H_{α} AND H_{β} PROFILES IN TOTAL EMISSION SPECTRA OF LASER-INDUCED PLASMAS USED FOR ANALYTICAL APPLICATIONS

K.Yu. Catsalap¹, E.A. Ershov-Pavlov¹, L.K. Stanchits², K.L. Stepanov²

¹B.I. Stepanov Institute of Physics of the NAS of Belarus, Nezalezhnastsi ave. 68, 220072 Minsk, Belarus, catsalap@imaph.bas-net.by

²A.V. Lykov Institute of Heat and Mass Transfer of the NAS of Belarus, Brouka st. 15, 220072 Minsk, Belarus, kls@hmti.ac.by

Introduction. The H_{α} and H_{β} hydrogen Balmer lines are commonly used for plasma diagnostics in many applications including chemical elemental analysis by optical emission spectroscopy with the sample excitation by laser radiation. For example, the determination of electron number density N_e from the profile of hydrogen Balmer line H_{β} (486.133 nm) is a well-established and widely used plasma diagnostic technique. In the range 10^{16} - 10^{17} m⁻³ the electron density can be determined from H_{β} line profile with 4-7% accuracy. Obviously, local measurements are necessary, which are very complicated, because of the laser plasma high inhomogeneity and instability. For such plasmas, total emission spectra (integrated along a line of sight during exposition time) can be used to compare them with the spectra pre-calculated for a due set of distributions of the plasma parameters using a numerical model.

In this paper, the numerical modeling of H_{α} and H_{β} profiles in the laser plasma emission spectra has been performed for space/time behavior of the plasma expressed in a parametrical form. The plasma properties correspond to the hydrogen-containing laser erosion plasma in excitation conditions typical for analytical applications. Modeled profiles of the lines in the plasma total emission spectra have been fitted to the measured ones by means of changes of the plasma parameters in the model entrance data. The fitting allows performing the plasma local diagnostics, particularly obtaining additional information on the plasma self-absorption in the lines considered and on the following changes in the line profiles and intensity.

Experiment. The laser beam of Q-switched Nd YAG laser (pulse energy and duration 50 mJ and 12 ns, respectively) is focused on a solid sample surface covered with water. Power density is 5 GW/cm² approximately. The conditions of an excitation are close to ones which are typical ones for the laser assisted nanoparticle forming. The observation axis is parallel to sample surface (copper). Two different spectral ranges are used - 400-550 nm (H_{β} and neutral copper lines) and 550-700 nm (H_{α} line). The grating PC-controlled spectrometer S150 has a spectral resolution of 0.12 nm. Laser repetition frequency is 10 Hz.

The spectra are averaged on 20 laser pulses. The sketch of the experimental setup is shown on Fig. 1



Fig. 1. Experimental setup. 1 – solid sample covered with water, 2 – laserinduced plasma, 3 – laser beam, 4 – condenser, 5 – light fiber, 6 – spectrometer, 7 - PC

Modeling. The laser plasma is assumed as having symmetrical monotonous temperature profile with one maximum along the observation direction. This temperature profile approximates ones of the laser plasma with good accuracy and can be easily parameterized. Halfwidths of local and integrated H_{α} and H_{β} line profiles were obtained by computer simulation and numerical solution of radiation transfer equation respectively. Plasma composition is calculated in assumption that the mixture of hydrogen and oxygen corresponding to thermally decomposed water is under atmospheric pressure and states in local thermodynamic equilibrium. The ratio calculated allows introducing an inhomogeneity correction to the procedure of electron concentration determining by the lines halfwidth. The calculation was performed for wide range of maximal plasma temperature and inhomogeneity parameter values. The broadening data for the hydrogen lines were taken from /1, 2/. The radiation transition model is described elsewhere /3, 4/.

Results and discussion. Inhomogeneity and emission self-absorption distort the measurement data. Generally, an optically "thick" line is broader than an optically "thin" line. Also, emission spectral lines of inhomogeneous plasma are narrower than ones of homogeneous plasma. In a simple case of homogeneous plasma and Lorentz profile one could get an analytical expression which connects line halfwidth and plasma optical thickness /5/.

However, in a case of inhomogeneous laser – induced plasma and complicated line profile of hydrogen line the analytical expression could not be obtained and the dependence can be calculated using a numerical model only. The numerical model we developed allows accounting for the distortions and to measure the values more correctly. Fig. 2. shows an experimental profile of H_{β}

line and its fitting by calculated one. While the recorded profile is slightly asymmetrical one, the spectra coincidence is satisfactory.



Fig. 2 – Profile of H_{β} line– experiment (dots) and fit (solid)

Using the numerical model the dependence of hydrogen lines reduced halfwidths on plasma inhomogeneity and optical thickness can be obtained. Actually, optical thickness τ affects a relative broadening of a line. Hence it is logical to consider a dependence of halfwidth $\Delta\lambda$ reduced to its optically "thin" value $\Delta\lambda_0$. The dependence for H_β is shown on Fig. 3.



Fig. 3. – Dependence of reduced halfwidth of H_{β} on plasma optical thickness for different inhomogeneity rates. Dots – theoretical approximation

The dependence is corresponding to parabolic temperature profile $T(y)=T_0[1+(y/y_0)^{\alpha}]^{-1}$, where T_0 – maximal temperature on a line of sight, $2y_0$ – effective plasma size, parameter α describes an inhomogeneity rate ($\alpha \rightarrow \infty$ corresponding to homogeneous plasma).

The profile is common for the laser plasma stages when neutral lines have a maximum of emissivity. From the Fig. 3 one can see that the inhomogeneity distorts line halfwidth greatly, while a calculation corresponds to homogeneous plasma tends to analytical approximation /5/. To perform a measurement one must determine a plasma optical thickness and evaluate a plasma inhmogeneity. The program we developed could fit a recorded profile by altering plasma optical thickness and inhomogeneity rate, allowing measurement of both values. Combining of data from both lines could greatly improve the fitting due to significant difference of optical thickness of H_{α} μ H_{β} lines.

Conclusion. At present paper the dependence of halfwidth of $H_{\alpha} \mu H_{\beta}$ lines in emission spectra of laser-induced plasma in water on optical thickness of the plasma. The correction factors accounting for plasma inhomogeneity are calculated. The factors allow getting a halfwidth of the hydrogen lines as if it would be in emission of optically "thin" plasma for diagnostics purposes. Investigation results could be used for a spectroscopic control and diagnostics of laser induced plasma in nanoparticle synthesis processes.

References

- 1. Ershov-Pavlov E.A., Catsalap K.Yu., Stepanov K.L., et al. J. of Appl. Spectros. 69 (2002) 395–401.
- 2. Ershov-Pavlov E.A., Katsalap K.Yu., Stepanov K.L., Stankevich Yu.A. Spectrochim. Acta. Part B 63 (2008) 1024–1037
- 3. **Griem H.R.** Spectral line broadening by plasma, Academic Press, New York, 1974.
- 4. **Kasabov G. A., Yeliseev V.V.** Spectroscopic tables for low-temperature plasma /[in Rus.] Касабов Г.А., Елисеев В.В. Спектроскопические таблицы для низкотемпературной плазмы. Москва, Атомиздат 1973
- 5. Amamou H., Bois A., Ferhat B. et al. JQSRT 75 (2002) 747–752