Laser ablation and spallation of crystalline aluminum simulated by Molecular Dynamics

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Abstract. The mechanical action of femtosecond laser pulse on a target may result in ablation of the irradiated frontal layer and spallation of the target rear side as well. The dynamics of expansion of the solid Al film after laser heating is studied by means of molecular dynamics (MD) simulations with the two EAM potentials (ours and Mishin et al.) and our parallel auto-balancing MPD\textsuperscript{3} code. It is found that the rear side spallation threshold is half as much again the frontal ablation threshold. The experimental and evaluated characteristics agree well in both crater depth and incident fluence on the ablation threshold.

The ultrashort laser-matter interaction at moderate fluence $\sim 1 \text{ J/cm}^2$ is important for fundamental physics of fast processes, warm dense matter physics as well as for a wide range of industrial applications [1–7]. Under action of an ultrashort pulse with duration $\tau < 1 \text{ ps}$ the heated layer $d_T$ is formed for the time less than the acoustic time $t_a = d_T/c_s$, where $c_s$ is a sound speed. Thus the ultrashort heating $\tau < t_a$ can be considered as an isochoric process. It gives rise to the stress confinement, which is a distinctive feature of the ultrashort laser heating.

For fluences in the vicinity of the ablation threshold $\sim F_a$, the time evolution may be divided into two consecutive stages: two-temperature (2T) and one-temperature (1T) stage [8]. In the beginning of the 2T stage the electron thermal energy $E^e$ is much larger than ion energy $E^i$ within the heated layer. As the thermal wave penetrates into the target, the electron temperature decreases and ion temperature increases. The 2T stage is completed when the electron and ion temperatures become equal. After that the 1T stage starts. The transition between 2T and 1T stages in Al occurs within the time slice 2–5 ps as pointed out in [8]. In the early time of the 1T stage the Gaussian curve can be used in fitting of the temperature profile which provides an approximation of the initial temperature $T_0(x)$ for MD simulations discussed below.

We use the MPD$^3$ algorithm [9] for large-scale parallel MD simulation of Al target with our own EAM potential and one given by Mishin et al. [10]. At $t = 0$ the motionless target is a perfect Al crystal having a face-centered cubic lattice at zero pressure and room temperature. The crystal is a rectangle with dimensions $l_x \times l_y \times l_z$ and periodical conditions imposed along the $y, z$-directions. The crystal direction [110] is aligned with $x$-axis, and $l_x = 689 \text{ nm}$. The maximal number of atoms in simulation was 76,296,000. The Gaussian initial temperature is formed by heating up Al atoms during $\sim 1–2 \text{ ps}$ by Langevin thermostat.

The dotted line in Fig. 1 is a pressure profile corresponding to the given temperature profile $T_0(x) = 4700 \exp(-x^2/d_T^2) + 300 \text{ K}$ with heated layer $d_T = 110.5 \text{ nm}$. The supersonic formation
Figure 1. The initial pressure $p(x, t = 1\text{ ps})$, and formation of the quasi-steady twofold pulse with two shocks $S_e$ and $S_f$.

Figure 2. The quasi-steady twofold pulse propagates into Al target heated up to $T_0(0) = 5\text{kK}$. In continuation of Fig. 1.

Figure 3. Hydrodynamic interaction between the initial pressure $p_0(x)$ (a) and the target boundary with vacuum 0 (b), resulting in the formation of twofold wave $p_{r+}$ (c). There are cavitation zone $[C_aC_b]$, the quiet zone $[C_be]$, and two shocks $S_e,S_f$ (d).
of pressure profile within a target gives rise to the rarefaction wave \( p_- \) and the compression wave moving from the left vacuum boundary into the bulk as illustrated by Fig. 1. With time the initial pressure transforms to quasi-steady twofold wave shown in Fig. 2, see also Fig. 3(d).

The Figure 3 illustrates how the initial triangular pressure profile can be decomposed in the two half waves \( p(x, t) = p_0(x + c_s t)/2 + p_0(x - c_s t)/2 = p_- + p_+ \) in linear acoustics approximation. Due to the reflection of \( p_- \) wave from the vacuum boundary \( x = 0 \) the pressure profile is a sum of three waves \( p_+ \), \( p_- \) and \( p_r \) for the period \( 0 < t < t_s \), as indicated by Fig. 3. The negative pressure area is placed within the section \( 0A = c_s t \) where the negative pressure wave \( p_r \) dominates the \( p_- \) wave. The negative pressure \( p_A = -p_{\max}/t_s \) at point A reaches its limit \( -p_{\max}/2 \) at \( t = t_s/2 \). If the cavitation pressure of material \( p_b \) is less than \( | -p_{\max}/2 | \) then the cavitation starts in the point A at the time \( t_b = t_s p_b/p_{\max} \). The wave \( p_- \) disappears after \( t > t_s \), and the quasi-steady twofold wave \( p_{r+} \) consisting of the compression \( p > 0 \) and the stretching \( p < 0 \) regions is formed as shown in Fig. 3(c). The nonlinear effect of focusing of characteristics results in the breaking of the compression wave \( p_+ \), as well as the breaking of the stretching wave \( p_- \) for the particular case \( F < F_a \). Wave breaking induces formation of two shocks, the first at point e (trailing front of the twofold pulse) and the second at f (leading edge of the pulse). In the case \( F > F_a \) a shock with \( p < 0 \) at the point e is created after appearance of the cavitation zone \( C_aC_b \) as illustrated by Fig. 3(cd).

To simulate a semi-infinite target we develop a new ”acoustic cut-off” procedure. The twofold pulse escapes from the boundary 0 leaving behind the quiet zone 0e or \( C_aC_e \) for the case \( F > F_a \). The terminating plane \( x_t \) begins to move with supersonic speed from the right boundary of a target to the quiet zone at the time \( t_s \), when the trailing front e has propagated a distance \( \sim d_T \). All atoms to the right of the terminating plane \( x_t \) are eliminated from MD simulation. The plane \( x_t \) stops at \( t = t_{ss} \) at the distance \( \sim 2d_T \) from the 0, as in Fig. 3(d). After that the free surface condition \( p_{\max} = 0 \) is established on the right boundary of remained target shown in Fig. 4. This procedure makes it possible to significantly reduce the simulation time per a step.

Figures 4 and 5 show the two MD simulations of complicated processes of ablation with the different heated depths \( d_T = 18.6 \text{ nm} \) and \( d_T = 110.5 \text{ nm} \), respectively. Bubble nucleation starts from \( t \approx 17 \text{ ps} \) at the pressure \( p \approx -2 \text{ GPa} \) between the right end of runaway layer and
Figure 5. Formation of a runaway layer on the left end of cavitation zone at 20.8 ps, and detachment of the runaway layer associated with decay of the foam at 71.6 ps.

The remaining Al target as shown in Fig. 5. The pressure in the cavitation zone grows up quickly to zero as the stretching stress is released. As a result the two shocks $S_e$ and $S_r$ are generated. Fig. 4 indicates that the surface tension of the binding foam slows down the runaway layer. Its final detachment from the target takes a long time for the ablation near threshold.

The MD simulation reveals that the ablated runaway layer appears first for the initial temperature $T_0(0) = 2590$ K. The evaluated ablation threshold is $F_a = 0.065$ J/cm$^2$, and the corresponding crater depth is 48 nm for the simulations with the number of atoms $N \approx 76 \times 10^6$ and the large $yz$-cross section $l_y \times l_z \approx 40 \times 40$ nm$^2$ of the MD box. The threshold temperature $T_a$ and $F_a$ are 2% higher for the MD simulations with smaller $yz$-cross section $7 \times 7$ nm$^2$.

The rear side spallation begins near the maximum of stretching stress $p_s \approx -9$ GPa for Mishin potential, as shown in Fig. 6. After first crack appears the pressure rapidly increases up to zero forming the two shocks. According our MD simulations the spallation threshold fluence is 0.17 J/cm$^2$ for Mishin potential, and $\sim 0.1$ J/cm$^2$ at $p_s \approx -7.5$ GPa for our EAM potential. It is should be noted that by contrast, the ablation thresholds are similar for the both potentials.

The experiments [8] give the ablation threshold for absorbed fluence $F_a = 0.064 - 0.07$ J/cm$^2$ and the corresponding crater depth 50 nm. The both characteristics are in a excellent agreement with evaluated ones presented above.

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