshold fluence as the pulse separation is changed. Section 3 describes the experimental results.

### 2. NUMERICAL MODELING

As described in the previous section, the two-temperature model can be used to simulate the conduction of heat during femtosecond laser irradiation of a metal. In this section, numerical results obtained by solving these equations are presented. The general two-temperature model is the non-equilibrium hyperbolic model represented by the set of equations:\textsuperscript{5}

\[
C_e(T_e) \frac{\partial T_e}{\partial t} = -\nabla \cdot \bar{Q} - G(T_e - T_l) + S \tag{1}
\]

\[
C_l \frac{\partial T_l}{\partial t} = \nabla [\kappa_l (\nabla T_l)] + G(T_e - T_l) \tag{2}
\]

\[
\tau \frac{\partial \bar{Q}}{\partial t} + \kappa_e \nabla T_e + \bar{Q} = 0 \tag{3}
\]

Eq. 1 is for the electron sub-system and it describes the energy absorption by the electrons directly from the laser source term $S$. This heat is then transferred by diffusion among the electrons and also coupled to the lattice by the coupling factor $G$. The second equation describes the energy transport in the lattice sub-system. Energy is coupled in from the electrons and then slowly diffuses throughout the lattice. The third equation provides for the hyperbolic or wave effect of heat conduction. The term $\tau$ in Eq. 3 represents the electron relaxation time, which is the mean time between electron-electron collisions. The value for this term has been shown to be of the order of 10 fs for gold by Qiu and Tien.\textsuperscript{19} As the pulse-widths considered in this study are an order of magnitude higher, the first term in the third equation is neglected to yield a simpler model. As such, the HTS model equations simplify to the PTS model shown below:

\[
C_e(T_e) \frac{\partial T_e}{\partial t} = \frac{\partial}{\partial x} \left( \kappa_e \frac{\partial T_e}{\partial x} \right) - G(T_e - T_l) + S \tag{4}
\]

\[
C_l \frac{\partial T_l}{\partial t} = \frac{\partial}{\partial x} \left( \kappa_l \frac{\partial T_l}{\partial x} \right) + G(T_e - T_l) \tag{5}
\]

It is to be noted that the equations have been further simplified to consider only one-dimensional heat conduction in the $x$-direction. This simplification is justified as the laser beam diameter is much larger than the heat penetration depth. The source term $S$ is as given below:
In Eq. 6 above, the form of the laser pulse has been assumed to be Gaussian in time. Time $t=0$ is taken to coincide with the peak of the pulse. In this equation, the various parameters are $R$ the sample reflectivity, $J$ the input fluence, $t_p$ the full width at half maximum (FWHM) pulse-width, $d$ the sample thickness, $\delta$ the absorption depth and $\delta_b$ the ballistic depth which provides for the ballistic transport of energy by the hot electrons as described in the previous section. Basically, the ballistic depth is added to the optical absorption depth.

Care has to be taken while considering the values of the other parameters like thermal conductivity and specific heat. The lattice parameters are fairly well-defined from the available literature.\textsuperscript{20, 21} However, the values for the electronic thermal properties need more careful attention as the electrons can reach fairly high temperatures under femtosecond irradiation. Usually, the electronic thermal conductivity is taken to be proportional to the ratio of the electron and the lattice temperatures as:\textsuperscript{19}

$$\kappa_e = \kappa_{\infty} \frac{T_e}{T_l}$$  \hspace{1cm} (7)

Eq. 7 is valid when the electron temperatures are much less than the Fermi temperature $T_F (= \varepsilon_F / k_b)$, where $\varepsilon_F$ is the Fermi energy and $k_b$ the Boltzmann constant. When the electron temperatures become comparable to this quantity, a more general expression given below has to be used:\textsuperscript{22}

$$\kappa_e = \chi \left( \frac{\partial_e^2 + 0.16}{\sqrt{\left( \partial_e^2 + 0.092 \right) \left( \partial_e^2 + \eta \theta_l \right)}} \right)^{25}$$  \hspace{1cm} (8)

where

$$\partial_e = \frac{k_b T_e}{\varepsilon_F}$$  \hspace{1cm} (9)

and

$$\theta_l = \frac{k_b T_l}{\varepsilon_F}$$  \hspace{1cm} (10)

Eq. 8 reduces to Eq. 7 when $T_e \ll T_F$. In the case of very high temperatures ($T_e > T_F$), the equation goes to the form $\kappa_e \sim T_e^{5/2}$ which is characteristic of low-density plasma.

The electronic specific heat is taken to be proportional to the electron temperature as:\textsuperscript{19}

$$C_e(T_e) = B_e T_e$$  \hspace{1cm} (11)

The simulation is started at time $t = -2t_p$. So the initial electron and the lattice temperatures are taken to be equal to the room temperature $T_0$ at that time, that is,

$$T_e(x, -2t_p) = T_l(x, -2t_p) = T_0$$  \hspace{1cm} (12)

The top and bottom surfaces of the target are assumed to be insulated, leading to the boundary conditions:

$$\frac{\partial T_e}{\partial x} \bigg|_{x=0} = \frac{\partial T_l}{\partial x} \bigg|_{x=0} = \frac{\partial T_e}{\partial x} \bigg|_{x=d} = \frac{\partial T_l}{\partial x} \bigg|_{x=d} = 0$$  \hspace{1cm} (13)

The governing equations 4 and 5 are then solved using the finite difference method. The electron temperature field is solved for using the semi-implicit Crank-Nicholson scheme. These numbers are then used to calculate how much energy is coupled to the lattice and the lattice temperature field is calculated using an explicit scheme. These values are fed back to calculate new electron temperatures and the process is continued until the errors in the electron and the lattice temperatures at each iteration are below certain tolerance levels. At this point, the calculation proceeds to the next time step. A detailed description of the numerical methods used is given in Chowdhury and Xu.\textsuperscript{23}
The calculations were carried out for a gold sample and the predicted melting threshold fluences are plotted in Fig. 1. The predictions are compared with experimental data published by Wellershoff et al. Two sets of results are plotted in the figure - one in which the ballistic depth $\delta_b$ is taken to be 200 nm and another in which the ballistic effect is neglected completely. It is seen that if the ballistic effect is not considered, the predicted melting threshold fluence is much smaller than the experimentally determined value. This is because, in the latter case, the incident laser energy is absorbed only in the absorption depth $\delta$ and hence leads to a higher energy density in the top part of the film that translates into higher temperatures. On the other hand, consideration of the ballistic depth leads to the incident energy being absorbed over a greater depth that gives a lower temperature and hence higher threshold fluence. The ballistic depth of 200 nm that is considered here is reasonably consistent with previous measurements of the depth that gave a value of 100 nm for much lower fluence pulses.

The inclusion of the ballistic effect gives a reasonably good fit to the experimental data. It is seen that the threshold fluence saturates at about 111 mJ/cm$^2$ for film thickness greater than 900 nm, which is due to the fact that the sample is thick compared with the electronic diffusion range. Also, it is noticed that the simulation over-estimates the fluence for thinner films. This may be due to the fact that multiple reflections that might occur for thinner films are not included in the model. The thermal conductivity of thin metal films has also been shown to be much smaller then the bulk value. Taking this effect into account would lower the predicted damage threshold for the thinner films. Also, it has been shown that the value of the electron-lattice coupling factor might change depending on the electron temperature. This has not been considered in the simulation and might improve model accuracy. Smith and Norris have shown that their numerical solution of the PTS model predicts higher lattice temperatures when the temperature-dependence of the electron-phonon coupling factor is taken into account.

This simulation was used to find out the effect of a pulse-train on the machining characteristics of a gold sample. In particular, the case of a quadruple pulse-train consisting of four equal fluence pulses separated by a certain time interval was considered. The effect on the melting threshold fluence was investigated and the results are plotted in Fig. 2. It is noticed that using a quadruple pulse reduces the average fluence per pulse needed to reach the melting temperature. This is of course expected, as the combination of the four pulses together lead to a higher temperature rise. Also, the fluence needed is seen to increase with increasing pulse separation. This is also as expected because when the separation is less,
the quadruple essentially behaves as a single pulse and the total melting threshold fluence is close to the single pulse threshold of 111 mJ/cm². Essentially, this calculation demonstrated that it is possible to use a combination of lower fluence pulses to cause thermal effects similar to that of a single higher fluence pulse. This effect is then used in experiments to see if there is any advantage in using a pulse-train instead of a single pulse.

3. EXPERIMENTAL RESULTS

The pulse-splitting setup used for the experiments is shown in Fig. 3 below. It is based upon a similar design of an \( n \)-fold Michelson interferometer reported by Siders et al.\textsuperscript{28} In their design, eight delay lines had been implemented to allow the splitting of each input pulse into a maximum of sixteen equally spaced pulses. In our design, only four arms are implemented giving the option to split the input pulse into four pulses. The input pulse gets split into two at the first beam-splitter (BS1). One pulse goes into the first delay arm (Delay 1) while the other is guided into the second beam-splitter (BS2) and further split into two pulses. One of these goes through a fixed arm while the other passes through a movable delay arm (Delay 2). Thus, the spacing between these two pulses can be varied and they are finally made collinear at the third beam-splitter (BS3). The pulse, which went into Delay 1, is also split into two pulses at BS2 which are subsequently made collinear at BS3 also to produce a train of four pulses moving collinearly.

The delay between the pulses can be controlled by the position of the movable mirrors in Delay 1 and 2. During the experiments, Delay 1 was set to twice the value of Delay 2 in order to obtain a train of four equally spaced pulses. The zero delay between the pulses was determined by observing the interference fringes at the path-matched position. In the present design, half of the total energy is discarded in a beam dump at the third beam-splitter so that the beams can be recombined to propagate in the same direction. This energy loss can be avoided by using a thin film polarizer to recombine the pulses.\textsuperscript{28} The only drawback in that case would be that half of the pulses would have a polarization perpendicular to the other half. In the present setup, the polarization stays constant for all the pulses.

![Figure 3: Schematic diagram of the pulse splitting setup](image)

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All the experiments were carried out using the fundamental 800 nm output pulses from a Spectra-Physics Spitfire amplifier seeded with a Tsunami femtosecond oscillator. The machining geometry was very simple - the pulse train was simply focused onto the sample using a gradient index lens having a focal length of 60.0 mm. An iris was used in front of the lens to control the incident energy. A silicon PIN detector along with a counter was used to count the number of pulses incident on the sample. Three different samples were tested - stainless steel, silicon, and glass. After machining, all the samples were examined under an optical microscope and optical micrographs were taken. Also, in some cases, the samples were examined with a Tencor Alphastep stylus profilometer in order to determine the machined profile.

The first set of results is for a 0.5 mm thick stainless steel sample and is shown in Fig. 4 above. The profiles obtained using the stylus profilometer are shown. Four different cases are considered. The first one shows the profile after machining with 1370 single pulses with 12 µJ energy each. The next two profiles are for quadruple pulses that are produced using the pulse-splitting setup. This quadruple pulse has a total energy of 12 µJ and each of the constituent pulses have ~3 µJ each. Two different separations are shown - 1 ps and 1 ns. It is noticed that the quadruple pulses separated by 1 ps (b) and by 1 ns (c) lead to a profile that is not very different from that of the single pulse cases (a) (d). The only difference is in the depth of the machined hole which is slightly smaller in the case of the quadruple pulses.

Fig. 5 shows some results for machining of a 0.635 mm thick silicon wafer. It is seen that there is not much difference between the profiles machined with a single 12 µJ pulse and a quadruple pulse with 1 ns separation. This suggests that the energy distribution in the silicon sample stays constant over this period. An interesting result is seen in Fig. 5(c) which shows that for a single 3 µJ pulse there is no appreciable material removal. This is seen from the optical micrograph in Fig. 5(d) also which shows some surface roughening only. This is evidence that the separation between the pulses affects how the machining proceeds. One possible reason might be a surface modification driven by the first pulse of the quadruple which leads to enhanced absorption of the subsequent pulses.
Results for the machining of a second silicon sample - a 13 µm thick film - are shown in Fig. 6. Optical micrographs of the front and rear surface of the film after irradiation with ~10,000 pulses are shown. The first set is for machining with a train of quadruple pulses having 1 ns separation while the second set is for a single pulse train at 1 kHz. It is noticed that the quadruplet gives a much cleaner entrance and exit hole and smaller taper angle. Machining with ~100,000 pulses was also carried out but showed no appreciable difference.
The final set of results is for machining of a glass microscope cover slide. The machining is done with a train of single pulses having 40 µJ each and also with a train of quadruplets which has ~10 µJ in each pulse. It is noticed from the profiles in Fig. 7 that the material removal seems to be a bit higher for the quadruple case. It was also seen that no damage could be observed when the glass slide was irradiated with single pulses at 1 kHz having 10 µJ energy each. Thus, the accumulated heating of the pulses over a period of several nanoseconds seems to have a significant effect on the machining as compared to single pulses repeated at 1 kHz. It is also possible that an incubation effect as discussed by Lenzner et al.9 and Kautek et al.30 In these papers, it is assumed that the pulses which come first cause a pre-ablation modification of the dielectric material. This leads to enhanced absorption of the subsequent pulses.

CONCLUSIONS

In conclusion, a numerical solution of the two-step heating model for femtosecond irradiation of a metal has been developed and has been used to predict the change in melting threshold fluence for the case of multiple pulses. Ablation experiments carried out with quadruple pulses show that the material removal characteristics can be affected by how the pulses are separated. Especially in the case of dielectrics, it was observed that quadruple pulses could lead to material removal while single pulses of the same fluence as each part of the quadruple could not, pointing to the possibility of an incubation effect.

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