

## Evolution of the Collision Rate in the Process of Propagation of Laser-Generated Plume into an Inert Gas

Y.L. Wang <sup>1,\*</sup>, G.I. Albokhary <sup>1,2</sup>, X.C. Ding <sup>1</sup>, L.Z. Chu <sup>1</sup>, Z.C. Deng <sup>1</sup>,  
W.H. Liang <sup>1</sup> and, G.S. Fu <sup>1</sup>

<sup>1</sup> College of Physics Science and Technology, Hebei University, Baoding 071002, China

<sup>2</sup> Faculty of Engineering and Technology, University of Gezira, Wad Madani, Sudan

\*E-mail: hdwangyl@mail.hbu.edu.cn

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**Abstract.** The quantity of collision rate (CR) is one of the most important parameters that need to be taken into account when dealing with gas dynamics of unsteady regime due to its role in determining the final conditions of the gas-phase formed clusters. In the present work, the so-called direct simulation Monte-Carlo (DSMC) method has been used as a computational tool to investigate the transportation modes of laser-generated Plume (LGP) of Si target into He inert gas. For this purpose, the spatial evolution of the quantity of CR has been temporally traced among the entire space of flow field. The existence of several previously reported phenomena using different physical quantities such as number density, temperature and velocity have been also explored and confirmed. An excellent agreement with our current works has been found. These previously noticed phenomena may include oscillating behavior of vapor front and interaction of shock compressed wave with both of substrate surface and vapor front.

**Keywords:** Pulsed Laser Deposition (PLD); Direct Simulation Monte-Carlo (DSMC); Nucleation and Growth; Nanoparticles (nPs); Collision Rate (CR)

### 1. Introduction

Pulsed laser deposition (PLD) has been considered, for more than one decade, as a much powerful tool for synthesizing a wide range of nanoscale materials for promising applications such as fullerenes and Si nanoparticles (nPs) [1].

In spite of the fact that the common setup of PLD apparatus arrangement is quite simple compared with many other thin films deposition techniques, the fundamental mechanisms of processes developing during the transfer of material from ablated target to a distant placed substrate are not completely explored and are consequently centered at the interest of much researchers in this field.

The physical phenomena of laser-target interactions and film growth are of great complexity. It involves all the physical processes that can be expected as a result of the impact of the high-power pulsed radiation on a solid material.

An ambient inert gas is often used for PLD setup to accelerate the collision rate of reactant ablated nucleus, inert gas molecules and, their combination groups thus makes it possible for the energy to be gradually exchanged and distributed between these species resulting in the nucleation and growth process of the gas-phase formed nanoclusters. The existence of an

ambient gas during laser ablation (LA) of target material is considerably altering gas dynamics behavior of ablated particles and lead to a much complicated expansion and propagation fashions. These include the formation of shock waves [2- 4], plume segregation into fast and slow components [5- 11] and oscillations of the vapor contact front accompanied with a high density mixed region [12, 13].

Numerous of fundamental informations have been successfully revealed by carrying out both experimental and theoretical approaches. These may include, optical emission spectroscopy [14-16], mass spectroscopy [17], laser-induced fluorescence [18], Langmuir probe flux measuring [19], shock wave model [2] and direct simulation Monte-Carlo [4, 12]. In all those above works researchers were seeking to explore the mean features of gas dynamic by reporting some common effective quantities of flow domain, in other words the velocity, temperature, density and pressure profiles have been extensively discussed in the context of nucleation and growth processes of the gas-phase synthesized nPs via these works.

However, to our knowledge none of those researchers has tried to discuss the dynamics mechanisms of plume expansion into an ambient gas in terms of the quantity of collision rate (CR). Although, CR quantity is the most important factor that related directly with the nucleation and growth process of gas-phase formed nPs. In this article, the plume dynamics during nPs formation and film deposition will be discussed, as well as the formation of the so-called shock waves occurred by the compressed background gas and its interaction with the thin film deposited onto apart located substrate.

## **2. Methodology**

Overall, one can speculate about the physical processes occurring during the operation of PLD units of single pulse as being composed of four distinct and consecutive stages. However, each of these stages involves different kind of interaction, thus the whole evolution of the system might only be treated by choosing more than one approach depending on the particular stages, which is focused on. These stages are listed below:

- 1) Laser radiation interaction with the target material, after an intense laser beam is focused onto the surface of the target.
- 2) Propagation and expansion of the laser-generated plume (LGP) which will be focused on through this article.
- 3) Deposition of the ablation material onto the substrate.
- 4) Nucleation and growth of a thin film on the substrate surface.

One of the most important stages, that play an important role on determining the final conditions of the so-formed nPs, is the stage of expansion and propagation of laser LGP. This stage is characterized by a relatively long period in comparing by other stages and many mechanisms that may alter the next two stages are dominant by it.

In this report, the so-called direct simulation Monte-Carlo (DSMC) is used as a computational tool to inspect and analyze the dynamics of propagation of LGP into an ambient inert gas in terms of collision quantity.

Here we will give a brief presentation for the basic features of our previously developed DSMC model for plasma plume expansion into an inert gas. As well as the obvious goal, the computational setup used through this study will also be described. We have developed a comprehensive DSMC approach for the propagation of LGP. The model has options to be driven in different configurations of geometric flow viz., planar, cylindrical and spherical flow. Diffuse reflected surface with a selectable value of accommodation coefficient has also been adopted throughout the model. Complete details for our model are given in our incoming paper that is still under revision [20].

In the simulation, a target material of Si element has been considered to be ablated by an ultraviolet laser source of solid-state type, which has wavelength, laser fluency and pulse duration of 532 nm, 5 J·cm<sup>-2</sup> and 10 ns, respectively. The radius of laser spot on the target surface is 0.5 mm. The ambient gas of He is pumped through the gas inlet and its pressure is kept to at a stable value of 1000 Pa. Under these conditions, which can be regarded as a typical condition of PLD experiment for common usages, M. Han *et al.* have used what is called the Knudsen layer (KL) analysis to deduce the idealized states of the gas just outside the KL [21]. One can summarize the quantities obtained by them with some trivial additional calculations performed by us as organized in Table 1.

**Table 1** Some of the parameters values used as an initial state in DSMC procedures

Number	Parameter Name	Parameter Value
1	Vapor Temperature (K)	628
2	Vapor Density (m <sup>-3</sup> )	$1.66 \times 10^{26}$
3	Flux Velocity (Km·s <sup>-1</sup> )	1.76
4	Vapor Flux (cm <sup>-2</sup> ·s <sup>-1</sup> )	$2.92 \times 10^{25}$
5	Flux Duration (ns)	4.4

Our DSMC procedures are started just after the gas cloud passed beyond the outside edge of KL. The spatial dimension of target substrate configuration is divided into cells and the cells are also subdivided into subcells of equally partitioned width. The ablated atoms are uniformly distributed within this space. The initial velocities of the ablated particles are distributed according to the well-known Maxwell-Boltzmann distribution function [22].

The ablated particles of the vapor cloud and the ambient gas may interact as an elastic hard spheres (HS) particles with a constant total collision cross section that is independent of the scattering angle and can be expressed as,

$$\begin{aligned}\sigma_{Tpq} &= \pi d_{pq}^2; \\ d_{pq} &= \frac{1}{2}(d_p + d_q),\end{aligned}\tag{1}$$

where  $d_p$  and  $d_q$  are the diameters of molecules  $p$  and  $q$ , respectively,  $d_{pq}$  is called the effective diameter of interacting molecules.

In the case of elastic scattering, the magnitude of the relative velocity in the center of mass frame of reference remains unchanged; this can be formulated mathematically as,

$$c_r^* = c_r,\tag{2}$$

where  $c_r$  and  $c_r^*$  are the pre-collision and post-collision relative velocities, respectively.

Based on the laws of conservation of energy and momentum, the post-collision relative velocity vector is evaluated stochastically and its components at the  $x$ ,  $y$  and  $z$  directions is given by,

$$\begin{aligned}c_{rx}^* &= c_r^* \cos \theta, \\ c_{ry}^* &= c_r^* \sin \theta \cos \varphi\end{aligned}\tag{3}$$

$$c_{rz}^* = c_r^* \sin \theta \sin \varphi,$$

respectively, where the angles  $\theta$  and  $\varphi$  are the scattering and azimuth angles. For HS model, these angles are uniformly distributed over the unit sphere. The azimuth angle  $\varphi$  can be easily selected from the range of interval  $[0, 2\pi]$  that it is uniformly distributed within this range. The angle  $\theta$  can be chosen from the interval  $[0, \pi]$  according to the following probability distribution function [23],

$$P(\theta)d\theta = \frac{1}{2}\sin \theta d\theta.\tag{4}$$

To insure that the contribution of the volume occupied by the molecules of dense gas is included to the final value of the quantity of collision rate (CR), the so-called consistent Boltzmann algorithm (CBA) has been adopted beside the main DSMC procedure [24].

### 3. Results and Discussion

In order to show and discuss our obtained results about the evolution modes of the quantity of collision rate (CR) in the context of propagation of laser-generated plume (LGP) into an ambient inert gas, we have presented the temporal snapshots of the quantity of CR covering the entire physical coordinate of the flow field. Figure 1 shows the temporal snapshots of the expended LGP of the Si target (solid line of red color) and the compressed He inert gas (solid line of green color) over the entire flow field. The snapshots denoted by the symbols a, b, c, d, e, f, g, h, i and j correspond to the elapsed times of values 0.0625  $\mu\text{s}$ , 2.5625  $\mu\text{s}$ , 15.8500  $\mu\text{s}$ , 20.5375  $\mu\text{s}$ , 22.8125  $\mu\text{s}$ , 28.1875  $\mu\text{s}$ , 34.8125  $\mu\text{s}$ , 41.4375  $\mu\text{s}$ , 45.1875  $\mu\text{s}$  and 50.0000  $\mu\text{s}$ , respectively.

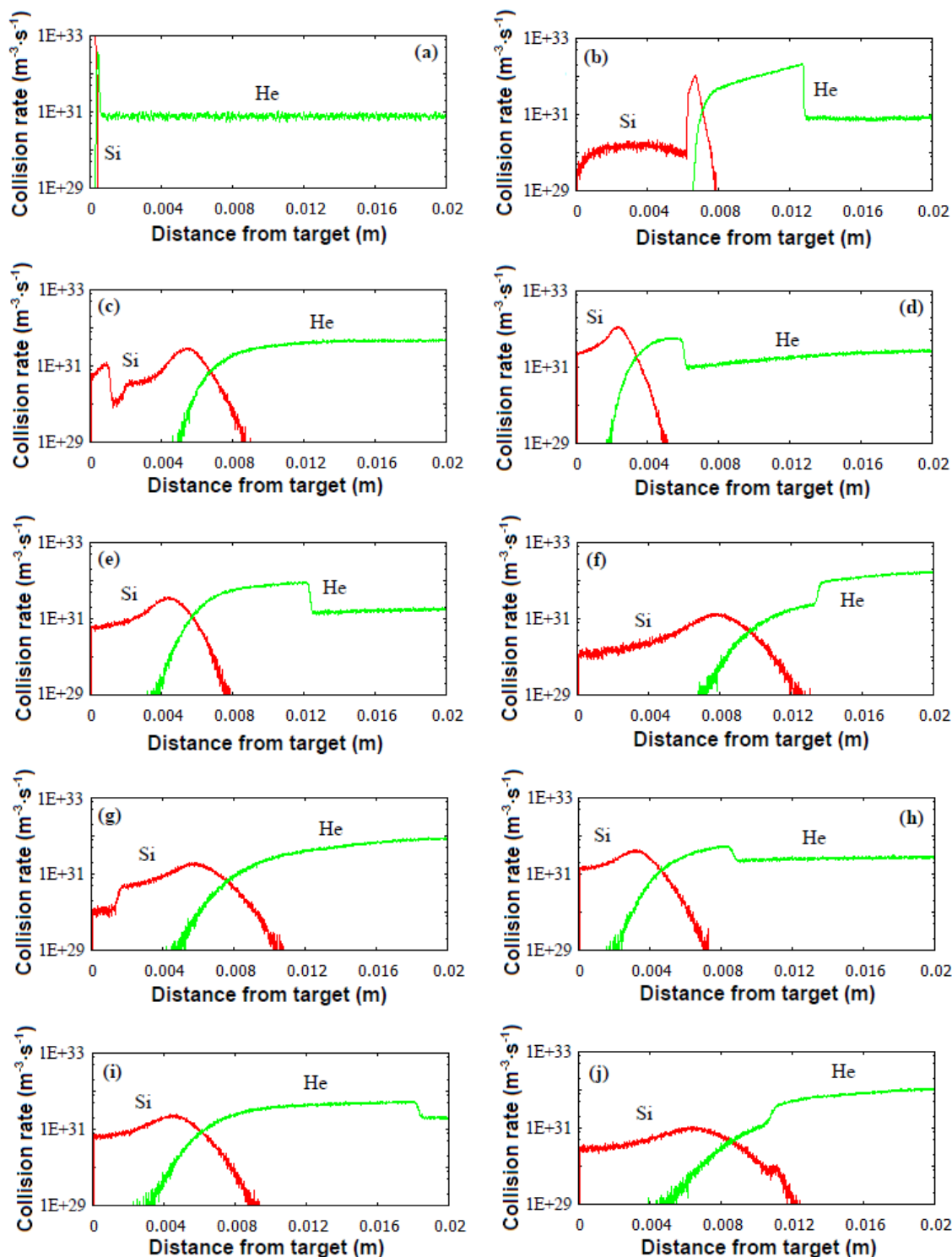
At an early phase of the expansion process, up to 0.0625  $\mu\text{s}$ , the density of Si vapor is still much higher than that of the ambient gas of He; this condition leads to a very high amount of CR between the particles of Si vapor.

However, the most portion of the flow field is dominantly packed by the molecules of the ambient gas of a relatively low density, in a consequence, a lower value of stationary CR between the molecules of He ambient gas can be obviously recognized, see Fig. 1 (a).

A few moments later, the propagation process proceeds at a high rate due to the large variations between the densities of interacting species; the vapor particles are expanded and the ambient gas molecules are compressed forward from the volume occupied by the vapor particles. As a result of these conditions, a high-density peak of the He gas is formed and start to propagate forward, see Fig. 1 (b). The formed peak of high-density can be seen as a compressed shock wave and has been previously investigated and reported in some reports [4, 10, 25].

However, formation of the so-called contact front, which is nothing but an enclosed fine layer containing a combination of the interacting species, can be clearly recognized. The contact front acts as a separated site between the ablated particles and the ambient gas molecules and identified by an imperceptible decline in the quantity of CR, as it can be distinctly seen in lots of snapshots of Fig. 1. The phenomenon of contact front formation and its oscillation behavior has been explored in different reports by following the evolution of some quantity else in the flow field such as density and number density [4, 20].

There is a noticeable action for the contact front happening during the period from 6.5250  $\mu\text{s}$  to 11.1625  $\mu\text{s}$ . One can be explained like that: after the position of contact front reaches to the farthest distance from the target at 6.5250  $\mu\text{s}$ , it didn't moves anymore and remains suspending at its position till the elapsed time become 11.1625  $\mu\text{s}$ , while the rest of ablated particles behind the contact front and the shock compressed wave of the ambient gas of He continue developing. During this period, the quantity of CR at the space close to the target becomes very low  $\sim 1\text{E}+29 \text{ m}^{-3} \cdot \text{s}^{-1}$  and this might refer to the sudden decline of number density of ablated particles. However, the plot showing this phenomenon has not showed here because it is suffered from some statistical fluctuation.



**Fig. 1** Temporal snapshots of the evolution of the quantity of collision rate (CR) of the expanded plume (LGP) of the Si target (solid line of red color) and the compressed He inert gas (solid line of green color) over the entire flow field. The snapshots of symbols a, b, c, d, e, f, g, h, i and j correspond to the elapsed times of values 0.0625  $\mu\text{s}$ , 2.5625  $\mu\text{s}$ , 15.8500  $\mu\text{s}$ , 20.5375  $\mu\text{s}$ , 22.8125  $\mu\text{s}$ , 28.1875  $\mu\text{s}$ , 34.8125  $\mu\text{s}$ , 41.4375  $\mu\text{s}$ , 45.1875  $\mu\text{s}$  and 50.0000  $\mu\text{s}$ , respectively.

Once the elapsed time of value  $11.1625\ \mu\text{s}$  is passed, the suspended contact front starts to propagates backward in the direction of the target reaching to the nearest point to the target and then changed its direction again and so forth, see plots (f) and (j) of Fig. 1. This oscillating behavior for the contact front has been noticed in more than one study [4, 10].

The existence of the substrate as a solid wall repulses the incoming shock wave of high-density peak leads to a successive reflections incident between the target and substrate. The forward propagating vapor front is also play an important role to induce and accelerate this phenomenon, see Fig. 1. The phenomenon of shock wave fashioning and reflection due to the existence of the ambient gas of He is of great importance determining the nPs formation progress and the final conditions of the so-deposited film.

There are some reports showed and suggested a featured model for the ablated particles condensation and gas-phase nPs nucleation process [26]. In accordance with the model of gas-phase nPs formation, the efficiency of cluster formation can be determined by the CR among cluster nucleus, ablated particles and inert gas molecules, which may be affected by the densities of the ablated particles, as well as the ambient gas molecules. However, we believe that the quantity of CR is one of the most important and comprehensive parameter that must be taken into account when dealing with the formation and growth process of gas-phase nucleated nPs. Furthermore think this quantity is governed not only by the densities of interacting molecules, but also by many other parameters such as molecules diameter and the mathematical relation between the masses of companied molecules.

#### 4. Conclusion

In this report, the so-called direct simulation Monte-Carlo (DSMC) technique is adopted as a powerful computational tool to investigate the transportation process of LIP into an inert gas. The quantity of CR is one of the most important and comprehensive parameter that must be taken into account when dealing with the formation and growth process of gas-phase nucleated nPs. To achieve our goals, the transportation process has been analyzed in terms of the quantity of CR as a main parameter. We have temporally and spatially traced the evolution of that quantity. A number of previously reported phenomena using different physical quantities such as number density, temperature and velocity, have been also confirmed and has found to be in excellent agreement with this work.

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