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Characterization of laser induced tantalum plasma by spatio-temporal resolved optical emission spectroscopy

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ABSTRACT

The spatial and temporal behavior of the tantalum plasma produced in air by third harmonic Nd:YAG laser (0.6 GW cm^{-2}) has been studied using optical emission spectroscopy. Excitation temperature and electron density have been estimated from the analysis of spectral data as well as their spatio-temporal evolutions. As the delay time increases from 400 to 2000 ns, the excitation temperature has found to decrease from 10,000 K to 7900 K. The value of N_e decreases continuously from 4×10^{18} near the zero position to 5×10^{17} in a linear approach along with the propagation axe of the plasma plume. Laser-supported consumption wave regime has been proposed to explain the propagation of the plasma.

1. Introduction

Laser Induced Plasma Spectroscopy (LIPS) is one of the analytical and diagnostic techniques that has been really boosted recently [1,2], depending on its original concept based on induced spectral emission after exciting a small quantity of the material by high power laser pulses. The emitted radiations were recorded and analyzed by optical emission spectroscopy (OES) system.

In order to investigate and monitor the emission spectra with time resolution of nanoseconds, an advanced triggered and gated intensified CCD camera in the nanosecond regime is needed. The time resolved LIPS has become the most popular way to improve the performance of the laser induced plasma spectrometry [3,4].

Tantalum is a rare metal, widely used in electronic components such as superconducting transition edge temperature sensor [5], capacitors and resistances in oxide form used in manufacturing a hearing devices [6]. Tantalum layers are used as a robust coating of surfaces requiring a high resistance against wear, corrosion as well as immunity to chemical and biological attack such as surgical tools and implants [7–9].

A variety of techniques including sputtering [10,11], e-beam evaporation [12], plasma-enhanced atomic layer deposition [13] and laser ablation [14–16] have been used for tantalum and its compounds thin films deposition.

In order to get a better control on the pulsed laser deposition processes, a spatio-temporal study by optical emission spectroscopy of Ta plasma in air, could be considered as a first step and it will be essential to characterize the plasma plume during the films deposition in further detailed study. The most important plasma parameters are excitation temperature and electron density. These parameters depend highly on the experimental conditions such as laser wavelength, laser fluence, ambient gas pressure, target materials and the geometrical set up of the collection optics [1,2].

Torrisi et al. have studied the Ta plasma in vacuum by nanosecond laser at different wavelengths and fluencies, where they explore the emission of neutrals and ionic species of the Ta plasma [17,18]. They found that the electron temperature and density of the plasma are about 50 eV and 10^{18} cm^{-3} respectively. In other work, Torrisi et al. have studied the spatio-temporal behavior of laser-generated pulsed plasmas at 1064 nm of Ta [19], by means of Langmuir probes, ion collectors, and ion energy analyzers. The electron density value in their work is similar to what it is obtained in our work.

Novodvorsky et al. have studied the erosion plume of tantalum targets in vacuum formed by excimer laser (308 nm) [20]. They studied the electron temperature distribution over the plume using Langmuir probe and optical emission spectroscopy. Their results showed that the excitation temperature of the Ta plume is not uniform and it has a maximum value in front of the plume.

This article presents, up to the best of our knowledge, the first spatial and temporal study of the tantalum plume produced in air. Intensity, excitation temperature, and electron density distribution as a function of space and time were investigated from a large number of optical emission spectra taken at different points of the plume along the propagation axe with different delay times.

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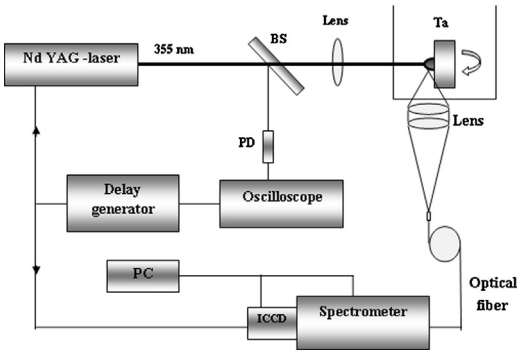

Fig. 1. Schematic diagram of the experimental set up.

2. Experimental set up

The experimental set up employed in this work is shown in Fig. 1.

The experiments were carried out in air with third harmonic (355 nm) of Q-switched Nd:YAG pulsed laser (Quantel YG series 820) with a 10 ns pulse duration at a repetition rate of 10 Hz. The laser beam was focused by a quartz lens of 75 cm focal length onto the rotated Ta target (CRM, 99.95% purity) surface at an incidence angle of 45°. The Ta target holder rotates (maximum speed: ~30 rev/min) during experiment in order to avoid non-homogeneous ablation for each pulse. A pyroelectric smart sensor detector (LM-10 LP Coherent Labmaster) was used to measure the laser pulse energy which is 12 mJ, with fluctuation of approximately 5%. The laser fluence at the focused spot area is 6 J cm^{-2} and a pulse power density or irradiance is 0.6 GW cm^{-2} on the target surface.

The emitted light from the plume was optically imaged 1:4 onto the optical fiber bundle entrance at right angle to the normal of the Ta surface (see Fig. 1). Imaging was done by using a confocal optical system consisted of a diaphragm and two quartz lenses (f_1 = 5 cm, f_2 = 20 cm). The emission light of the plasma collected by the optical system into the optical fiber was dispersed by high resolution spectrometer (SCIENCETECH-9150) with 2400 g mm^{-1} grating, coupled with an intensified CCD camera (Andor i-Star DH740-18F-03) to detect the dispersed spectral emission. The entrance slit width of the spectrometer was set at 50 μm.

Stanford DG 535 pulse delay generator was used to trigger the laser and the ICCD camera. The ICCD camera and the Nd:YAG laser were synchronized by using a fast photodiode (SV_2 – THORLABS). A portion of the laser beam is reflected by glass slide to the photodiode; its signal was overlapped with the signal resulting from the ICCD camera and monitored by an oscilloscope (Tektronix TDS 3054C).

The spatial study was done by moving the entrance of the optical fiber bundle (mounted on *X*, *Y*, *Z* micro-displacement) in the plane of the image, which permit to study the plasma along its axial direction.

The temporal study was controlled by the ICCD software (Andor Solis), the gate width was fixed at 300 ns in order to get a good signal to noise ratio necessary for a precise measurement of the spectral line intensity, this gate width will produce an averaged excitation temperature value over the gate width as we add different signal intensities during the gate but it will produce a better reproducibility of the signals. Due to the high intensity of the continuum emission in the initial stage of the expansion plasma, the temporal study was done starting from 400 ns delay. Each spectrum was background corrected and 200 spectra were taken for each measurement. All spectra were normalized with respect to the spectral response of the grating and to the spectral response of the ICCD.

The excitation temperature and electron density values reported in this work are calculated from spectra measured by integrating the intensity over the line-of-sight in the plasma perpendicular to the target on the center of the laser spot. No Abel inversion or intensity deconvolution was applied, so the resultant excitation temperatures and electron densities are population-averaged values. This effect is more important near the surface where the plume is extremely inhomogeneous.

3. Results and discussion

3.1. General intensity profile

The emission spectrum of the Ta plasma was recorded in the range of 380–600 nm after 400 ns from the plasma creation. It is dominated by the emission of Ta I, Ta II, no more Ta ion states were expected due to the low laser irradiance [17–19], as well as the used delay value. The contribution of air atoms is not notably appeared in the emission spectra.

Cristoforetti et al. [21] have mentioned that at laser irradiance less than 8×10^8 W cm^{-2}, the plasma begins in the target vapor and it expands in air like a piston, simply pushing out the air molecules so there is almost no mixing between air atoms/molecules and target atoms. Therefore, the formed plasma is mainly composed by the target species. Many authors [22,23] have discussed the dynamics of formation and propagations of the Laser Supported Absorption Wave (LSAWs). They are roughly, in the low and medium laser irradiance, divided into two regimes: laser-supported consumption wave (LSC) and laser-supported detonation wave (LSD). These two regimes are not always easy to be distinguished. However, many parameters such as irradiance, wavelength, target material and ambient atmosphere significantly influence these regimes [1,2]. In the first regime (LSC) the layer of compressed background gas in contact with the vapor plume remains transparent for laser radiation pulse, thus the laser beam propagates through this layer and is absorbed by the vapor plume under the shocked gas. For the second regime (LSD), the shocked background gas absorbs laser radiation. Then secondary breakdown occurs in this layer leading to high ionized gas in the plasma and a good mixing between the background and the target gases is present. Since the laser absorption by shocked background gas occurs mainly through electron-ion Inverse Bremsstrahlung effect, where the absorption increases as a function of λ^3 [23] thus, it is much less efficient for the UV lasers. As we have mentioned above that the emission spectrum of our plasma is composed mainly from the Ta species and there is almost no emission from the air atoms/molecules and since we are working in the UV range it might be considered that the plasma studied in this work propagates in LSC regime and the plume is composed mainly from the tantalum atoms and ions.

For the small intensity peaks that are not analyzed, we assume that these peaks are due to the trace elements in the target even though XPS analyses of the Ta target did not show other elements rather than tantalum.

Since all the emission lines showed the same intensity profile evolution, only two lines are chosen to have a better 3D figure appearance. The spatial evolution of the emission spectra is shown in Fig. 2. The spectra are measured at delay time of 400 ns corresponding to the end of the continuum radiation. It is clear that the intensity increases rapidly versus distance until it reaches its maximum at 2 mm which corresponds to the highest concentrations of the neutral excited Ta atoms and then it starts to decrease gradually. The reason for this decrease in the intensity is the plasma thermalization by the collision with the atmospheric pressure and by the recombination collision process [24]. The same behavior is present for Ta II too.