Langmuir probe investigation of plasma expansion in femto- and picosecond laser ablation of selected metals

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Abstract. A time resolving Langmuir probe was used to study the plasma plumes produced by the ablation of Ag, Ni and Al targets with laser pulses of different pulse durations (0.2 – 10 ps). These metals were chosen because their electron-phonon relaxation times, $\tau_{e-ph}$, are of the order of the pulse durations used. The time of flight (TOF) signals have been used to establish the threshold fluences and plume expansion dynamics of the laser produced plasmas for the different pulse durations. The angular dependence of the magnitude of the ion flux was analysed on the basis of Anisimov’s self-similar model of the plasma expansion. The amount of charge in the ablation plume is compared for the different pulse durations.

1. Introduction

The advantages of ultrashort pulses in laser machining arise mainly because the laser energy is deposited before thermal equilibrium is reached between the electrons and the lattice. Hence, the electron-phonon relaxation time, $\tau_{e-ph}$, is a key parameter. It determines the amount of time required for the lattice to respond to the laser pulse and is highly dependent on the strength of the electron-phonon coupling constant, $g$, of the material [1,2]. The extent of the non-equilibrium reached between the electrons and the lattice determines the rate of the lattice heating. For pulse durations shorter than $\tau_{e-ph}$, the electrons are initially heated to very high temperatures, compared to the cold lattice, and therefore the subsequent lattice heating rate is high. For pulse durations longer than $\tau_{e-ph}$, the extent of the non-equilibrium reached between the electrons and the lattice subsystem is lower and thus the lattice-heating rate is reduced. As a result, for ultrashort pulses, there are more photomechanical effects involved in the ablation process and there is less thermal damage. This is contrary to long pulses, where the heating rate of the lattice is lower and the ablation is mainly due to thermal processes. A possible interaction between the trailing edge of the beam and the emerging (nascent) vapor cloud could also occur.

The effect of the pulse duration on the ablation process is observed by investigating the ablation rate, which can be associated with the ablation efficiency, and by analysing the plasma expansion, particularly the number of ionized species in the plume and the expansion ratio, $\kappa$, which defines the shape of the plume.

Langmuir ion probes, which enable the ionic component of the ablation plume to be studied relatively easily, have been used extensively to investigate the plasma parameters of laser produced ablation plumes from metal targets [3,4,5,6,7]. The behavior of expanding laser produced plasma
plumes has previously been explained using the self-similar, adiabatic model of plume expansion developed by Anisimov et al [8,9]. Strictly, this model applies to the expansion of a neutral gas but it has been shown before that, with the correct choice of parameters, the model can be used to describe the dynamics of laser ablation plasmas [10].

2. Experiment

The ablation was performed in 10⁻⁶ Torr using linearly polarized pulses from 200 fs to 10 ps duration from a commercially available Ti-sapphire laser (λ = 775 nm). Adjusting the distance between the gratings in the compressor stage allowed the pulse duration to be varied. A repetition rate of 50 Hz was used for all experiments. A 0.011 cm² flat Langmuir probe, biased at –25 V was positioned 2.7 cm from the target, perpendicular to the plasma flow direction. The ion current signal was determined by measuring the voltage response across a load resistor. The angular position of the probe relative to the target normal was varied. The experimental set-up is described in detail in earlier works [11,12]. Typical time of flight signals are presented in [12], where it has been shown that metal samples are normally covered with a thin layer of surface impurities. These impurities are removed by the first few laser pulses and signals are acquired directly after cleaning to avoid any surface recontamination.

The metals investigated in this paper were chosen to have an electron-phonon relaxation time greater than, and lower than, the laser pulse durations. Their properties are listed in Table 1 where values for τₑ-ₚₙ have been extracted from [1], using a value for the electron temperature of 10⁴ K.

| Metal | g [W m⁻³K⁻¹] | τₑ-ₚₙ [ps] |
|-------|-------------|------------|
| Ag    | 3.1 × 10¹⁶  | 20.0       |
| Ni    | 3.6 × 10¹⁷  | 7.0        |
| Al    | 4.9 × 10¹⁷  | 1.5        |

Table 1. Strength of the electron-phonon coupling constant, g and electron-phonon relaxation time, τₑ-ₚₙ, for the selected metals.

3. Results

In order to highlight the effect of the pulse duration on the ablation process, the ablation rate, the plasma formation threshold and the plasma expansion dynamics were investigated as a function of the pulse duration.

3.1.1. Ablation rate

The effect of the pulse duration (200 fs and 10 ps) on the ablation rate, expressed in terms of the depth removed per pulse, for the different metals investigated is presented in figure 1.

Figure 1. Ablation rate curves for (a) Ag, (b) Ni and (c) Al.

The ablation threshold is the same for pico- and femtosecond laser pulses and there are two distinct ablation regimes following a logarithmic dependence [11]. The influence of τₑ-ₚₙ can be explained in the following way. In the case of Ag (figure 1), both pulses are shorter than the electron-phonon relaxation time, τₑ-ₚₙ (Ag) = 20 ps, and the heating rate of the lattice is almost the same. There is a slight reduction in the ablation rate for the 10 ps pulses. For Ni and Al (figure 1), the shorter pulse is well below τₑ-ₚₙ and so the heating rate of the lattice is much higher. This results in a more concentrated laser energy deposition and the lattice heating is developed under conditions of inertial stress confinement leading to material removal mainly by photomechanical effects [2]. Therefore,
more efficient ablation process is observed for 200 fs as compared to 10 ps pulse, where an interaction between the trailing edge of the beam and the emerging (nascent) vapor cloud could also reduce the ablation rate.

Figure 2 shows the ion energy distribution for the three metals at a peak fluence of 0.66 J/cm². The results show an increase in the ion energy for the 10 ps pulses, which is more significant for Ni and Al, compared to Ag, due to their faster lattice response times. This observation is consistent with the smaller ablation depth, leading to higher energy per unit mass in the ablated material, for laser pulses shorter than \( t_{\text{on}} \). Moreover, previous experiments [12] have shown that the plasma expansion speed is of the order of \( 1 \times 10^6 \) cm/sec. Therefore in 10 ps the vapor cloud would have traveled a distance of the order of 100 nm. The optical penetration depth of Ni, for example, is \(~15\) nm for the present conditions, and so it is possible that some of the laser energy in the trailing edge of the beam is absorbed by the emerging vapor/plasma leading to more energetic ion emission.

3.1.2. Laser induced plasma threshold determination
In order to determine the threshold fluence for the onset of the laser induced plasma, TOF signals were obtained for a range of fluence values as outlined in [12]. The total charge flux, measured at a fixed probe position (\( 5^\circ \) off the target normal) depends on laser fluence. Figure 3 shows that the threshold for plasma formation is not dependent on pulse duration and the behavior of the experimental data suggests a log dependence with two regimes present, as observed elsewhere [7].

The Ni and Al data shows that there is significantly more charge collected for the longer pulses, where a higher lattice temperature and the vapor-laser interaction are responsible for the ionization of the ablated material.

3.1.3. Plasma expansion dynamics
The angular dependence of the magnitude of the ion flux is analysed on the basis of Anisimov’s self-similar adiabatic model of the plasma expansion [8]. The model describes the plasma expansion based on the conditions established at the end of the laser pulse, where the plasma exists as a thin semi-ellipsoidal layer on the target surface. The expansion is driven by pressure gradients, which depend on the thickness of the initial plasma layer, and can be characterised by a quantity known as the expansion ratio, \( \kappa \), which is the ratio of longitudinal and transverse semi-axes of the semi-ellipsoidal plume in the asymptotic limit. This is a measure of the forward peaking of the plasma plume. The pressure gradients are higher for thinner initial plasmas resulting in higher \( \kappa \) values. The angular
dependence of the magnitude of the ion flux is presented in figure 4 for the different materials at the peak fluence values and pulse durations shown.

In terms of the expansion dynamics, there is little difference between the 10 ps and 200 fs pulses for Ag, as evidenced by the values obtained for the expansion ratio, $\kappa$ (figure 4). The data obtained for Ni and Al shows that the 200 fs plumes are more forward peaked than the 10 ps plumes. This result suggests that the 10 ps pulses produced thicker initial plasmas than the 200 fs pulses, for these two metals. This is consistent with the fact that the laser energy deposited into the electronic system can be high enough for a solid dense plasma formation at high fluences in the case of pulse durations shorter than $\tau_{\text{e-ph}}$ resulting in a more forwardly peaked plasma plumes.

![Figure 4. Angular plasma dependence for Ag, Ni, and Al.](image1)

Using the data presented in figure 4 the total amount of charge per pulse is determined by integrating over the half solid angle subtended by the plume, approximately $2\pi$. For Ag, there is 2 times more charge detected in the 10 ps plasma compared to the 200 fs case. In Ni and Al, the increase in the amount of charge is more significant, a 5.6 fold increase for Ni and a 3.3 fold increase for Al.

When collected by the probe at a fixed fluence value and angular position, the charge flux is presented as a function of the pulse duration (figure 5) for the different targets. A sharp increase is observed at a value around 4 ps for Ni, and around 0.8 ps for Al, close to the estimation of the respective $\tau_{\text{e-ph}}$ values. No such increase could be observed for Ag on this scale.

![Figure 5. Ion flux versus incident pulse duration for Ag, Ni, and Al.](image2)

4. Conclusions

The threshold for plasma formation coincides with the ablation threshold for Ag, Ni and Al. The charge flux for 200 fs and 10 ps pulses displays a logarithmic dependence on the laser fluence with two separate regimes, similar to what was observed for the ablation rate. A weaker non-equilibrium between electrons and phonons during the 10 ps pulses reduces the ablation rate, especially for Ni and Al. The amount of charge present in the ablation plumes increases for the 10 ps pulses and the ions are more energetic than for the 200 fs case. These effects are more pronounced for Ni and Al, because they have shorter lattice response times than Ag. For Ag, where both pulse durations are less than $\tau_{\text{e-ph}}$ (Ag) = 20 ps, there was very little difference in the expansion dynamics for the 10 ps and 200 fs plasma plumes. However, for Ni and Al, the 200 fs plasmas were more forward peaked than the 10 ps plasma. This suggests that the 10 ps pulses produced thicker initial plasmas than the 200 fs pulses, for
these two metals. Moreover, an increase in ion emission is observed at a value close to the estimation of the respective $\tau_{\text{e-ph}}$ values.

References
[1] Demsar J, Averitt R D, Kabanov V V and Mihailovic D 2003 Phys. Rev. Lett. 91 027401
[2] Ivanov D S and Zhigilei L V 2003 Phys. Rev. B 68 064114
[3] Hansen T N, Schou J and Lunney J G 1999 Appl. Surf. Sci. 139 184-187
[4] Toftmann B, Schou J, Hansen T N and Lunney J G 2002 Appl. Surf. Sci. 186 293-297
[5] Thestrup B, Toftmann B, Schou J, Doggett B and Lunney J G 2002 Appl. Surf. Sci. 197 175-180
[6] Amoruso S, Wang X, Altucci C, de Lisio C, Armenante M, Bruzzese R and Velotta R 2000 Appl. Phys. Lett. 77 3728-3730
[7] Ye M and Grigoropoulos C P 2001 Journal of Appl. Phys. 89 5183-5190
[8] Anisimov S I, Lukyanchuk B S and Luches A 1996 Appl. Surf. Sci. 96-8 24-32
[9] Anisimov S I, Bauerle D and Lukyanchuk B S 1993 Phys. Rev. B 48 12076-12081
[10] Hansen T N, Schou J and Lunney J G 1999 Appl. Phys. A 69 S601-S604
[11] Mannion P T, Magee J, Coyne E, O'Connor G M and Glynn T J 2004 Appl. Surf. Sci. 233 275-287
[12] Mannion P T, Favre S, O'Connor G, Doggett B, Lunney J G and Glynn T J 2005 Proc. SPIE 5827 457-466