Molecular Dynamics Simulation of Ablation and Spallation of Gold irradiated by femtosecond laser

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Ablation

Spallation
Experimental Background

Typical experimental set-up:

Ti-Sapphire laser system
- $\lambda \sim 800 \text{ nm}$
- $\angle_{\text{center}} \sim 45^\circ$
- $t_{\text{pulse}} \sim 100 \text{ fs}$
- $F_{\text{incident}} < 1 \text{ J/cm}^2$

• Ablation threshold:
  - $F_{\text{absorbed}} \approx 0.13 - 0.18 \text{ J/cm}^2$

• Crater depth at threshold:
  - $d_{\text{crater}} \approx 110 - 130 \text{ nm}$

• Simulation models experiment by splitting it into two parts
  1. 2-temperature (ions and electrons) short time scale regime
  2. 1-temperature (ions) long time scale regime Molecular Dynamics
Physical processes in 2T regime

1. Transmission of energy from laser pulse to conduction electrons within skin layer

2. Transport of heat by electrons through Gold target

3. Exchange of energy between electron and ion subsystems until thermal equilibrium is established

Fit shape to functional form and use as initial profile for 1-temperature regime (MD simulation)
Physical processes in 1T regime
(simulated by MD)

4. Formation of pressurized anterior layer corresponding to initial temperature profile

5. Propagation of compression wave and rarefaction wave through Gold target

6. Negative pressure of rarefaction wave causes stretching and eventually void formation

7. Growth of voids can lead to ablation of melted frontal layer
Molecular Dynamics

- How does MD work?
  1. Determine forces on particles from an appropriate potential function

\[ F(r_i) = -\nabla V(r_i) \]

2. Solve Newton’s equations of motion for each particle for next time step, \( t + \Delta t \), using velocity Verlet algorithm

\[ F_i = m_i \ddot{r}_i \quad \text{integrate} \quad v_i(t + \Delta t) \text{ and } r_i(t + \Delta t) \]

3. Calculate desired physical properties and repeat

- Why do we use MD?
  - Short time and length scales in femtosecond laser ablation favors atomistic simulation.
  - Ultrafast physics of ablation is difficult for experiment as well as hydrodynamic simulation
Simulation set-up and method

- New many-body embedded atom method (EAM) potential for Gold,

\[ E_{tot} = \sum_i F(n_i) + \sum_{i<j} V(r_{ij}) \]

where \( F(n_i) \) is an embedding function, \( n_i(r) \) is a density function and \( V(r_{ij}) \) is a pair potential

- Three-dimensional MD cell with periodic boundary conditions along lateral dimensions

- Principal axis is located along crystal direction [110]

- Temperature distribution has the form of a mixed Gaussian,

\[ T(x) = T_1 \{ a \times e^{-x^2/d_{T1}^2} + (1-a) \times e^{-x^2/d_{T2}^2} \} + T_0 \]

where \( d_{T1} \) and \( d_{T2} \) are heating depths, \( T_1 \) is the surface temperature at left boundary and \( T_0 \) is a constant equal to room temperature.

- Simulated laser heating is accomplished by a Langevin Thermostat, which runs near the start of the simulation for \( \sim 20 \) ps
Verification of a new EAM potential

- Vacancy formation energy* – the energy required to rearrange an atom within the crystal lattice so a vacancy is formed

\[ E_{vf} = E_2 - \frac{N - 1}{N} \times E_1 \]

- Vacancy migration energy* – the energy required to move a nearest-neighbor atom to a vacancy position

* Important for defect and void formation processes that occur in ablation and spallation
Results for $E_{vf}$ and $E_{vm}$

<table>
<thead>
<tr>
<th>$E_{vf}$ (eV)</th>
<th>$E_{vm}$ (eV)</th>
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</thead>
<tbody>
<tr>
<td>0.94 ± 0.09\textsuperscript{a}</td>
<td>0.90 ± 0.04\textsuperscript{d}</td>
</tr>
<tr>
<td>0.97 ± 0.01\textsuperscript{b}</td>
<td>0.85 ± 0.03\textsuperscript{e}</td>
</tr>
<tr>
<td>0.98 ± 0.03\textsuperscript{c}</td>
<td>0.87</td>
</tr>
</tbody>
</table>

\textsuperscript{a} Simmons and Baluffi  
\textsuperscript{b} Triftshauser and Kauffman  
\textsuperscript{c} Bauerle and Koehler  
\textsuperscript{d} Sharma, Lee, and Koehler  
\textsuperscript{e} Kino and Koehler  

\textcolor{red}{\textbf{red}} - our EAM potential

Approaches constant value for increasing cell size

![Vacancy Migration Energy vs. Cell Size](image-url)
Determination of ablation threshold

• \( l_x \times l_y \times l_z = 800 \times 16 \times 10 \text{ nm} \)

• \( d_{T1} = 109 \text{ nm} \)  \( \cdot N_{\text{atoms}} = 7,977,600 \)

• \( d_{T2} = 460 \text{ nm} \)  \( \cdot T_0 = 300 \text{ K} \)

1. \( T_1 = 2400 \text{ K} \)
no void formation

2. \( T_1 = 2600 \text{ K} \)
one void forms, no detachment

3. \( T_1 = 2800 \text{ K} \)
many voids form, partial detachment

- From small simulation it is not clear if \( T_1 = 2600 \text{ K} \) corresponds to ablation threshold

- Large simulation is necessary to determine if ablation occurs at \( T_1 = 2600 \text{ K} \)

- Mass density profile shows many voids and detachment, therefore \( T_1 \approx 2600 \text{ K} \) at ablation threshold

\[ N_{\text{atoms}} = 170,962,180 \text{ atoms} \]

\[ t = 208 \text{ ps} \]
Fitting crater depth

- \(l_x \times l_y \times l_z = 800 \times 16 \times 10 \text{ nm}\)  
- \(N_{\text{atoms}} = 7,977,600\)  
- \(T_I = 2800 \text{ K}\)

1. \(d_{T_I} = 109\text{nm}: \quad d_{\text{crater}} = 88\text{nm}\)

2. \(d_{T_I} = 139\text{nm}: \quad d_{\text{crater}} = 103\text{nm}\)

3. \(d_{T_I} = 165\text{nm}: \quad d_{\text{crater}} = 122\text{nm}\)

4. \(d_{T_I} = 180\text{nm}: \quad d_{\text{crater}} = 150\text{nm}\)

- \(d_{T_I} = 165\text{nm}\) is in best agreement with experimental crater depth
- Large simulation with \(T_I \approx 2600 \text{ K}\) is needed to get values for \(d_{\text{crater}}\) and \(F_{\text{absorbed}}\) that can be compare directly to experiment

\(d_{\text{crater}} = 112\text{nm}\)

\(F_{\text{absorbed}} = 0.137 \text{ J/cm}^2\)

\(v_x = 120 \text{ m/s}\)

\(170,962,180 \text{ atoms}\)
Spallation processes and results

• Occurs for much higher $T_1$ than is necessary for ablation

• Compression wave from pressurized frontal layer propagates to rear-side Gold-vacuum interface and is reflected

• If rarefaction wave is strong enough crystal undergoes permanent deformation (fractures, voids) and rear layer is ejected

• Spall strength ($\sigma^*$) – maximum stress material can withstand before spallation

• Simulation for $T_1 = 5000 \text{ K}$ ($F_{\text{absorbed}} = .289$, above spallation threshold)
  
  $-\sigma^* = 11.8 \text{ GPa}$

• $\sigma^*$ theoretical limit ($T=0 \text{ K}$) $\approx 21 \text{ GPa}$
Conclusions

- Simulated values for $E_{vf}$ and $E_{vm}$ are in good agreement with experiment, therefore new EAM potential is well-suited for MD simulation of ablation and spallation.

- Ablation occurs in melted Gold, whereas spallation occurs in solid Gold crystal.

- Ablation threshold values:
  - $F_{\text{absorbed}} = 0.137 \text{ J/cm}^2$ (simulated)  \quad 0.13 – 0.18 J/cm$^2$ (experiment)
  - $d_{\text{crater}} = 112 \text{ nm}$ (simulated)  \quad 110 – 130 nm (experiment)

- Future work will determine $F_{\text{absorbed}}$ at spallation threshold, which should be greater than $F_{\text{absorbed}}$ at ablation threshold.

- Experimental results for $\sigma^*$ under conditions created by a femtosecond laser are needed to compare to simulation result of 11.8 GPa.
Acknowledgements

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