Analysis of effectiveness of subpicosecond multi-pulse laser ablation of metals

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Motivation: drilling, LIBS, nanoparticles, etc.
Phase diagrams & single-pulse trajectories, 20 J/cm²

Duration: 100 fs
Wavelength: 800 nm
Target: aluminum

![Phase diagram of aluminum with laser pulse parameters](image)
Ablation by ultrafast bursts of femtosecond pulses

C. Kerse et al., Nature 537, 2016

- Burst of pulses with duration ~ 60 ns
- Total fluence of the burst 10-30 J/cm²
- Number of pulses in the burst 5 – 200
- Each pulse: wavelength 800 nm, duration 800 fs
- Repetition rate from 86 MHz (12 ns) to 3.5 GHz (290 ps)

Predictions:
- Ablation cooling – increase of repetition rate reduce the heating of surrounding regions – the ablation efficiency is higher
- Energy of each pulse can be decreased without a reduction in the ablation efficiency
Two-temperature hydrodynamics

\[ \frac{\partial (1/\rho)}{\partial t} - \frac{\partial u}{\partial m} = 0 \]

\[ \frac{\partial u}{\partial t} + \frac{\partial (P_{\text{ion}} + P_{\text{el}})}{\partial m} = 0 \]

Conservation of mass and momentum

\[ \frac{\partial e_{\text{ion}}}{\partial t} + P_{\text{ion}} \frac{\partial u}{\partial m} = \gamma_{ei} (T_{\text{el}} - T_{\text{io}}) / \rho \]

\[ \frac{\partial e_{\text{el}}}{\partial t} + P_{\text{el}} \frac{\partial u}{\partial m} = \frac{\partial}{\partial m} \left( \rho \kappa_{\text{el}} \frac{\partial T_{\text{el}}}{\partial m} \right) - \gamma_{ei} (T_{\text{el}} - T_{\text{ion}}) / \rho + Q_L / \rho \]

Energy equations for ions and electrons

Laser energy absorption (Helmholtz equation)

\[
\frac{\partial^2 E}{\partial z^2} + k_0^2 [\varepsilon(z) - \sin^2 \theta] E = 0.
\]

\[
\frac{\partial^2 B}{\partial z^2} + k_0^2 [\varepsilon(z) - \sin^2 \theta] B - \frac{\partial \ln \varepsilon(z)}{\partial z} \frac{\partial B}{\partial z} = 0
\]

\[
Q_L(t, z) = I(t) \frac{\omega_L}{c} \text{Im}\{\varepsilon(t, z)\} |E(t, z)/E_L(t)|^2
\]

\[
\varepsilon_{\text{met}}(\omega_L, \rho, T_i, T_e) = \varepsilon_{\text{bb}} + 1 - \frac{n_e}{n_{\text{cr}}(1 + i \nu_{\text{eff}, \rho}/\omega_L)}
\]

\[
\varepsilon_{\text{pl}}(\omega_L, \rho, T_e) = 1 - \frac{n_e}{n_{\text{cr}}} \left[ K_1(\xi) - i(\nu_{\text{pl}}/\omega_L) K_2(\xi) \right]
\]

Nucleation model

\[ \tau_{\text{exp}} = (J_1 N)^{-1} \exp \left( \frac{W}{k_B T} \right) \]

\[ J_1 \approx 10^{10} \text{ s}^{-1} \]

\[ W = \frac{16\pi\sigma^3}{3(P_g - P_l)^2} \]

\[ \sigma = \sigma_0 \left(1 - \frac{T}{T_{\text{CP}}} \right)^{1.25} \]

\[ \tau_{\text{nuc}} = \tau_{\text{exp}} + \tau_{\text{grow}} \]

\[ \frac{1}{\rho} \frac{\partial \rho}{\partial t} \approx 10^{10} \text{ s}^{-1} \]
Ablation depth decreases with the delay between pulses.

Second pulse returns the ablated material on the target.

After the first pulse

After the second pulse

Multiple pulses: contour plots of density for 20 J/cm$^2$ burst

We neglect absorption in the plume from the previous pulse (big delay between pulses or weak pulses). Ablation depth grows with the number of pulses; for 200 pulses each pulse is below ablation threshold.
Crater depth dynamics for 20 J/cm² burst

Crater depth grows with the number of pulses in the burst
Crater depth is up to 15 times bigger in comparison with that for the single pulse
200-pulse ablation on the phase diagram

- The process for subthreshold pulses is almost isobaric
- Phase trajectories tend to the critical point where the phase explosion occurs
Temperature profiles in the target, $t = 60$ ns

Temperature gradient is the biggest for 200 pulses – ablation cooling
Almost linear dependence on the burst fluence
Bigger number of pulses in the burst gives bigger efficiency
Conclusions

• Single pulse ablation: produces population of different-size nanoparticles
• Double-pulse ablation: crater depth drops with the delay (even below the SP level)
• Multi-pulse ablation: rate increases with the repetition rate growth
• Material surface does not cool down substantially between successive pulses
• On the other hand, hot layers of the target are rapidly ablated forming steep temperature gradient – ablation cooling
• To prevent the shielding effect: the fluence of each pulse in the burst has a sub-threshold value
• Multiple pulses can guide thermodynamic trajectories along the binodal to the critical point