# A SPECTROSCOPIC STUDY OF THE ABLATION PLASMA PRODUCED ON $Er^{3+}$-DOPED Ti:$LiNBO_3$

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*Prezentăm un studiu experimental privind dependența temperaturii plasmelor de ablație in funcție de iradianța fasciculului laser. Plasmele de ablație au fost generate prin focalizarea pulsurilor laser cu durata de 4.5 ns şi lungimea de undă de 532 nm incidente normal pe o țintă solidă de $Er^{3+}$:Ti:$LiNbO_3$ aflată în aer. Iradianța laser din focar a fost variată, prin modificarea energiei pulsului, în domeniul 0.2 - 2.5 TW/cm$^2$, în jurul pragului de ionizare al vaporilor din plasmă.Am măsurat intensitățile liniilor Li neutru de-a lungul direcției axiale în plasmă prin scanarea axială a imaginii plasmei cu ajutorul unei fibre de 10 microni care a fost cuplată la intrarea monocromatorului. Rezultatele obținute arată că intensitățile liniilor Li neutru scad cu distanța față de suprafața țintei. Temperatura plasmei calculată cu ajutorul intensităților celor două linii este aproximativ constantă (~16000 K) axial de-a lungul plasmei şi nu depinde de iradianța laser când aceasta este în jurul pragului de ionizare.*

*Here we present an experimental study on the dependence of the temperature within the ablation-plasmas as a function of laser irradiance. The ablation plasmas were generated by focusing 4.5 ns laser pulses with 532 nm wavelength at normal incidence on a solid target of $Er^{3+}$-doped Ti:$LiNbO_3$ in atmospheric air. The laser irradiance in focus was varied, by changing the pulse energy, in the range of 0.2 - 2.5 TW/cm$^2$ which is around the ionization threshold of the vapour plume. The intensity of the neutral Li lines along the axial direction within the plasma plume was measured by scanning axially the plume image with a 10 microns fiber that is coupled to the monochromator entrance. The results indicate that the intensity of the neutral Li lines increases with the distance from the target surface. The plasma temperature calculated by accounting for these lines intensities is almost constant (~16000 K) along the plasma plume axis and do not depend on laser-irradiance when this is around the ionization threshold.*

**Keywords:** ablation plasma, $Er^{3+}$:Ti:$LiNbO_3$.

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## 1. Introduction

Many theoretical and experimental papers using dealt in the last years with the characterization of the $LiNbO_3$ targets due to its attractive piezoelectric, acousto-optical, electro-optical and non-linear optical properties. This makes from the $LiNbO_3$ ferroelectric crystal one of the main materials used in manufacturing optical devices such as wave-guides, optical switches, modulators, nonlinear wavelength converters by second harmonic generation, and surface acoustic wave devices [1-5]. These applications have been implemented mainly on bulk materials but, from technological point of view of integrated optical devices, $LiNbO_3$ thin films are also very promising [1, 8].

Pulsed laser deposition is the main method for complex stoichiometry structures such as $LiNbO_3$ thin films deposition. The basic phenomena involved in laser deposition is laser ablation which imply target heating, melting, evaporation, ionization, and re-solidification/re-crystallisation [1, 3]. The ablation rate depends on the photon energy, fluence, pulse duration and diameter of the laser beam, optical and thermal penetration depths, composition and pressure of the medium, reflectivity of the target surface, etc.

Laser-induced breakdown spectroscopy is a precise and powerful tool for real-time analysis of the laser ablation process because in many cases it is simple, very fast and sensitive [3]. The purpose of this work consist in studying by laser-induced breakdown spectroscopy the characteristics of the ablation plasma generated on $Er^{3+}$-doped Ti:$LiNbO_3$ targets with nanosecond laser pulses.

We determine the spatial distribution of the electrons temperature along the axial direction as a function of laser irradiance. The results can be very valuable for applications like deposition of $LiNbO_3$ thin films by pulsed laser ablation where the control of the plasma temperature at the substrate is very important to obtain uniform films [7].

## 2. Experiment

The experimental set-up used for the spectroscopic characterization of the pulsed laser ablation plasmas of $Er^{3+}$:Ti:$LiNbO_3$ targets is presented in Fig. 1. The second harmonic of a Q-switched Nd:YAG laser (4.5 ns pulse duration, 532 nm wavelength) was used for laser ablation and plasma ignition.

The laser pulses were focused on the target surface at normal incidence with convergent lens ( $f/6$, $f$ = 18 cm). The energy of the beam was varied in the range of 0.5 to 6.5 mJ by using a variable attenuator, giving an irradiance of 0.2 to 2.5 TW/cm$^2$ in the focus. The experiments were carried in open air, at normal conditions.

The maximum length of the plasma plume is about 1 mm. The 4 times magnified image of the plasma obtained with the imaging lens (Fig. 1) was scanned axially with an optical fiber connected to the Acton Research monochromator. Thus, we recorded the lines intensities of different atomic species at different positions within the plume.

Behind the rotating grating of the monochromator the light was detected by a Hamamatsu photomultiplier. At each position of the diffraction grating corresponding to a certain wavelength, the light intensity was integrated during at least a time equal with the inverse of repetition rate of the laser shots. Then the emission spectrum is recorded by a computer.

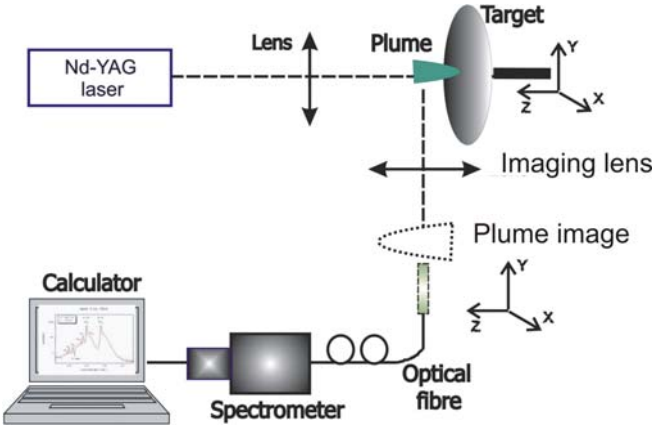



Fig. 1 The scheme of the experimental setup for emission plasma spectroscopy.

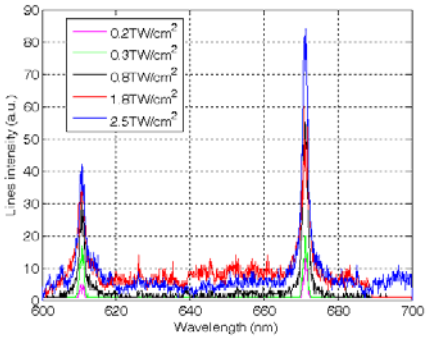
### 3. Results and discussion

Fig 2 presents typical spectra obtained at different positions within the plasma plume generated on $Er^{3+}$-doped Ti:$LiNbO_3$ at different laser irradiances. Each point of the spectra was obtained by averaging the signal coming from maximum 10 consecutive laser pulses incident on the same spot, creating the condition of approximate constant ablation rate during this process. The figure indicate the two main lines of neutral Li atoms at 610 and 671 nm that we used for calculating the plasma temperature.

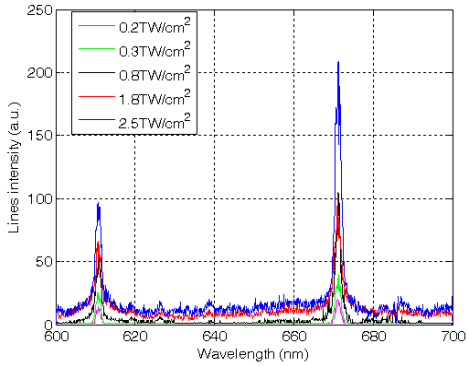
Figure 3 presents the relative intensities of the two Li lines as a function of laser irradiance. Increasing laser irradiance leads to the increase of the lines intensity whereas the ratio of the 671 to 610 nm lines intensities remains almost constant (~1.8).

When moving the observation point into the plume at larger distances (i.e. 1 and 2 mm away) from the target surface, the lines intensities obtained at a given

laser irradiance become larger. These observations could suggest that the plasma is either richer in neutral Li atoms or hotter when increasing the laser irradiance and the distance from the target [9].




a)

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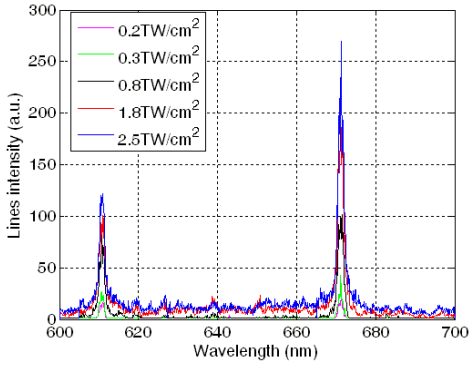


c)

Fig. 2 The visible spectrum of the $Er^{3+}$:Ti:$LiNbO_3$ plasmas at different irradiances (in $TW/cm^2$) and different axial positions within the plasma plume: a) 0 mm, b) 1mm, c) 2 mm away from the target.

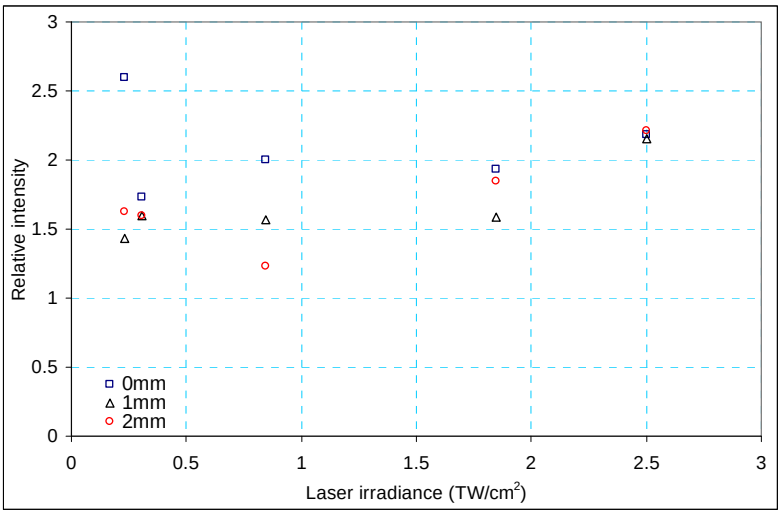


Fig. 3 The ratio of the 671 to 610 nm lines intensities vs laser irradiances at different axial positions within the plume of $Er^{3+}$:Ti:$LiNbO_3$ target as determines from the graphs presented in Fig. 4: near target (0 mm), 1 mm, and 2 mm away from target.

In order to decide what the case we deal with is, the electron temperature was derived from the relative intensities of these atomic lines of Li I, at 610 and 671 nm, presented in Fig. 3. We took into account the Boltzmann's distribution of the atoms on the energetic levels for a thin plasma. The atomic data for these lines is given in Table 1 [6].

*Table 1*

**Atomic data used for the treatment of LiI experimental spectrum.**

| λ(nm) | $E_u\left(\text{cm}^{-1}\right)$ | $J_u$ | $E_l\left(\text{cm}^{-1}\right)$ | $J_l$ | $A_{ul}\left(\text{s}^{-1}\right)$ |
|---|---|---|---|---|---|
| 610.35 | 31283 | 1.5 | 14899 | 0.5 | $6.00\cdot10^7$ |
| 610.37 | 31283 | 1.5 | 14899 | 1.5 | $1.20\cdot10^7$ |
| 610.37 | 31283 | 2.5 | 14899 | 1.5 | $7.10\cdot10^7$ |
| 670.78 | 14904 | 1.5 | 0 | 0.5 | $3.72\cdot10^7$ |
| 670.79 | 14904 | 0.5 | 0 | 0.5 | $3.72\cdot10^7$ |

Due to a very limited spectral resolution of the measurement system, no line profile analysis was possible. So, no flattening of spectral profile in its centre for homogeneous plasma and no absorption dip for a inhomogeneous plasma could be observed. But, due to the very probable self-absorption of the resonance lines radiation at 671 nm in the plasma, the emission coefficient corresponding to

these lines is under-estimated. This fact leads to an estimated value of the electron temperature greater than the real value.

The results presented in Fig. 4 indicate that the plasma temperature as obtained from Li I lines is about 16000 K, being non-dependent on the laser irradiance or the position within the plume. Thereby, the difference in the lines intensities observed in Fig. 3 comes only from the richness in neutral Li atoms that are obtained at high laser irradiances. In fact, increasing laser irradiance leads to larger volumes and densities of ablated material rather than heating the plume [10, 11].

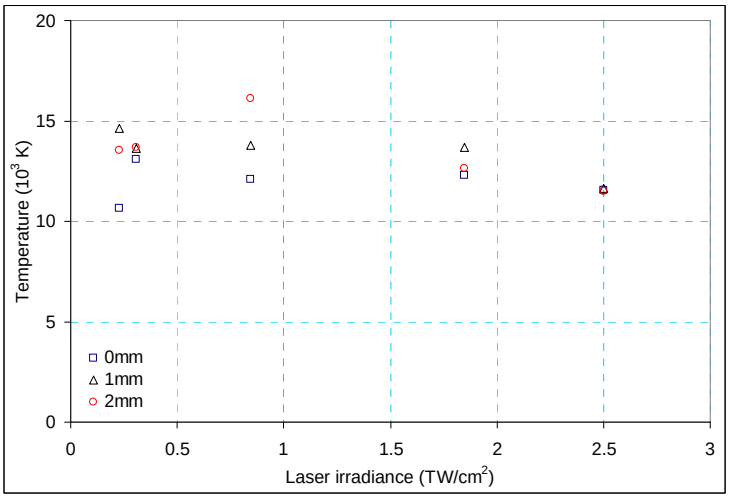


Fig. 4 Plasma temperature as a function of laser irradiance on an $Er^{3+}$:Ti:$LiNbO_3$ target for different positions within the plasma.

## 4. Conclusion

We presented an experimental study on the dependence of the temperature within the ablation-plasmas generated on a solid target of $Er^{3+}$-doped Ti:$LiNbO_3$ as a function of laser irradiance. The ablation plasmas were generated by using 4.5 ns laser pulses with 532 nm wavelength at normal incidence on the solid target in atmospheric air. The laser irradiance in focus was varied, by chancing the pulse energy, in the range of 0.2 - 2.5 TW/cm$^2$ which is around the ionization threshold of the vapour plume.

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neutral Li lines increases with the distance from the target surface. The plasma temperature calculated by accounting for these lines intensities is almost constant (~16000 K) along the plasma plume axis and do not depend on laser-irradiance when this is around the ionization threshold.

The results are very valuable for applications such as deposition of $LiNbO_3$ thin films by pulsed laser ablation for integrated optoelectronic devices where the control of the plasma temperature at the substrate is very important to obtain uniform films.

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