Ablation into water: Fragmentation of metal via Richtmyer–Meshkov instability

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Abstract. The consistent molecular dynamics and smoothed particle hydrodynamics (SPH) simulations of gold ablation into water are performed in a quasi two-dimensional setup. Both approaches demonstrate accretion of the nucleated gold to the gold–water interface, producing sound waves which propagate to the water. Jets growth provided by SPH simulation is similar to the Richtmyer–Meshkov instability development.

1. Introduction

The investigation of the ultrashort laser–matter interaction is important both for understanding fundamental processes in high energy density physics and industrial applications [1]. The last ones include various surface processing methods such as pulsed laser deposition of thin films, fabrication of nanostructures and nanoparticles.

The process of laser irradiation of a material surface is accompanied by a variety of physical phenomena: laser absorption by electrons, melting and nucleation, surface tension, extreme strain rates, development of instabilities, and many others. That is why fluid dynamics simulations are usually quite difficult to perform: one should use a thermodynamically complete equation of state with stable and metastable states and proper phase boundaries [2], two-temperature hydrodynamic model for electrons and ions [3], a surface tension implementation, and a fragmentation model. However, most of the required physics may be naturally simulated via molecular dynamics (MD) [4–7].

In this research, we extend the results obtained in the paper [8] for the ablation of a gold layer into the water at nanoscales. By emulating a laser pulse absorption with a heating source in a two-dimensional (2D) setup we explore the development of instabilities under ablation, that can be useful in determining nanoparticles size distribution. Simulations are performed via MD and smoothed particle hydrodynamics (SPH) methods.
2. Contact SPH equations

The approach for medium representation with smoothing particles has many advantages. In contrast to usual grid methods, the meshless nature of SPH does not require special algorithms to handle shock fronts, contact surfaces, or free boundaries. Thus, there are many opportunities for SPH application in simulations of materials in extremes: explosion, ablation, fragmentation, high-speed collision, etc.

The smoothed particles approximation of the function $f$ at the point $r$ using its known values $f_1, \ldots, f_N$ at $N$ points $r_1, \ldots, r_N$ within the smoothing distance $h$ is given by

$$ f(r) = \sum_{j=1}^{N} f_j \Delta V_j W(|r - r_j|, h), \quad (1) $$

where $W(r, h)$ is the smoothing function (kernel), $\Delta V_j$ is the volume related to the point $j$. The smoothing kernel is usually given in a form of a gaussian or polynomial function with conditions

$$ \int_{r<h} W(r, h)d^3r = 1, \quad \lim_{h \to 0} \int_{r<h} W(r, h)d^3r = \delta(r). \quad (2) $$

This approach provides not only the function approximation at a given point $r$, but also its derivative:

$$ \nabla f(r) = \sum_{j=1}^{N} f_j \Delta V_j \nabla W(|r - r_j|, h), \quad (3) $$

where the kernel gradient is

$$ \nabla W(|r - r_j|, h) = W'(|r - r_j|, h) \frac{r - r_j}{|r - r_j|}. \quad (4) $$
The described approximation technique may be directly applied to represent a medium via a set of particles which keep values for the following functions: the density $\rho$, the velocity $U$, the specific internal energy $e$, the pressure $P$. Evolution of medium is described via the set of equations:

$$\frac{d\rho_i}{dt} = -\rho \nabla \cdot U,$$

$$\frac{dU_i}{dt} = -\frac{1}{\rho} \nabla P,$$

$$\frac{de_i}{dt} = -\frac{P}{\rho} \nabla \cdot U,$$

which may be rewritten for smoothed particles of masses $m_i$. By denoting $r_{ij} = r_j - r_i$, $e_{ij} = r_{ij}/r_{ij}$, $\nabla W_{ij} = W'(r_{ij}, h)e_{ij}$, $\Delta V_i = m_i/\rho_i$ for the particles $i$ and $j$, the equations (5)–(7) are transformed to the smoothed particles hydrodynamics (SPH):

$$\frac{d\rho_i}{dt} = \rho_i \sum_{j=1}^{N} \frac{m_j}{\rho_j} (U_i - U_j) \cdot \nabla W_{ij},$$

$$\frac{dU_i}{dt} = -\sum_{j=1}^{N} \frac{m_j}{\rho_i \rho_j} P_i + P_j \nabla W_{ij},$$

$$\frac{de_i}{dt} = \frac{1}{2} \sum_{j=1}^{N} \frac{m_j}{\rho_i \rho_j} (P_i + P_j)(U_i - U_j) \cdot \nabla W_{ij}.$$

Unfortunately, the given approach provides non-monotonous solutions at interparticle contact discontinuities. The introduction of the artificial viscosity is quite popular approach to avoid that, but we utilize the Riemann problem solution at interparticle contacts [9]. Instead of using the velocity $U_j$ of particle $j$ in (8)–(10) it is more accurate to obtain the velocity of the interparticle contact $U_j^*$ as the solution of 1D Riemann problem between $i$ and $j$ particles along the direction $e_{ij}$ (figure 1). The more accurate contact pressure $P_{ij}^*$ should also be placed instead of $(P_i + P_j)/2$ resulting in the contact SPH (CSPH) equations:

$$\frac{d\rho_i}{dt} = 2\rho_i \sum_{j=1}^{N} \frac{m_j}{\rho_j} (U_i - U_j^*) \cdot \nabla W_{ij},$$

$$\frac{dU_i}{dt} = -\frac{2}{\rho_i} \sum_{j=1}^{N} \frac{m_j}{\rho_j} P_{ij}^* \nabla W_{ij},$$

$$\frac{de_i}{dt} = \frac{2}{\rho_i} \sum_{j=1}^{N} \frac{m_j}{\rho_j} P_{ij}^*(U_i - U_j^*) \cdot \nabla W_{ij}.$$

Materials in extremes may be dramatically compressed or expanded what requires SPH particles to have variable smoothing length $h$. The particle size is defined as $d_i = \sqrt{m_i/\rho_i}$, and the smoothing length is set to $h_{ij} = (d_i + d_j)/2$. Interaction distance is then defined by a condition $W((r_{ij})^2, h_{ij}) > 0$, or $|r_{ij}| < \kappa h_{ij}$, where $\kappa$ is the smoothing kernel width. Further simulations are performed using the Wendland C$^2$ kernel [10] with $\kappa = 1.936$.

3. EOS for water and gold

The set of equations (11)–(13) is enclosed by the equation of state (EOS) $P = f(\rho, e)$, which is taken in the form of Mie–Grüneisen:

$$P - P_r = \gamma \rho (e - e_r),$$

where $P_r$ is the reference pressure, $e_r$ is the reference energy, $\gamma$ is a constant that characterizes the compressibility of the medium. This equation is usually solved numerically due to the non-linearity of the EOS.
Figure 2. The gold (a) and the water (b) reference data fit (15) for SPH simulation and the EAM potentials [5] for MD simulation.

Table 1. Gold and water properties.

| Material property          | Gold  | Water |
|----------------------------|-------|-------|
| Normal density $\rho_0$, kg/m$^3$ | 19320 | 1000  |
| Bulk sound speed $c$, km/s  | 3.07  | 1.498 |
| Hugoniot coefficient $a$   | 1.54  | 1.95  |
| Gr"uneisen parameter       | 2.05  | 0.11  |

where $P_r$, $e_r$ are the reference pressure and the specific energy for the shock Hugoniot in the form $u_s = c + au_p$, where $u_s$ is the shock velocity, $u_p$ is the particle velocity, $c$ is the bulk sound speed, $a$ is the coefficient:

$$P_r(x) = \rho_0 c^2 \frac{1 - x}{[1 - a(1 - x)]^2}, \quad e_r(x) = \frac{P_r}{\rho_0} \frac{1 - x}{2}, \quad x = \rho_0/\rho. \quad (15)$$

One should notice that a laser pulse induces the high thermal pressure at normal density in a gold layer which is then releases to the water. Therefore, the cold curve for gold in our simulation should be well fitted for the densities below normal $\rho_0$. For the ab-initio reference data in figure 2(a), there are the embedded atom model (EAM) potential [5] and our shock Hugoniot fit (15).

The EAM potential developed to emulate mechanical properties of water provides a good agreement with shock-wave data [11, 12] in a wide range of compressions. However, the data could hardly be approximated via linear $u_s-u_p$. Fortunately, MD simulations predict water shock amplitude to be about 10 GPa and less, what can be approximated using linear $u_s-u_p$ Hugoniot as demonstrated in figure 2(b).

The summary of EOS parameters for the SPH-simulated gold and water are given in table 1.

4. Simulation setup

MD and SPH simulations of gold ablation into water are preformed in rectangular domains $L_x \times L_y \times L_z$ where $x$ is the main axis, along which layers of water and gold are placed, $y$ and $z$ are in periodical boundary conditions. Instability development requires several nanoseconds
Figure 3. Pressure profiles at $t = 23.04$ ps after irradiation from SPH and MD simulations of the gold–water interface. The water layer is limited with the LP boundary which position, velocity and material state are recorded in preliminary simulations of system with a large $x$-dimension and small $y$–$z$ cross-section.

during which a shock propagates in water for several micrometers. Transversal sizes $L_y$ and $L_z$ should contain several instability modes, so the reasonable choice is about 100 nm. The complete MD simulation thus requires about $10^{10}$ atoms that is huge amount even for the top supercomputers. SPH is more flexible and independent of spatial scales, but also requires quite good resolution to observe the development of instabilities.

To overcome the described difficulties the spatially reduced, but physically equivalent problem is simulated. First, the reasonable simplification [13] is to replace the full three-dimensional (3D) ($L_z = 100$ nm) setup with the quasi 2D ($L_z = 10$ nm). Second, the unnecessary gold and water particles, which are far away from the interface, are removed.

The gold interface layer is quite easy to separate. High temperature and release induces nucleation in gold at the distance about 100 nm from the interface. Nucleation zone is represented by the two-phase state of gold with relatively low sound speed. Thus, the removal of the gold atoms at some boundary $x_{Au}$ in this zone does not produce sound waves which may affect the gold–water interface.

In contrast to gold, the water layer could not be separated at some boundary $x_w$ without producing a rarefaction wave moving back to the interface. But it is possible to place the lagrangian particle (LP) boundary, which represents the properties of a water at the position $x_w(t)$ during shock propagation (figure 3). Thus, if the particles behind this boundary are removed, for those particles at the interface there will be no difference whether they interact with the real particles or such boundary.

To setup the LP boundary we first need to track the loading history of real particles. It may be done in quasi one-dimensional (1D) simulation with the $L_y = 10$ nm, where the water layer length is set to 1.5 $\mu$m. The initial progress is shown in figure 3. Water–gold interface is at $x = 100$ nm, the length of the gold sample is 500 nm. In SPH the laser pulse is emulated via the heating source for gold with the heating rate in the form $w(t, x) = w_1 + w_2$, where

$$w_k(t, x) = w_{0k} \exp \left(-\frac{t}{\tau} \right) \exp \left(-\frac{(x-x_0)^2}{d_k^2} \right),$$

(16)

$\tau = 5$ ps, $x_0 = 100$ nm, $w_{01} = 5 \times 10^{17}$ W/kg, $d_1 = 50$ nm, $w_{02} = 2.8 \times 10^{17}$ W/kg, $d_2 = 250$ nm. The two-gaussian form describes both the skin layer energy deposition ($w_1$) and the thermal wave propagation ($w_2$). In molecular dynamics, we use a Monte-Carlo model for electrons which provides a proper heat transfer from electrons to ions. The model parameters are adjusted to provide a proper coefficient of thermal conductivity in liquid gold. The resulting pressure profiles simulated in MD and SPH are shown in figure 3.
The LP boundary is tracked for the water particles at $x = -400$ nm. The time histories for velocity and density in figure 4 are the result of SPH simulation. To perform further simulations with bigger samples these profiles are fitted with the following functions:

$$\rho = \rho_0 + \Delta \rho \exp\left[-\left(\frac{t - t_0}{\Delta t_\rho}\right)^{0.475}\right],$$  \hspace{1cm} (17)

$$U = -\Delta U \exp\left[-\left(\frac{t - t_0}{\Delta t_U}\right)^{0.415}\right],$$  \hspace{1cm} (18)

where $\rho_0 = 1 \text{ g/cm}^3$, $\Delta \rho = 0.6518 \text{ g/cm}^3$, $\Delta U = 2.08 \text{ km/s}$, $t_0 = 67 \text{ ps}$, $\Delta t_\rho = 311.6 \text{ ps}$, $\Delta t_U = 185.4 \text{ ps}$. The functions have the proper asymptotes at $t \to \infty$: $\rho \to \rho_0 = 1 \text{ g/cm}^3$, $U \to 0 \text{ km/s}$.

In MD the LP boundary can be emulated with the potential barrier which propagates with the approximated velocity (18). SPH particles at boundaries should have properly calculated strains what cannot be done with such barrier. Thus, the pack of particles with the predefined density and velocity history is placed at the LP boundary, so that “real” particles interact with them properly.

SPH simulation of gold ablation into water is performed using CSPH-VD$^3$ software with the dynamic load balancing parallel algorithm [14] in accordance with the MD simulation performed using MD-MPD$^3$ software [15] with the same balancing algorithm. In quasi 2D simulations we use $L_y = 260$ nm, $L_z = 10$ nm and the same heating properties.

Before we proceed to results, some remarks about SPH and MD setup should be added. Except the electron thermal conductivity, MD also naturally takes into account the solid–liquid phase transition and the surface tension in gold. SPH simulations are rather simplified: no phase transition (liquid gold and water), no surface tension, and no electron–ion energy transport. But this first step is necessary to analyze the influence of these effects.

5. Simulation results

MD simulation demonstrates the development of instabilities with mode coupling. In figure 5 one can notice that at late times ($> 1$ ns) there is the stable mode with $\lambda = L_y/3$. CSPH
Figure 5. MD and SPH simulation results in the quasi 2D setup: gold is represented in yellow, water in blue. In both approaches the formation of nucleation zone is observed with the accretion of gold to a liquid water–gold interface. The jets growth in SPH corresponds RM instability. In MD jets collapse into droplets due to surface tension.

method is too viscous, so it blocks the growth of small instabilities. In our SPH simulation we introduced the mode $\lambda = L_y/3$ with the initial perturbation amplitude $p = 5$ nm. The instability development is shown in figure 5.

Nucleation of gold in SPH appears due to limiting the tensile stress by 0.1 GPa which is observed in MD. This process is consistent with MD with the only difference in the surface tension. Another similar phenomenon is the gold accretion: as soon as the gold–water interface slows by the water layer, the gold from nucleation zone move towards the interface what increases the width of the liquid layer. The accretion also induce waves which propagate to the interface and beyond: in figure 4 one may notice oscillations on density and velocity histories. It is not yet understood whether this process also affects the instability development.

Finally, the SPH simulation provides the gold jets in the form of Richtmyer–Meshkov instabilities. With the surface tension jet tips would collapse into a drop (similar to MD) what would result in nanoparticle of specific size corresponding the chosen mode $\lambda$.

6. Conclusion
The consistent MD and SPH simulations of the gold ablation into water are performed in quasi 2D setup. Both approaches demonstrate accretion of the nucleated gold to the gold–water interface, producing sound waves which propagate to the water. Jets growth in SPH looks similar to the well-known Richtmyer–Meshkov problem. The surface tension, phase transitions, and electron–ion energy transport in SPH will be introduced in future works.

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