Measurements of ions and neutral particles in a plume generated by laser ablation of copper

J B de Matos¹, C A B da Silveira² and N A S Rodrigues²

¹Technological Institute for Aeronautics-ITA, P.O. Box 6050, 12.228-900, Brazil
²Institute for Advanced Studies-IEAv, P.O. Box 6044, 12.231-970, Brazil

E-mail: julianamatos_ita@yahoo.com.br

Abstract. This paper presents the results for the measurement of neutral atoms and ion density in a Cu plume generated by laser ablation using electrostatic probes and a piezoelectric sensor. A tripled frequency Nd YAG laser (355 nm, 2 kHz, 23 ns, 10 kW) has been used to produce a plasma plume of copper in vacuum atmosphere (10⁻⁴ Torr). It will be shown the effect of laser fluence on the relative densities of neutrals and ions departed from target. It was also observed that the ion density increases with laser fluence while neutral atoms remain roughly constant in the range of our experimental conditions. It was measured a drift velocity around 3-7 km/s and translational temperature in the range of 2.5-4.4x10⁴(K).

1. Introduction
The ejection of particles from a solid or liquid surface by laser pulses is well known as laser ablation. In this process short laser pulses with high power densities lead to the formation of a plume made of neutral atoms, ions, clusters and droplets of liquid material [1]. Laser ablation is widely present in different fields of science and industry and a number of techniques have been employed over decades to analyze the plasma plume composition and dynamics, such as electrostatic probes [2], optical emission [3], spectroscopy [4], and spectrometry [5] among others. In this paper, we present results of study of relative population of neutral and ionized atoms of copper samples irradiated by Nd:YAG laser using pressure transducer device and electrostatic probes.

2. Experimental setup
A plasma plume was produced by laser ablation of copper samples inside a vacuum chamber kept at 10⁻⁴Torr. Nd:YAG laser pulses were focused onto the metallic target at 45° as sketched in figure 1. In order to measure total particles of plume, a pressure transducer, PVDF, was used at 150 mm from the target surface. The polyvinylidene fluoride (PVDF) film is a ferroelectric polymer which has pyro- and piezoelectric properties and gives information about plume drift velocity and temperature. Two parallel electrostatic plates were placed at 140 mm far from the target surface. For those distances no internal mechanism of plume acceleration was detected, it means that it remained in thermal equilibrium and expands according to a Maxwellian Velocity Distribution. A power supply (up to 1kV) was used to bias the electrode plate with negative voltages to extract ions of the plume.

Two experiments were performed, one using only PVDF to measure drift velocity and translational temperature of all particles of the plume, with values corresponding to 3-7 km/s and 2.5-4.4x10⁴ K, respectively. In order to investigate the ratio of neutrals particles to ions in the plume, another
experiment was accomplished with PVDF and simultaneously a pair of electrodes biased with different polarization voltages. The PVDF electrical signal was recorded in an oscilloscope for each polarization voltage as function of the incident laser fluence (20 up to 40 J/cm²). In order to prevent degradation on the copper surface by prolonged radiation, the target was constantly in movement. Each measurement set was performed only after shot surface cleaning. Copper target was attached in controlled XY table to insure suitable velocity and precision to produce a plume with same characteristics for all laser ablation experiments. Figure 2 shows two typical PVDF curves concerning the effect of potential applied between the electrostatic plates.

**Figure 1.** Experimental setup.

**Figure 2.** PVDF signal with polarized plate (solid line curve) and without potential between plates (dashed line curve).

Nd:YAG laser was used due to some special characteristics, in particular because it emits in low energy and high repetition rate, as shown in table 1. The laser beam illuminated a 25 µm diameter spot size by a set of two 100 mm focal length lens. It is well-known that not only laser characteristics but
also chemical and physical properties of target and surrounding atmosphere play a very important role in laser ablation experiments. Cu samples were systematically polished and washed with acetone prior to the ablation experiments in order to remove any surface roughness, impurity or contamination.

### Table 1. Nd:YAG laser parameters for laser ablation experiments

| Characteristics          | Value | Unit |
|--------------------------|-------|------|
| Wavelength               | 355   | nm   |
| Repetition Rate          | 2     | kHz  |
| Pulse Width              | 23    | ns   |
| Pulse Energy             | 124-240| µJ   |
| Peak power               | 5-10  | kW   |
| Intensity                | 0.8-1.7| GW/cm² |
| Fluence                  | 20-40 | J/cm²|

3. Results and discussion

It was shown that the PVDF electrical signal is proportional to the linear moment transferred by particles of the plume to the sensor surface. The time-of-flight (TOF), when triggered by laser pulses, is given by [6]

\[
S(t) = \frac{1}{t^5} \cdot \exp \left[ -\frac{m}{2K_B \cdot T} \left( \frac{l}{t} - v_0 \right)^2 \right]
\]

where \( l \) is the distance between target and sensor, the particles arrive to the sensor surface at the instant \( t \) with velocity \( v = \frac{l}{t} \), \( m \) being the mass and \( K_B \) Boltzmann constant. We assume the plume as a bunch of free particles far from Knudsen layer (150mm), and it expands according to Maxwellian Velocity Distribution with a drift velocity \( v_0 \) and translational temperature \( T \). By adjusting the equation (1) to the PVDF electrical signal it is possible to infer the drift velocity and translational temperature, as it is shown in figure 3.

Some summarized results of \( v_0 \) and \( T \) are presented in table 2 as function of laser fluence.

### Table 2. Drift velocity \((v_0)\) and translational temperature \((T)\) measured with PVDF sensor as function of fluence in laser ablation experiments of copper target.

| Fluence (J/cm²) | \( v_0 \) (km/s) | \( T \) (10⁴K) |
|-----------------|------------------|------------------|
| 20              | 3                | 3.2              |
| 27              | 5.3              | 2.5              |
| 40              | 7.05             | 4.4              |

The relative abundance in ion density and neutral atoms present in laser ablation depend strongly on laser fluence. PVDF sensor was employed together with electrostatic plates to show how the laser energy density may affect directly on the ratio of ions to neutral atoms. Results present in figure 4 were obtained taking into account the maximum PVDF electrical signal and its dependence with plate polarization for different fluences. Each data represents the maximum peak of PVDF signal and all of them correspond to an average of 64 curves measured three times. As expected, it was observed that
the amplitude of PVDF electrical signal tends to decrease as function of polarization in the electrode plates. Despite fluctuation observed in first data for 40 J/cm², it is evident a decreasing and it tends to a value of 90% of total particles as plate polarization increases up to 650V. It is possible to infer that the others 10% of the plume are made of neutral particles which are not affected by plates potential, as one can notice in the plateau for all curves. The same trend is observed for fluences between 20 J/cm² and 27 J/cm², and the reduction is in the range of 70% and 80% of total particles of the plume up to 30 V and 250 V applied, respectively. To our experimental conditions we conclude that ion density increases with fluence while neutral atoms remain roughly the same.

**Figure 3.** The theoretical curve (dashed line) was adjusted to PVDF signal (solid line) to obtain \( v_0 \) (m/s) and \( T(K) \).

**Figure 4.** Effect of laser fluence in laser ablation experiments by using PVDF as a pressure sensor and electrostatic plates.
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