Fluid simulation of plume head-on collision dynamics during pulsed laser ablation

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Abstract. Expansion dynamics of plume after irradiation of the target material is essential to prepare nanoparticles by pulsed laser ablation and it can be modified by collision of two plumes. In the present paper, effect of head-on collision on the expansion dynamics is discussed by numerical simulation based on the fluid dynamics and compared with the experimental results of plume emission. Suppression of plumes by collision with counter plume observed by experiment is reproduced by numerical simulation. Results of the numerical calculation indicate that shockwave induced by the irradiation of the opposite target suppress vapor expansion. The vapors do not mix around the center of the targets when the two targets are irradiated simultaneously and unstable flow is seen when delay between laser pulses was applied for irradiation of two targets. The results of the numerical simulation suggest that formation of combined and alloy nanoparticles are expected for former and latter cases.

1. Introduction
Formation of nanoparticles during pulsed laser ablation (PLA) in background gas is reported for various materials. Strong pulsed laser irradiation of target induces high-density, -pressure and -temperature plasma, plume, and it expands to the background gas. The analysis of point explosion well describes dynamic process of the plume expansion [1–3]. Nano-droplets are thought to be formed during the free expansion of the plume by the adiabatic cooling followed by nano-crystallization on further cooling [4, 5]. The control of nanostructure is expected by varying dynamic plume expansion processes. The dynamic expansion process can be modified by the background gas pressure and laser fluence, and correlation between the preparation condition and structure of nanoparticles has been discussed so far [6, 7]. Introducing synchronized two laser beams enable us to collide two plumes. The collision of the two plumes modifies plume expansion. For example, a layer of stagnated plasma is formed at the collision front when two plumes collide under appropriate conditions [8, 9]. One of the practical applications of colliding two plumes is formation of high quality droplet free films [10]. Recently, a method to form novel nanoparticles by colliding two plumes is proposed [11, 12].

Not only for the material processing, PLA has many similarities with astrophysical phenomena such as point source explosion, expansion of plasma, formation of shockwave and so on. The effect of
collision of two plasmas is also important to reveal the evolution of structures within supernova remnants [13, 14]. In the present paper, we focused on the effect of plume collision on the nanoparticle formation. Irradiation of a target by a pulsed laser and that of two targets by two pulsed lasers are referred to as “single pulsed laser ablation (SPLA)” and “double pulsed laser ablation (DPLA)”, respectively. Effect of collision on the expansion dynamics is discussed by comparing numerical and experimental methods and a way to control nanoparticle formation is proposed.

2. Experiment
Figure 1 shows experimental setup of DPLA. Si and Ge targets were placed parallel to each other at distance of 9 mm. The targets were irradiated by individual two YAG lasers. The wavelength, pulse width, repetition rate and fluence were 355 nm, 7 ns, 10 Hz and 1.6 J/cm² for ablation of the Si target and 266 nm, 7 ns, 10 Hz and 1.9 J/cm² for that of the Ge target, respectively. Helium gas was introduced as a background gas and kept constant value during the ablation. The plume expansion dynamics was monitored by gated ICCD equipped with monochromer. The ICCD camera is positioned at the front side of the sheet in figure 1. The details of experimental set up are shown in Ref. 11.

3. Numerical method and initial conditions
We perform two-dimensional hydrodynamics calculations in the cylindrical coordinate system. We use following ordinary set of equations:

\[
\frac{\partial \rho}{\partial t} = - \nabla \left( \rho \cdot \vec{v} \right), \quad (\rho = \rho_{\text{Si}} + \rho_{\text{Ge}} + \rho_{\text{He}}), \tag{1}
\]

\[
\rho \left( \frac{\partial \vec{v}}{\partial t} + (\vec{v} \cdot \nabla) \vec{v} \right) = - \nabla (P + q), \tag{2}
\]

\[
\frac{\partial e}{\partial t} = - \nabla ((e + P) \rho \cdot \vec{v}), \tag{3}
\]

\[
e = \frac{1}{\gamma - 1} \frac{P}{\rho}. \tag{4}
\]
Here we solve continuity equations for three species (suffix \(i\) in (1) denotes He, Ge and Si). As shown in (1), \(\rho\) represents the total density of the fluid, while \(\vec{v}\) is the velocity, \(P\) and \(e\) denote the thermal pressure and the internal energy, respectively. \(q\) denotes the additional pressure term originated from the von-Neumann type artificial viscosity. In practice, we combine (3) and (4) to solve the energy equation. Substituting (4) into (3), we have

\[
\frac{\partial P}{\partial t} + \vec{v} \cdot \nabla P = -\left(\gamma P + (\gamma - 1)q\right)(\nabla \cdot \vec{v}).
\]  

We solve (5) instead of (3). Even though the plume of PLA is plasma state, neutral gas is assumed here for simplicity and electromagnetic effects are not taken into account. From this reason, we use the term ‘vapor’ instead of ‘plume’ for the result of calculation.

Here, we adopt an equation of state of ideal gas. We assumed the initial densities, pressure, temperature to reproduce the laser fluence and ejected mass per laser shot from experimental results [7]. The adopted parameters are summarized in table 1.

**Table 1.** Initial conditions used in the calculation.

|        | Density (kg/m\(^3\)) | Pressure (Pa) | Temperature (K) |
|--------|-----------------------|---------------|-----------------|
| Si     | 0.038                 | 6.3 \times 10^7 | 5.2 \times 10^6 |
| Ge     | 0.038                 | 7.4 \times 10^7 | 1.4 \times 10^7 |
| He     | 0.042                 | 260           | 300             |

In order to treat a shock wave, we applied Constrained Interpolation Profile (CIP) scheme in which advection and non-advection terms in the equations are separately solved. We also employ von-Neumann type artificial viscosity in order to capture the shock front with a given grid size [15-17]. The additional pressure term \(q\) in (2) and (5) is given as

\[
q = \alpha \left(\rho C_s(\nabla \cdot \vec{v}) + \frac{\gamma + 1}{2}(\nabla \cdot \vec{v})^2 \lambda^2\right) \quad \text{if} \quad \nabla \cdot \vec{v} < 0, \tag{6}
\]

\[
q = 0 \quad \text{if} \quad \nabla \cdot \vec{v} > 0, \tag{7}
\]

where \(C_s\) is the sound velocity of the fluid, \(\lambda\) is the assumed thickness of the shock front which is set to be the grid size in the present simulation. We adopt \(\alpha=0.75\) being appropriate for shock waves.

4. Results and discussion

The images of the plume expansion dynamics observed by the gated ICCD camera when two targets are irradiated simultaneously are shown in figure 2. Contour plot images of the total gas pressure and densities of the Si and Ge vapors obtained by numerical simulation are shown in figures 3a and 3b, respectively. The profiles of densities of the Si and Ge vapors and temperature along symmetric axis under background gas pressure of 270 Pa at 100 ns after the laser irradiation are shown in figure 4. The high-density region roughly corresponds to the high temperature region. Slight increase in the temperature around the center of targets is observed by collision. However the temperature around the center of targets is about one order smaller than the highest temperature, it increases to considerable value by selecting initial condition.
Figure 2. The images of plume emission under background gas of 270 Pa at 100, 300, 500 and 700 ns. The red and blue lines in the figures show the positions of the Si and Ge targets, respectively. The distance between targets is 9 mm.

Figure 3. (a) Contour plots of total pressure and (b) Contour plots of Si and Ge vapor densities at 100, 150, 250 and 2000 ns after targets are irradiated simultaneously. The Si and Ge targets are at 0 and 9 mm, lower and upper side of the figure.
Figure 4. The densities of the Si and Ge vapors and temperature under background gas pressure of 270 Pa at 100 ns after the laser irradiation.

We define front edge of vapors at the position where density decreases to $1 \times 10^6$ kg/m$^3$. The position of vapor front-edges is plotted in figure 5 versus time after the pulsed irradiation. Positions of the Si and Ge targets are 0 and 9 mm, respectively, on vertical axis of these figures. Even though the agreement of absolute value of time scale is not good, results of calculation are qualitatively similar to those of experiments [12]. The effect of DPLA is remarkable above 140 ns. The vapor front-edge moves backward at this time. The positions of the shockwave and vapor front-edge can be estimated from figures 3a and 3b, respectively. The shockwave induced by irradiation of the opposite target reaches the vapor front-edge at around 150 ns as shown in figure 3. Since this value is close to 140 ns, the backward motion observed in figure 5 should be the effect of counter shockwave. As we can see in the figure 3, the Ge and Si vapors are spatially separated even at 2000 ns after irradiation. This suggests formations of the Si and Ge nanoparticles in the spatially separated Ge and Si vapors individually. Formation of aggregates of combined Si and Ge nanoparticles is expected at later stage.

The results of calculation are suggestive also for position of substrate. The gas flow perpendicular to the symmetric axis around the center of targets is observed by calculation. This suggests that collection of nanoparticles is possible by placing substrate perpendicular to the substrate apart from symmetric axis as shown in figure 1.

Figure 5. Time evolution of the front edges of vapors under background gas of 270Pa. The Si and Ge targets are at 0 and 9 mm, lower and upper side of the figure.

Delay-time between the two laser pulses is easily controlled parameter both for the experiment and calculation. Effect of the delay-time on the vapor collision is shown in figure 6. This is a result when
the Ge target is irradiated following the irradiation of the Si target and the delay-time is set at 700 ns. The extensive unstable flow of the vapors begin to seen after 1000 ns of Si target irradiation, which is after 300 ns of Ge target irradiation, and clearly seen at 2000 ns as shown in figure 6. This time corresponds to the time when the Ge vapor reaches the Si target. The position of Ge vapor front as a function of the time after irradiation of Ge target when delay time is applied is shown in figure 7. However the effect of the counter vapor seen in figure 5 is observed at delay-time of 100 ns, such effect is not seen at delay-time of 700 ns or higher. The results of vapor expansion profile indicate that the delay time of 700 ns corresponds to the time when the Si vapor front reaches the Ge target. The very unstable feature is found at delay-time of 700 ns as shown in figure 6. A similar feature is observed at higher delay-time. One of the possible reasons of the very unstable feature is that the density of the Si vapor becomes lower with time, and the more compact dense Ge vapor penetrates the Si vapor like a bullet at higher delay-time. As a result, strong shear flow arise at the contact surface of Si and Ge vapors, and Kelvin-Helmholtz instability becomes prominent. Unfortunately, this unstable flow is not observed in the experimental plume emission. It is presumably because unstable flow grows at later stage of the plume expansion and the plume emission vanishes at this stage. We can expect mixture of two plumes by the extensive unstable flow shown here and formation of Si-Ge alloy nanoparticles if the temperature is not below the nanocrystallization at this time scale.

![Figure 6](image1.png)

**Figure 6.** Contour plots of the Si and Ge vapor densities when the Ge target is irradiated 700 ns after irradiation of the Si target. The Si and Ge targets are at 0 and 9 mm, lower and upper side of the figures. Right and left half of the figures show densities of Si and Ge vapors, respectively.
5. Conclusion
We show effects of collision of two vapors by numerical simulation. The vapor expansion dynamics is qualitatively similar to experimental plume expansion. The effects of shockwave on the plume expansion dynamics are discussed from the simulation. Results of numerical simulation show possibility to control nanostructure by controlling the delay-time between the laser pulses. The shockwave induced by irradiation of the opposite target suppress expansion of vapors. The two vapors are confined by counter shockwave and mixture of the vapor is not seen when the targets are irradiated simultaneously. The extensive unstable flow of the vapors is observed when the delay was applied between the laser pulses. These results give valuable information for design of the hybrid nanoparticles generation.

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