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Adiabatic, Non-Isothermal Plume Expansion into Vacuum in Terms of Knudsen Layer

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This article presents a theoretical analysis of laser plume formation in terms of Knudsen layer when a femtosecond laser pulse was irradiated on a thin gold foil in terms of gas dynamics equations. The laser spot was assumed to be noncircular laser spot radius, giving an ellipsoidal form of expansion. The profile of the plume will be discussed depending on the laser fluence and beam waist. Analytical solution of this expansion will be presented for the density of vapor plume.

Keywords: Femtosecond laser pulse, Thin film, Gold foil, Laser plume expansion, Gas dynamics.

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1. INTRODUCTION

Femtosecond laser pulse irradiated on a thin gold film can initiate the plume on the surface due the thermal expansion and high temperature gradient as a part of laser ablation. This thermal expansion (plume expansion) can play an important role in determining the Knudsen layer characteristics. Deposition and/or ablation can be of great interest in vast fields of physics, e.g., thin film deposition, material evaporation. A clear understanding of the physics defining the plume expansion is of fundamental importance in such applications of laser ablation as thin films deposition and mass spectrometry. Quality of the deposited films in pulsed laser deposition [1, 2] and quality of mass spectra in matrix assisted laser desorption ionization [3, 4] are critically dependent on the velocity distributions of the ejected species and their spatial distributions in the plume. Theoretical and computational investigation of the laser ablation phenomenon is challenging due to the complex collective characters of the involved processes that occur at different time and length scales [5, 6].

Plume behavior can be described as a two-stage process: a "violent" plume expansion due to the absorption of the laser beam energy (during the laser pulse) followed by a fast adiabatic expansion in the ambient gas (after the end of the laser pulse). Plasma plume may last a few microseconds and may have densities 10 times lower than the solid densities at temperatures close to the ambient temperature [7]. Many theoretical models can be found in the literature concerning the plume expansion, some have been in vacuum and others in gas atmosphere. A clear understanding of the physics defining the plume expansion is of great importance in many laser-matter interactions and laser applications. As soon as the local Knudsen number in the ablated plume becomes high enough to provide the rarified conditions, the long-term plume evolution can be modeled using one of the methods of the rarified gas dynamics [8].

Laser plume dynamics will be discussed based on gas dynamics equations into vacuum in terms of Knudsen layer depending on the initial dimensions of the plume and the laser fluence. Knudsen layer thickness is assumed to be smaller than the laser spot size of femtosecond pulse duration. The expansion into a vacuum will be presented by three dimensional analyses with an assumption that an ellipsoidal expansion is initially formed. The 3D profile will be presented for two values of laser fluence. The density of vapor plume will be given for many values of noncircular laser spot radius. Analytical solution of the expansion of plasma plume described by gas dynamic equation for adiabatic expansion on the basis of Anisimove et al. [9] was solved numerically for the profile of vapor density.

2. LASER PLUME DYNAMICS

In this section we are going to discuss theoretically the equations of laser plume gas dynamics. Equation (1) represents the surface temperature distribution for the plume of gold film irradiated by Gaussian femtosecond laser pulse [10]:

$$T(r,t,z)=\frac{F_{\max}}{K}\left(\frac{\kappa}{\pi}\right)^{1/2}\int_0^t\frac{p\left(t-t'\right)d^2\left(t'\right)dt'}{t'^{1/2}\left(4\kappa t'+d^2\left(t'\right)\right)}\times$$
$$\times\exp\left[\frac{-z^2}{4\kappa t'}-\frac{r^2}{4\kappa t'+d^2\left(t'\right)}\right] \qquad (1)$$

with F_{max} is the laser peak flux density. The symbols given in the above equation is for thermal properties of gold. K is the thermal conductivity, κ is the thermal diffusivity $\kappa = K/\rho\cdot c$, ρ is density, c is heat capacity, $p(t)$ is the normalized pulse magnitude ($0 \le p(t) \le 1$). Table 1 summarize the thermal properties of gold used in the numerical simulation of this article.

Table 1 – Parameters of gold used in the simulation

Property	κ	K	ρ	w
Definition	Thermal diffusivity	Thermal conductivity	Density	Thermal wave speed
Numerical value	1.2×10^{-4} m²/s	315 W / (m.K)	19.3×10^3 kg/m³	1.9085 m/s

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Equation (1) was solved numerically for laser fluence of (13.4 & 23.4 J/m^2) and for laser pulse duration of 96 fs, as shown in Fig. 1.

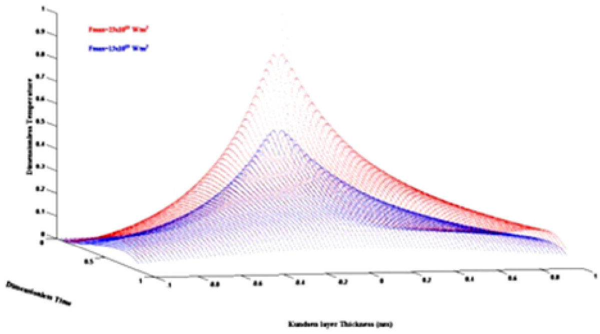


Fig. 1 – Illustratese the expansion of plume (in terms of temperature distribution of vapour plume) for two different values of laser fluence of 13.4 & 23.4 J/m^2 as a function of Kundsen layer thickness

The expansion of the plume was taken into account the effects of laser fluency in order to show how this can influence the expansion in terms of Knudsen layer thickness which had been evaporated by thermal effect of laser pulse. Laser ablation has to concider this result in its application.

Laser spot radius was found to be effective on the plume expansion as shown in Fig. 2. Two values were used in the numerical simulation. As can be seen from the graph, higher value of laser spot gives higher temperature distribution and expansion.

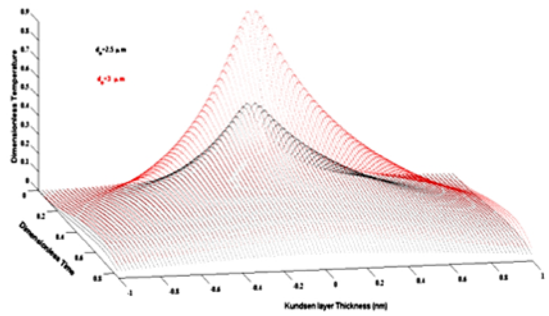
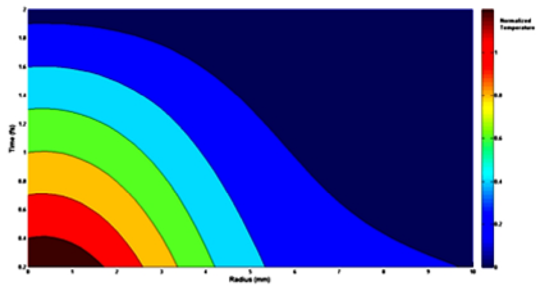
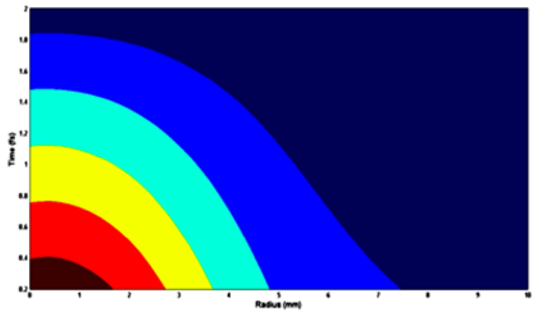


Fig. 2 – Representce the influence of laser spot diameter on the temperature distribution of the plume in terms of Knudsen layer thickness

Fig. 3 shows the numerical simulation of temperature distribution on the surface of the gold film irradiated by (96 fs) laser pulse duration and fluence of (13.4 Jm^{-2}) by solving the characteristic equation of temperature distribution given in equation (1). The heated zones of the gold film were drawn in terms of time – varying peak temperature of heat transport in the gold film during laser irradiation. Temperature of the heated zones were found to be symmetric when a Gaussian laser pulse was assumed in the numerically solved equation (1). The figure shows the influence of laser spot diameter on the temperature distribution as in Fig. 3a is for laser spot diameter 2.5 mm, while figure 3b is for 3 mm.



a



b

Fig. 3 – Time-varying of temperature distribution of plume expansion for Knudsen layer evaporation when irradiated by femtosecond laser pulse. (a) laser spot diameter 2.5 mm and (b) laser spot diameter 3 mm

The density of the plume will increase as long as the laser irradiation is on and the gold film is being ablated until a shielding is formed on the surface. As the plume expands the electron density decreases allowing again the ablation to occurs on the target and so raising the density of electron. To investigate the electron density, the plume expansion can be considered to be expanded thermodynamically adiabatic as in the equation:

$$T[X(t)Y(t)Z(t)]^{(\gamma-1)} = constant \qquad (2)$$

where T is the temperature of the plume, $X(t)$, $Y(t)$ and $Z(t)$ are the dimension of the plume at a time t and γ is the ratio of specific heat capacity at constant pressure and volume. Due to this expansion, the plasma density N is decreasing, as well as its temperature T,

$$T \propto N^{\gamma-1} \qquad (3)$$

To model the plume dynamics, Anisimov proposed an isentropic equation for a spatially constant temperature with a Gaussian density profile N at any point and any time as [9]:

$$N(x,y,z,t) = \frac{N_T}{I_1(\gamma)X(t)Y(t)Z(t)} \exp\left[-\frac{x^2}{X(t)^2} - \frac{y^2}{Y(t)^2} - \frac{z^2}{Z(t)^2}\right]^{1/\gamma-1} \qquad (4)$$

where N_T is the total number of evaporated particles at the end of the laser pulse , with plume expansion in three orthogonal directions, and $I_1(\gamma)$ is an integral expressed in terms of the Γ function [9].

Equation (4) gives the density of all species in the plasma plume expansion, i.e, atoms, ions, neutrals, small clusters, etc. The density profile of these species will be found by solving numerically equation (4), and to estimate the total number of evaporated particles at the end of laser pulse.

Fig. 4 represents the numerical solution of equation (4) for the plasma plume expanded from the surface of gold target assuming isentropic case. The total ablated particles density was plotted in terms of ablation depth Z and laser spot diameter. The ablated mass in unit volume of the target was found to be dependent on the laser beam waist. We have assumed that a minimum energy was delivered by the laser source in order that evaporation occurs. The melted surface layer was calculated from the laser pulse duration and the thermal diffusivity of gold as:

$$L_{th} = \sqrt{2D\tau_p} \qquad (5)$$

with D is the thermal diffusivity of gold (1.2×10^{-4} m^2/s), and τ_p is the laser pulse duration (96 fs).

The target temperature was assumed at (300 K), and laser fluence (13.4 Jm^{-2}). The ablation depth was calculated from the following equation:

$$\delta Z = (1-R)\left(\frac{F_l - F_{th}}{h}\right) \qquad (6)$$

where R is the gold reflectivity, F_l is the laser fluence for ablation, and F_{th} is the threshold energy of evaporation, given by:

$$F_{th} = \left(\frac{\rho C_T \Delta T L_{th}}{1-R}\right) \qquad (7)$$

Where ρ is the density of gold, C_T is the specific heat capacity of gold (12.9 J/kg·K), and ΔT is the temperature difference between melting temperature of gold (1337 K) and ambient temperature.

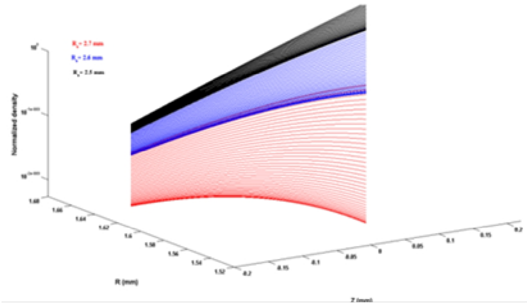


Fig. 4 – Density of evaporated particles from the target surface as a function of ablation depth and at different laser beam waist

3. CONCLUSIONS

Laser plume expansion from thin gold film was investigated in this article in terms of Knudsen layer for adiabatic, non-isothermal case of gas dynamics. The results showed that the plume expansion was completely dependent on the laser pulse parameters, the pulse duration, fluency, and beam waist. This dependence had been shown implicitly in the plots given. The analytical equations were solved numerically for special case as spatially constant temperature of the laser plume.

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