JOURNAL OF OPTOELECTRONICS AND ADVANCED MATERIALS Vol. 12, No. 3, March 2010, p. 674 - 676

Simulation of ultrashort double pulse laser ablation of metals

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We perform a hydrodynamic simulation of laser ablation of metals to explain the results of recent experiments with subpicosecond double pulses (2 × 2 J/cm^2). In the experiments, the time delay τ_{del} between the pulses varies from 0.1 ps to 200 ps. When the delay is much shorter than the electron-ion relaxation time τ_{ei} ($\tau_{\mathrm{delay}} << \tau_{\mathrm{ei}}$) the crater depth is the same as for a single pulse of energy 4 J/cm^2. For the intermediate delays ($\tau_{\mathrm{delay}} \sim \tau_{\mathrm{ei}}$) the crater depth monotonically drops. For the long delay $\tau_{\mathrm{delay}} >> \tau_{\mathrm{ei}}$ the crater depth corresponds to that obtained with a single pulse. The results of the hydrodynamic two-temperature simulations performed with realistic absorption and multiphase equation of state explain the observed behavior.

(Received June 30, 2009; accepted January 8, 2010)

Keywords: Laser ablation, Femtosecond, Double pulse, Hydrodynamic model

1. Introduction

Pulsed laser ablation in combination with the optical emission spectroscopy is an effective method for surface and bulk analysis of different materials. An efficient optimization of this technique when subpicosecond pulses are used can be achieved by using a series of successive pulses. It was demonstrated that the ultrashort laser ablation of metals gives the craters with higher precision in comparison with nanosecond pulses while the intensity of plasma spectral lines is lower than those obtained with nanosecond pulses. This result can be explained by the fact that the laser pulse interacts with the cold condensed matter before the formation of the hot and spreading plasma on the surface of the target. For technical applications, it means that the fast expansion of the ablated matter can result in rapid temperature decay leading to the decrease in the spectral line intensity. To improve the resolution, the double pulse technique was proposed [1, 2]. In these experiments, unexpectedly, the decrease in the crater depth with the increasing delay between pulses was observed. The main objective of this work is to describe the involved physical processes in details in order to define the dominant effect, which results in the reduction of the ablation depth.

2. Model

Double pulse experiments are studied with the aid of the developed hydrodynamic model [3], which describes two-temperature effects, laser energy absorption, electron thermal conductivity, electron-ion coupling and hydrodynamic motion with shock and rarefaction waves and mechanical spallation. Previously, we used the Beer's approximation for the simulation of the laser energy absorption in one-pulse experiments. For the double pulse configuration, this approximation is invalid since the second pulse interacts with hot plasma plume generated by the first pulse. To solve the problem, we use a one-dimensional Helmholtz equation to correctly describe the laser-matter interaction (*s*-polarized pulse).

A wide-range model for permittivity is taken from Ref. 4. Models used for thermal conductivity and electron-ion collisions are similar to these described in Ref. [5].

For completeness of the model we apply a semi-empirical multi-phase equation of state (EOS) accounting for melting and evaporation effects. The free energy is used as a thermodynamic potential and has the form $F(\rho, T) = F_c(\rho) + F_a(\rho, T) + F_e(\rho, T)$, composed of three terms, which describe an elastic part of interaction at $T = 0$ K (F_c) as well as the thermal contribution of atoms (F_a) and electrons (F_e). Here, ρ is the material density, T is the temperature.

3. Results and discussion

Two femtosecond pulses with full width at half maximum (FWHM) of 100 fs and the laser fluence of 2 J/cm^2 (each) interact with a Cu target. The simulation results show that the ablated material consists of both gas and liquid phases. During the first pulse the temperature of electrons rapidly grows up to 10 eV within the skin layer and the lattice temperature starts to increase due to the electron-phonon collisions. Then, lattice melting sets in and the melting front moves into the bulk. The thickness of the melted layer depends on the laser fluence and metal and was of the order of 100 nm in our simulations. After the electron-ion temperature relaxation during the characteristic time τ_{ei} (which of the order of tens picosecond for different metals) the pressure of ions increase up to 40 GPa. The sharp growth of pressure results in formation of intensive shock wave (SW), which also moves into the bulk. At the same time, the rarefaction wave (RW) is formed. After the propagation of the SW, the RW goes through the melted layer and a negative (tensile) pressure can appear. The amplitude of the negative pressure reaches up to -4 GPa in the RW. According to our spallation criteria and recent MD simulations [6], such negative pressure can result in the spallation effect in the liquid phase. Our previous results [7] show also that the main ablation mass originates from the melted layer due to such spallation (or, fragmentation) processes. When the delay between the pulses is small ($\tau_{delay} < 1$ ps) the regime is similar to a single pulse ablation (Fig. 1 for 0 ps delay). With the increasing delay the second pulse interacts with a preheated target material. The efficiency of this interaction depends on the time delay between the pulses. One can see in Fig. 1 a consecutive growth of gas phase with the increase in the delay between pulses, in agreement with the experimental results [2]. On the other hand, the velocity of the ablated melted layer decreases with the delay between the pulses. We attribute this effect to the ambient pressure work, which can slow down the products of ablation generated by the first pulse. It is clearly seen in Fig. 1 for 100 ps delay that the second pulse can totally stop the rarefaction of material in the liquid phase.

To calculate the ablation depth, we performed an integration of the mass flux through the cross section $x = 0$ during and after the pulses using the expression

$$\delta(t) = \frac{1}{\rho_0}\int_0^t (\rho u)|_{x=0}\, dt' .$$

Here ρ is the material density, ρ_0 is the initial material density and u is the material velocity. The dynamics of the crater growth is seen in Fig. 2. One can see that the ablated mass drops with time if the delay between the pulses increases. Already for 10 ps delay the crater depth is close to that obtained for a single pulse (short dashed yellow curve and dashed-doted black curve, respectively). For longer delays (50 and 100 ps) the crater depth is even smaller than that for a single pulse with energy 2 J/cm^2. The second pulse absorption in the nascent plasma plume results in the reheating and acceleration of the external part of the plume. At the same time the deceleration of internal layers of the plume ($\tau_{delay} = 100$ ps) results in formation of the recoil flux, see Fig. 2 for 100 ps curve between 170 and 280 ps. Thus, the main mechanism of the reduction in the ablation depth is the work of pressure in the reheated nascent plum, which suppresses the ablation process initiated by the first pulse.

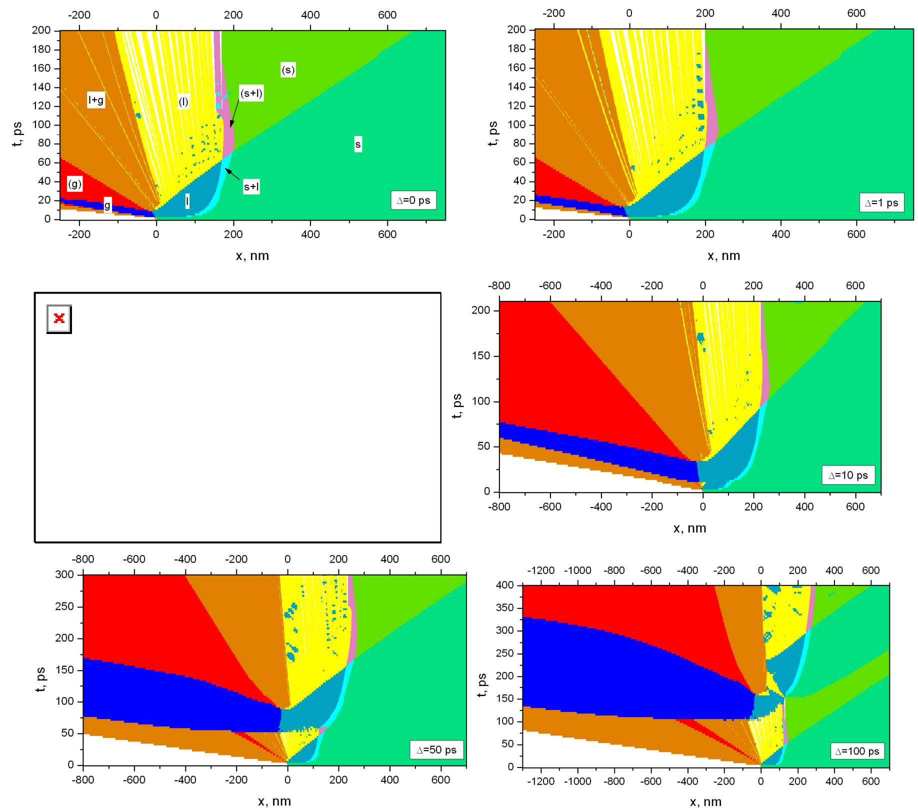

Fig. 1. x-t diagram for phase distribution in Cu irradiated by the double pulse. Different delays between the pulses are presented: 0, 1, 5, 10, 50 and 100 ps. Phase states are marked: s – solid, s+l – melting zone, l –liquid, g – gas, l+g – liquid-gas mixture, (s) – metastable solid, (s+l) – metastable melting, (l) – metastable over heated liquid, (g) – metastable oversaturated gas.

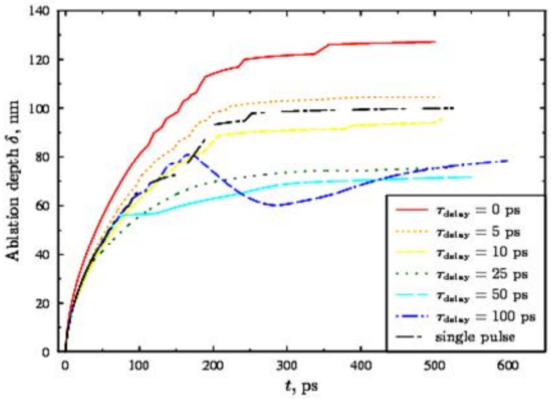

Fig. 2. The crater depth as a function of time for different delays between the pulses.

4. Conclusions

We have developed a self-consistent physical model for ultrashort laser interactions with metallic targets. The model is used for the simulation of the double pulse experiments with moderate laser fluences. Melting front propagation and phase transitions during the heating by the first pulse and the spreading of the target material is considered by using the multi-phase EOS in tabular form with separated components of electrons and heavy particles. The transport properties of the laser-irradiated materials are calculated by using the wide-range model of electron-ion collisions. The fragmentation of the liquid phase due to a strong tensile stress and high strain rate leads to the production of the liquid-gas mixture. The second pulse interacts with this rarefied zone and reheats it. The negative (tensile) pressures appearing in the solid phase during the propagation of the rarefaction wave can be strong enough (up to -5 GPa) but their life-time is typically too short to cause the spallation of the material. The work of pressure from the reheated plasma is shown to be the main reason of the decrease in the ablation depth with the increasing delay between the pulses.

Acknowledgments

This work is supported by the French-Russian cooperative project (CNRS DRI/Academy of Sciences of Russian Federation N 21276).

References

[1] A. Semerok, C. Dutouquet, Thin Solid Films **453-454**, 501 (2004).
[2] S. Noël, J. Hermann, Spectrochimica Acta Part B **94**, 053120 (2009).
[3] M. E. Povarnitsyn, K. V. Khishchenko, P. R. Levashov, Appl. Surf. Sci. **255**(10), 5120 (2009).
[4] M. B. Agranat, N. E. Andreev, S. I. Ashitkov, M. E. Veysman, P. R. Levashov, A. V. Ovchinnikov, D. S. Sitnikov, V. E. Fortov, K. V. Khishchenko, JETP Lett. **85**(6), 328 (2007).
[5] K. Eidmann, J. Meyer-ter-Vehn, T. Schlegel, S. Hüller, Phys. Rev. E, **62**(1), 1202 (2000), p..
[6] A. Yu. Kuksin, G. E. Norman, V. V. Stegailov V. V. et al. In: Proceedings of the Forth International Conference on Multiscale Materials Modeling (MMM-2008), (2008) p.442.
[7] M. E. Povarnitsyn, T. E. Itina, M. Sentis, P. R. Levashov, K. V. Khishchenko, Phys. Rev. B. **75**, 235414 (2007).

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