

APPEARANCE OF Be II 436.1 nm LINE WITH FORBIDDEN COMPONENT IN LIBS PLASMA

B. D. STANKOV¹, M. R. GAVRILOVIĆ BOŽOVIĆ² and M. IVKOVIĆ¹

¹*Institute of Physics, University of Belgrade, 11080 Belgrade, P.O. Box 68, Serbia
E-mail biljanas@ipb.ac.rs*

²*Faculty of Engineering, 34000 Kragujevac
E-mail marijana.gavrilovic@kg.ac.rs*

Abstract. In this work study of LIBS on BeO target in low pressure gas mixture of Ar (97 %) and H₂ (3%) using a nanosecond pulsed laser with 266 nm wavelength is presented. The appearance of forbidden component of Be II 436.1 nm was observed and presented in comparison with the previously acquired results for pulsed gas discharge plasma.

1. INTRODUCTION

The results of the experimental study of the singly charged beryllium spectral line 436.1 nm, transition $3p^2 P^0 - 4d^2 D$, and its forbidden component, transition $3p^2 P - 4f^2 F^0$, were previously reported, e.g.: see Stankov et al. 2018.a In this paper beryllium lines were recorded from gas discharge running in pulsed regime, after ablation of beryllium oxide (BeO) discharge tube. The plasma source was described in detail, e.g.: see Stankov et al. 2018.b.

The reason for the necessity of devising another experiment, to examine Be II with forbidden line, is twofold: a) there were dust particles in the pulsed discharge plasma observed, e.g.: see Stankov et al. 2018.b.; b) published experimental results can not be used for testing of the overall line shape modeling because the influence of the allowed line optical thickness and additional electric field has not been accounted for, e.g.: see Stankov et al. 2018.a.

LIBS stands for Laser Induced Breakdown Spectroscopy technique that uses a short laser pulse to create plasma on the sample surface. Analysis of this plasma enables determination of material's elemental composition, without sample preparation and with simple experimental setup. Difficulties that arise during analysis are mainly associated with the fast evolving plasma which demands well temporally and spatially resolved measurements.

This paper explores the possibility of recording Be II line with forbidden component in LIBS plasma. As a comparative study, LIBS method was chosen because it is assumed there will be no dust particles production. Besides, experimental setup used for LIBS plasma creation gives the possibility for spatially resolved measurements and experimental testing of self-absorption, which could not be performed in gas discharge experiment. Aim of this study is to check the possibility of spatially and temporally resolved measurements of Be line with forbidden component in dust free environment.

2. EXPERIMENT

Experimental apparatus for pulsed gas discharge plasma recordings, e.g.: see Stankov et al. 2018.b, was set up as for the standard end-on linear discharge plasma observation. Axial image of the plasma source was projected onto the entrance slit of a spectrometer (Andor Technology, Shamrock 303), by the use of a focusing mirror and achromatic lens. For line shape recordings the imaging spectrometer was equipped with iCCD camera (Andor Technology, model DH734). Line shape recordings were performed with full vertical binning and gate width of 50 ns at various delay times.

For the LIBS method, the fourth harmonic of Nd:YAG Q-switched laser (Quantel, Q-smart 450) at 266 nm, with repetition rate of 10 Hz, was used. The pulse energy at 266 nm was 70 mJ, on the average. The laser beam was focused perpendicular to the BeO target by the means of biconvex achromatic lens L1 (f = 100 mm), see Figure 1.

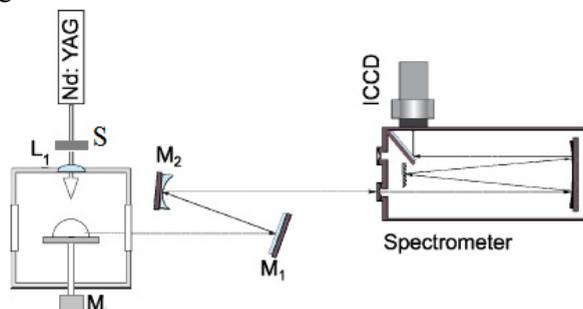


Figure 1: LIBS setup

Target made from BeO ceramics was glued to the carrier, which was rotated by the motor (M), in the low pressure chamber. 1 : 1 image of the plasma plume was projected by optical mirrors M1 and M2 on the entrance slit (20 μ m wide) of a 0.5 m Ebert-type spectrometer ($f/8.6$ equipped with a grating of 1180 grooves per mm). The spectrometer was mounted with an iCCD camera (Andor Technology, model DH734I-18U-03, with 1024 x 1024 pixels, 13 x 13 μ m size, 18 mm intensifier diameter). The iCCD camera was operated in the image mode and controlled using a pulse generator (DDG 535, Stanford Research Systems) triggered optically by the occurrence of plasma on the BeO target. Fast photodiode placed towards the target was used to convert the light signal. The shatter (S), placed between the laser and the lens L1, was changing open/close position for every 16 laser shots. Images of the plasma were accumulated over 16 laser shots and the final signal was the product of 10 such accumulations. Accumulation of the signal was performed to exclude the influence of the eventual shot-to-shot changes in the signal. The acquisition gate width was 100 ns.

3. RESULTS AND DISCUSSION

Diagnostics of plasma parameters, the electron number density, N_e , and electron temperature, T_e , was used for characterization of two experimental methods.

For optimal conditions in the pulsed discharge plasma electron number density, N_e , was determined from the peak separation $\Delta\lambda_{ps}$ of the H_β line using formula (6) from e.g.: see Ivković et al. 2015. T_e was estimated from the ratio of Be II 467.3 nm/Be I 457.3 line intensities, using formula (1):

$$\frac{I_1}{I_2} = \frac{h^3}{2(2\pi mk)^{3/2}} \frac{(gA)_1 \lambda_1 N_e}{(gA)_2 \lambda_2 T_e^{3/2}} \exp\left(\frac{E_2 - E_1 + E_1^{ion} - \Delta E}{kT_e}\right), \quad (1)$$

where E_1^{ion} is ionization potential and ΔE is ionization potential lowering.

After several iterations N_e and T_e were determined, and presented in Table 1, e.g.: see Stankov et al. 2018.a. Maximum values of N_e and T_e are measured to be $9.3 \cdot 10^{22} \text{ m}^{-3}$ and 16800 K.

For LIBS method, the chamber was filled with 10 mbar of Ar (97%) and H_2 (3%) mixture. The previously mentioned experimental conditions were chosen in order to achieve maximum Be and H line intensity.

Experimental profile of the $H\alpha$ line fitted with Voigt function was used for determination of N_e in LIBS plasma. Stark halfwidth, w_S , is determined by assuming $w_S = w_L$ and introducing this result in Eq. (12), e.g.: see Konjević et al. 2012. Instrumental line profile was measured at several wavelengths using Oriol penlight calibration lamp. T_e was estimated from the ratio of Be II 467.3 nm/Be I 457.3 line intensities, equation (1). The electron density and plasma temperature are displayed in Figure 2a as functions of time. Temporal evolution of Be II nm with forbidden component is presented in Figure 2b. Forbidden component, transition $3p^2 P - 4f^2 F^0$, is easily seen on the blue wing of Be II, transition, $3p^2 P^0 - 4d^2 D$.

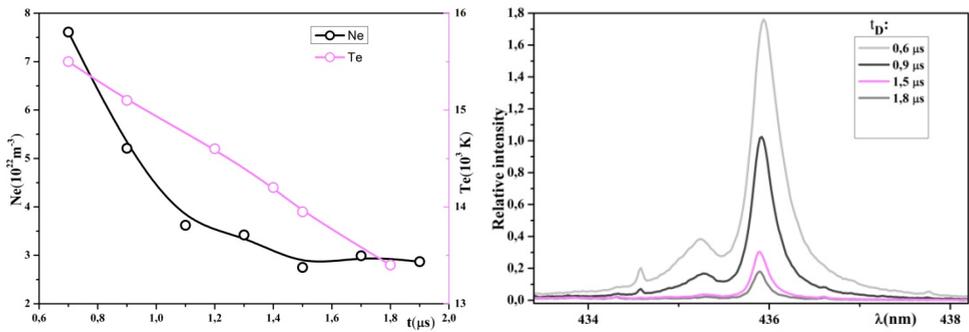


Figure 2: a-Temporal evolution of Ne and Te for LIBS on BeO target, b-Temporal evolution of Be II with forbidden component for LIBS on BeO target

The dependence of wavelength peaks separation s and peaks intensity ratio F/A upon N_e for Be II allowed and forbidden component is shown in Figure 3.

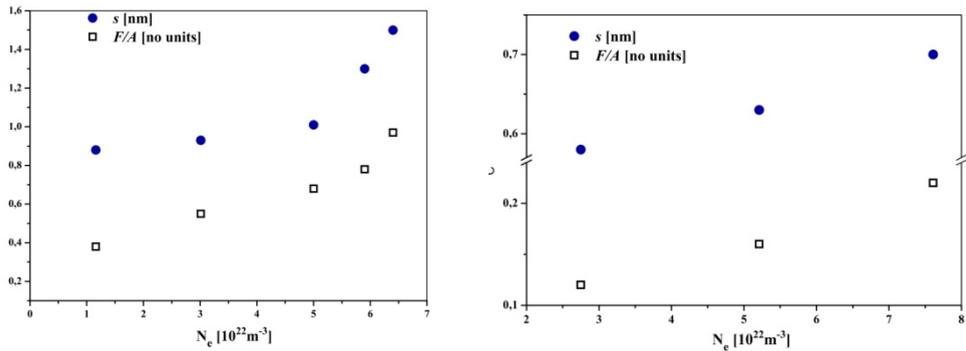


Figure 3: wavelength peaks separation s and peaks intensity ratio F/A dependence upon N_e for Be II Line with forbidden component for: a) pulsed discharge plasma b) LIBS plasma

Slight discrepancy between $s[\text{nm}]$ values for two experimental setups exists. More notable discrepancy is observed for F/A values. Reason for this may be presence of self-absorption and spatial inhomogeneity in both experimental setups. The example of spatially resolved Be II 436.1 nm line is presented in Figure 4.

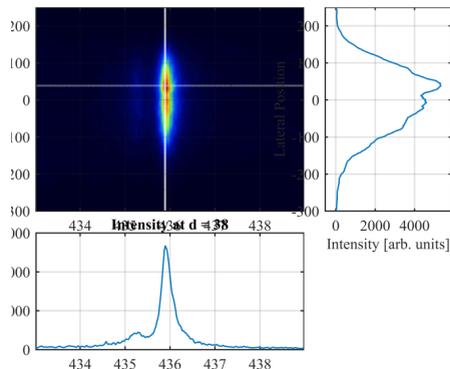


Figure 4: Spatially resolved Be II 436.1 nm line in LIBS plasma

In the next step of investigation, N_e and T_e will be determined using spatially resolved measurements of Be and H lines. Since there were no dust particle observed, as assumed, results obtained from LIBS experiment may be used to evaluate the effect of dust particles on Be line parameters obtained from gas discharge.

References

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