A LETTERS JOURNAL EXPLORING
THE FRONTIERS OF PHYSICS

LETTER

Forbidden component of the Be II 436.1 nm line recorded from pulsed gas discharge plasma

A LETTERS JOURNAL EXPLORING
THE FRONTIERS OF PHYSICS

September 2018

EPL, **123** (2018) 63001
doi: 10.1209/0295-5075/123/63001

www.epljournal.org

Forbidden component of the Be II 436.1 nm line recorded from pulsed gas discharge plasma

B. D. STANKOV[1,2], M. IVKOVIĆ[1(a)], M. VINIĆ[1] and N. KONJEVIĆ[3]

[1] *Institute of Physics, University of Belgrade - P.O.Box 68, 11080 Belgrade, Serbia*
[2] *University of Novi Sad, Faculty of Sciences, Department of Physics - Trg Dositeja Obradovića 4, 21000 Novi Sad, Serbia*
[3] *Faculty of Physics, University of Belgrade - P.O.Box 368, 11000 Belgrade, Serbia*

received 27 June 2018; accepted in final form 13 September 2018
published online 10 October 2018

PACS 32.70.Jz – Line shapes, widths, and shifts
PACS 52.70.Kz – Optical (ultraviolet, visible, infrared) measurements

Abstract – We report results of the experimental study of the singly charged beryllium spectral line 436.1 nm, transition $3p^2P^o$-$4d^2D$, and forbidden component, transition $3p^2P$-$4f^2F^o$, which is for the first time identified in this work. Beryllium lines are recorded from gas discharge, after ablation of a beryllium oxide discharge tube, running in pulsed regime with the following gases: helium with traces of hydrogen, argon with traces of hydrogen and pure krypton. The ratio of line intensities and wavelength separation of the Be II 436.1 nm line and neighbouring line located at the blue wing are followed in the electron density range $(1.16–6.4) \times 10^{22}\,\mathrm{m}^{-3}$ determined from the hydrogen Balmer beta line (H_β) in the electron temperature interval between 10500 K and 15500 K. The functional dependence of the wavelength separation range and peak intensity ratio of these lines upon electron number density suggests the complex profile of the forbidden and allowed line, which can be used for diagnostics of low-temperature beryllium containing plasmas.

Introduction. – The subject of this experimental study is an investigation of the shape of the Be II 436.1 nm line which appeared with a strong component, located at the blue wing, in our gas discharge [1]. The assumption was that this may be the forbidden component of the allowed line 436.1 nm, and this work is an attempt to prove this hypothesis. If proven, this result will partially fill the gap between the investigations of this type of transitions along a lithium isoelectronic sequence, with already published data for Li I [2–4], C IV [5–7] and N V [7].

The spectral lines with forbidden components attracted attention some time ago, see, *e.g.*, [8], because of numerous applications in the field of Stark broadening theory testing, for electron number density, N_e, laboratory plasma diagnostics and in astrophysics for the analysis and modelling of the star atmosphere, see, *e.g.*, [9]. In addition, in the case of beryllium lines there are two more specific applications. One is related to the study of the inner plasma-wall interaction in ITER (International Thermonuclear Experimental Reactor) since this wall is considered to be covered with beryllium [10]. The other important field of application is in astrophysics, considering that beryllium is a naturally occurring element in metal-poor stars [11].

Here, it should be emphasized that under a forbidden component (transitions with $\Delta l \neq \pm 1$, where l is the angular momentum quantum number) we consider lines which occur as a result of the breakdown of the parity selection rules induced by ambient plasma electric microfield. This effect has nothing to do with the forbiddeness associated to magnetic dipole, electric quadrupole or higher multipole transitions. A forbidden line starts to appear close to the allowed one when wave functions become mixed. It happens when plasma broadening of the allowed line becomes comparable with the energy levels separation between the allowed transition and the nearest dipole allowed perturbing level or levels. With an increase of the electric field (*i.e.*, an increase of the charged particles density), the mixing of the wave functions becomes stronger, and the wavelength difference between the allowed and forbidden components peaks becomes more pronounced. A further increment of the electric field brings an overall profile of the line shape close to the one of a hydrogen-like emitter with linear Stark effect. The conclusion here is that the overall shape of these lines is sensitive to the charged particle

(a) E-mail: milivoje.ivkovic@ipb.ac.rs (corresponding author)

density. Therefore, it can be used for the N_e plasma diagnostics. It should be stressed that plasma conditions when a forbidden component becomes significant differ from element to element and even from line to line. The overall shape of lines with forbidden components to a smaller extent depends also on the electron temperature, reduced mass and ratio between electron and gas temperature [12], which opens possibilities for other plasma parameters diagnostics. The most frequently studied spectral lines of this type belong to the visible spectrum of He I, see, *e.g.*, [8,13,14]. The application of parameters of these lines for electron density diagnostics is demonstrated in several publications and references therein [12,15–21]. In addition, parameters of these lines are applied also for DC electric-field measurements, see, *e.g.*, [22,23].

To achieve the aim of this work, we recorded the overall shape of the Be II 436.1 nm line, together with nearby lines, in a discharge tube with different carrier gases. Simultaneously, N_e and the electron temperature, T_e and the dependence of the line profile parameters upon N_e were determined.

Experiment. – In this section the experimental setup, the measurement procedure as well as the method and results of plasma diagnostics will be described. The experimental apparatus was set up as for the standard end-on linear discharge plasma observation, see, *e.g.*, [1]. For data acquisition two spectra recording systems were used. In both cases the 1:1 axial image of the plasma source was projected onto the entrance slit of a monochromator, by the use of a focusing mirror and achromatic lens, see fig. 6 in [24]. Almost all spectral line shape recordings were performed using an imaging spectrometer (Shamrock 303 Andor), having instrumental half-width of 0.09 nm equipped with ICCD camera DH734 (Andor). These line shape recordings were performed with full vertical binning and gate width of 50 ns at various delay times. Delay times from the beginning of the current pulse, monitored by the Rogowski coil were determined with a digital delay generator (Stanford Research Systems, DG535). In order to check the influence of the instrumental broadening to the line shape, the cross-check with a second high-spectral-resolution recording system was performed. The spectra were obtained using a 1 m monochromator (McPherson Model 2051), having the instrumental half-width of 0.02 nm. This monochromator was supplied with a stepping motor and photomultiplier radiation detector, PMT (EMI 9658 R). The signal from the PMT was observed by the digital oscilloscope Tektronix TDS360 (bandwidth 200 MHz), triggered by the signal supplied from the Rogowski coil. The step motor rotates the diffraction grating of the monochromator so that, at different wavelengths, the radiation intensity can be recorded. The computer simultaneously collects data from the oscilloscope and controls the step motor. The end signal represents the mean value of 4 consecutive signals. This shot-to-shot technique was justified considering that the pulse-to-pulse current reproducibility was better than 2%. More details about data acquisition and data manipulation can be found in [24].

Plasmas are created in a BeO ceramic discharge tube with inner diameter of 2.6 mm, outer diameter of 10 mm and length of 130 mm. Beryllium spectral lines are detected in the discharge after evaporation of Be off the inner tube wall by ablation induced by the discharge itself. Here, it should be emphasized that due to the toxicity of beryllium, a special procedure in handling the discharge tube and ablation products was always adapted [1]. The most important result of the previous study [1] is that the optimum conditions for excitation of Be spectral lines in our discharge are achieved when the capacitor $C = 5\,\mu\text{F}$ is charged up to 7 kV in argon with 3% of hydrogen at $p = 1.2$ mbar. At lower discharge voltages only lines of the carrier gas appear in spectra [1]. In addition, only at low gas pressure, $p < 5$ mbar, the percentage of the beryllium atoms in the discharge becomes significant and Be becomes the main plasma constituent and most of the Be spectral lines appear in the recorded spectra.

In order to characterize the plasma source, diagnostics of N_e and T_e were performed. The electron number density, N_e, was determined from the peak separation $\Delta\lambda_{ps}$ of the H_β line using formula (6) from [25]:

$$\log N_e[\mathrm{m}^{-3}] = A + B \log \Delta\lambda_{ps}[\mathrm{nm}] = 22.65 + 1.53 \log \Delta\lambda_{ps}[\mathrm{nm}]. \tag{1}$$

The values of parameters A and B are taken from [25] for a presupposed electron temperature of 13000 ± 3000 K. For electron number densities determined in such a way, the electron temperature, T_e was estimated from the ratio of Be II 467.3 nm/Be I 457.3 line intensities using the formula

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

After several iterations N_e and T_e were determined. The required transition probabilities were taken from the NIST database [26].

Since the Be lines have noticeable intensity in a very short time interval, the plasma diagnostics was performed only for times between 8 μs and 18 μs from the beginning of the current pulse. It should be stressed that N_e was determined only with a gas mixture of Ar with 3% H_2, since the intensity of the H_β line under other experimental conditions was negligible. Namely, in the stated delay times interval, lines of other elements have very low intensity with the exception of O II lines (originating from the ablation products of BeO ceramics) and H_β. The recorded O II lines have a width equal to the instrumental width and they are located on the broad wings of the studied Be II line shape, see fig. 2, and, therefore, they are not convenient for N_e diagnostics. The intensity of H_β line

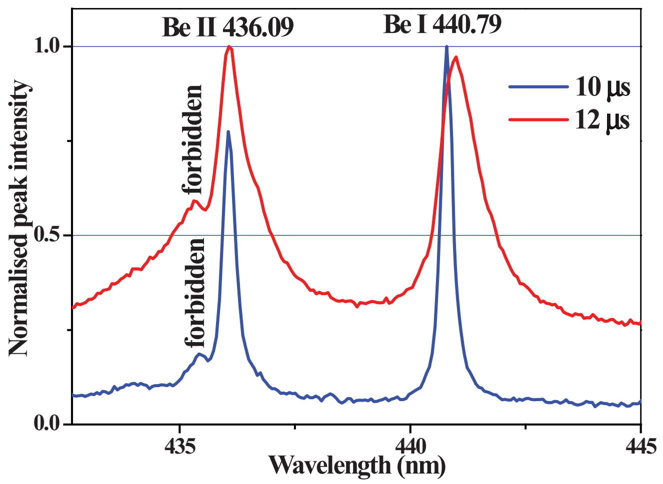

Fig. 1: (Colour online) Beryllium spectrum between 429 nm and 445 nm with normalised peak line intensities recorded in 1.2 mbar of He + 3% H_2 gas mixture at discharge voltage of 7 kV at different delays after beginning of the discharge current pulse.

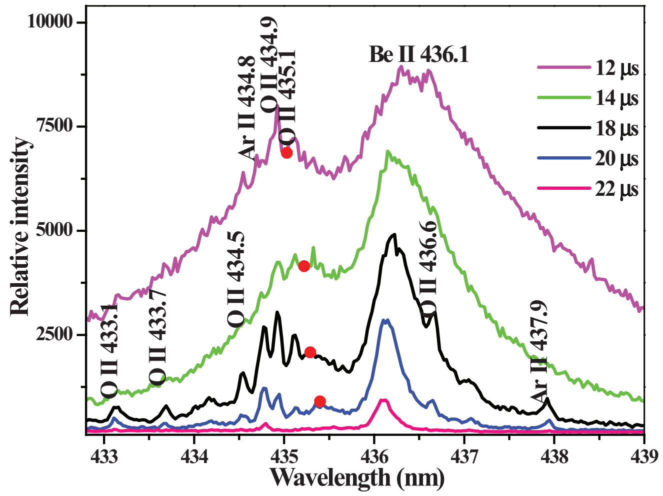

Fig. 2: (Colour online) Temporal evolution of the Be spectrum between 433 nm and 439 nm, recorded at discharge voltage of 7 kV at 1.2 mbar of Ar with 3% of H_2. The wavelength positions of the peaks are taken from [25]. Peak intensity and wavelength position of the forbidden component are denoted with circles and presented in table 1.

at the time of interest in helium with 3% of hydrogen gas mixture was very weak and not usable for diagnostics as well. Small contribution of hydrogen traces in krypton plasma prevented detection of H_β line, also. The use of strong Be lines for N_e diagnostic has a significant drawback since their Stark widths may introduce inaccuracy due to pronounced self-absorption and even self-reversal in the case of resonant lines 313.0 nm and 313.1 nm.

Results and discussion. – The main result of this experimental study is the identification of the forbidden component $3p^2P$-$4f^2F^o$ allong with allowed component $3p^2P^o$-$4d^2D$ of the Be II line at 436.1 nm in various gases. Although it has been previously stated [1] that the best conditions for excitation of beryllium lines were achieved when the carrier gas was argon, we first present the results in He with 3% H_2 plasma, see fig. 1. The reason for this being that in spectra recorded when the carrier gas was He with 3% H_2, only beryllium lines appeared, *e.g.*, there are no other lines that could entail confusion in the determination of a forbidden line shape and peak wavelength position.

However, in such discharge, beryllium lines appeared in a shorter period of time (10–n12 μs). In order to confirm that the recorded line belongs to singly ionized beryllium and to analyse the possible influence of the Ar II and O II lines on the wavelength position and intensity of the forbidden component, spectra recordings were performed for Ar and Kr using the same discharge conditions that were employed for He with 3% H_2.

In fig. 2 recordings made from Ar plasma are presented. It can be seen that on the blue side of Be II 436.1 nm the appearance of other spectral lines can call into question the presence of the forbidden Be II line in the spectra presented in fig. 2. Lines which may interfere with the Be II 436.1 nm overall line profile belong to hydrogen Balmer gamma (H_γ) 434.0 nm, Ar II at 434.81 nm and O II at 434.91 nm and 435.1 nm.

Having in mind the Stark shift and intensity of the H_γ, this line influence on the Be II 436.1 nm profile, at our experimental conditions, is small. Namely, for the estimated T_e of 10500 K–15500 K the H_γ line has at least twice lower peak intensity than the H_β line, recorded at the same experimental conditions, see fig. 5 in [1], and even smaller intensity at the position of the forbidden component due to the $\Delta\lambda^{-5/2}$ line wing dependence.

In order to resolve the influence of other non-hydrogenic plasma constituents, spectra recordings presented in fig. 2 were performed with smaller instrumental broadening, *i.e.*, the apparatus with better resolution, see section "Experiment". From the results in fig. 2 one can conclude that the Ar II and O II lines may interfere in the determination of the peak wavelength position of the forbidden component. Fortunately, the difference in line widths (the forbidden line is much broader) confirms the existence of the forbidden component.

In the case of Kr plasma, fig. 3, the forbidden line is clearly visible, but its peak intensity and wavelength position are different in comparison with Ar plasma, under the same excitation conditions. Also, the appearance of the forbidden component in the Kr plasma is prolonged.

Differences between spectra recorded in various gases are illustrated by the change of ratio between the intensity of the allowed Be II line 436.1 nm and Be I line 440.79 nm, see figs. 1 and 3, thus indicating that the plasma parameters are incomparable.

In order to use the Be II line 436.1 nm with forbidden component $3p^2P$-$4f^2F^o$ for plasma diagnostics, the functional dependence of the wavelength separation between

Table 1: Temporal variation of N_e, T_e, s and F/A in argon with 3% of hydrogen for $p = 1.2$ mbar and $U = 7$ kV.

t (μs)	N_e (10^{22} m^{-3})	T_e (K)	s (nm)	F/A
10	6.40	15500	1.5	0.97
12	5.90	14200	1.3	0.78
14	5.00	13500	1.01	0.68
16	3.01	11980	0.93	0.55
18	1.16	10500	0.88	0.38
20			0.74	0.26

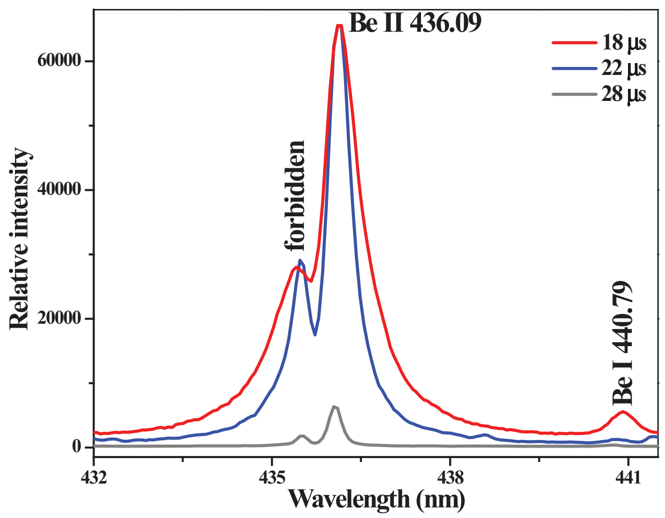

Fig. 3: (Colour online) Temporal evolution of the Be spectrum between 429 and 445 nm, recorded at discharge voltage of 7 kV at 1.2 mbar of Kr.

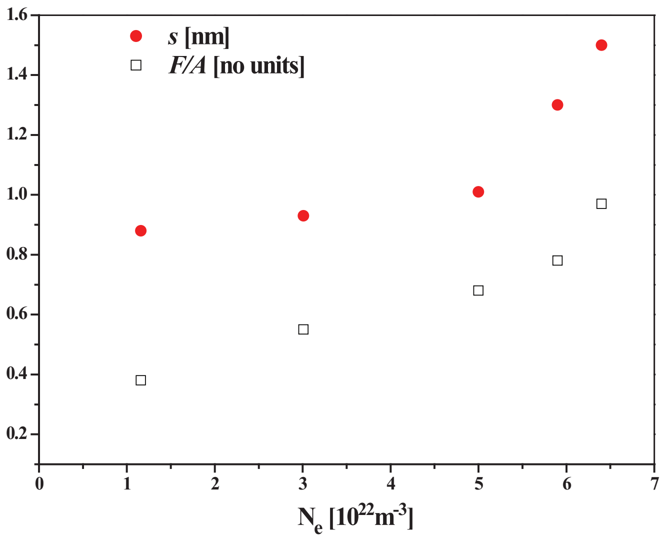

Fig. 4: (Colour online) The dependence of wavelength peaks separation s and peaks intensity ratio F/A dependence upon N_e for Be II allowed $3p^2P^o$-$4d^2D$ component and forbidden $3p^2P$-$4f^2F^o$ component at $p = 1.2$ mbar of Ar + 3% H_2 at $U = 7$ kV.

their peaks s, and/or the ratio between the maximum intensities of the forbidden component and allowed component, F/A, upon N_e should be determined. Unfortunately for our experimental conditions the Be II lines appear in a short time interval in which diagnostics of N_e is very difficult. Nevertheless, in spite of all the difficulties to determines the $s(N_e)$ and $F/A(N_e)$ dependences, we made an attempt to demonstrate the potential of this line with the forbidden component for the plasma diagnostics application, see fig. 4 and table 1.

The range of s and F/A and the corresponding N_e determined in the case of Ar with 3% H_2 suggests the possibility of using the Be II 436.1 nm line with forbidden component for plasma diagnostics purposes. Namely, it is shown that under our experimental conditions ($p = 1.2$ mbar, $U = 7$ kV and $C = 5\,\mu$F) during plasma generation and decay s changes between 0.74 nm and 1.5 nm, while F/A changes between 0.26 and 0.97 in the range of electron densities $(1.16$–$6.4) \times 10^{22}$ m^{-3} and electron temperatures 10500 K–15500 K. The aforesaid functional dependence of wavelength separation and intensity ratios of two lines upon electron density are typical for lines with forbidden components. Unfortunately, a small N_e range does not allow the determination of the best-fit formulas $N_e = f(s)$ or $N_e = f(F/A)$ for reliable plasma diagnostics.

Conclusions. – On the bases of our experimental study we conclude that the spectral line located at the blue wing of the Be II 436.1 nm line, transition $3p^2P^o$-$4d^2D$, has all the characteristics of the forbidden transition. In order to prove that this line originates from the forbidden $3p^2P$-$4f^2F^o$ transition we carried out several studies like wavelength analysis of plasma impurities, measurement of the wavelength separation and ratios of two line intensities *vs.* electron number density. All results are indicating that the forbidden line $3p^2P$-$4f^2F^o$ is present in spectra of our discharge, which is well illustrated in figs. 1–3. The functional dependence of the wavelength separation and intensity ratios of two lines upon electron density are typical for lines with forbidden components, see, *e.g.*, He I lines [12,18–21]. Thus, on the basis of all the presented results one may conclude that the line at the blue wing of Be II 436.1 nm line, is a forbidden line belonging to the $3p^2P$-$4f^2F^o$ transition. Finally the wavelength separations and the ratios of peak line intensities in table 1 cannot be used for the testing of the overall line shape modelling of this beryllium allowed line with forbidden component since the influence of the allowed line optical thickness and additional electric field has not been examined in this work. For the same reason the data in table 1 may be applied for low-temperature plasma diagnostics with great precautions. One interesting application can be for the N_e determination during *in situ* examination of plasma facing materials in tokamak (containing beryllium) by laser-induced breakdown spectroscopy (LIBS) [27], in which electron density and temperature ranges are close to the values studied in this work.

On the basis of the analysis of energy levels along a lithium isoelectronic sequence for Li I [2–4], C IV [5–7], NV [7] and our results for Be II, we propose the study of the B III line 195.38 nm from the same transition $3p^2P^o$-$4d^2D$ in order to check whether the forbidden components, $3p^2P$-$4f^2F^o$ and $3p^2P$-$4f^2P^o$ will appear. In the present study of the Be II lines the forbidden component $3p^2P$-$4f^2P^o$ was not detected.

* * *

This work has been financed by the Ministry of Education and Technological Development of the Republic of Serbia under the Project 171014. The authors thank the technician Stanko Milanović for technical assistance in the preparation and setting-up of the experiment.

REFERENCES

[1] Stankov B. D., Vinić M., Gavrilović Božović M. R. and Ivković M., *Rev. Sci.*, **89** (2018) 053108.

[2] Jovićević S., Gigosos M. A., Ivković M., Gonzalez M. A. and Konjević N., *Contributed Papers of the 22nd Summer School and International Symposium on the Physics of Ionized Gases (XXII SPIG, Kopaonik)* (Institute of Physics, Belgrade, Serbia) 2006, p. 315, ISBN 86-82441-18-7.

[3] Cvejić M., Gavrilović M., Jovićević S., Ivković M. and Konjević N., *EMSLIBS 2011 Euro Mediterranean Symposium on Laser Induced Breakdown Spectroscopy, Izmir, Turkey, 2011, Book of Abstracts* (İzmir Institute of Technology, Turkey) 2011, p. 128, `http://www.worldcat.org/title/euro-mediterranean-symposium-on-laser-induced-breakdown-spectroscopy-emslibs-2011-izmir-turkey-11-15-september-2011/oclc/75675399`.

[4] Cvejić M., Stambulchik E., Gavrilović M. R., Jovićević S. and Konjević N., *Spectrochim. Acta B*, **100** (2014) 86.

[5] Werner K., Rauch T., Hoyer D. and Quinet P., *Astrophys. J.*, **827** (2016) L4.

[6] Werner K., Hoyer D., Rauch T., Kruk J. W. and Quinet P., in *20th European White Dwarf Workshop*, edited by Tremblay P.-E., Gänsicke B. and Marsh T., *ASP Conference Series*, Vol. **509** (Astronomical Society of the Pacific) 2017.

[7] Bottcher F., Musielok J. and Kunze H.-J., *Phys. Rev. A*, **36** (1987) 5.

[8] Griem H. R., *Spectral Line Broadening by Plasmas* (Academic Press, New York) 1974.

[9] Beauchamp A., Wesemael F. and Bergeron P., *Astrophys. J. Suppl. Ser.*, **108** (1997) 559.

[10] How J. and Reichle R., International Thermonuclear Fusion Experimental Reactor Project 2009 ITER Report, ITER D 2X6K67 v1.0 Plant Description (PD) Cadarache.

[11] Gilmore G., Gustafsson B., Edvardsson B. and Niessen P. E., *Nature*, **357** (1992) 379.

[12] Gigosos M. A. and Gonzalez M. A., *Astron. Astrophys.*, **503** (2009) 293.

[13] Bekefi G., Deutch C. and Yaakobi B., *Principles of Laser Plasmas* (John Wiley & Sons, New York) 1976.

[14] Chernichowski A. and Chapelle J., *J. Quant. Spectrosc. Radiat. Transfer*, **33** (1985) 427.

[15] Uzelac N. I. and Konjević N., *Phys. Rev. A*, **33** (1986) 1349.

[16] Uzelac N. I., Stefanović I. and Konjević N., *J. Quant. Spectrosc. Radiat. Transfer*, **46** (1991) 447.

[17] Perez C., De La Rosa I., Aparicio J. A., Mar S. and Gigosos M. A., *Jpn. J. Appl. Phys.*, **35** (1996) 4073.

[18] Ivković M., Jovićević S. and Konjević N., *Spectrochim. Acta B*, **59** (2004) 591.

[19] Ivković M., Gonzalez M. A., Jovićević S., Gigosos M. A. and Konjević N., *Spectrochim. Acta B*, **65** (2010) 234.

[20] Gonzalez M. A., Ivković M., Gigosos M. A., Jovićević S., Lara N. and Konjević N., *J. Phys. D*, **44** (2011) 194010.

[21] Ivković M., Gonzalez M. A., Lara N., Gigosos M. A. and Konjević N., *J. Quant. Spectrosc. Radiat. Transfer*, **127** (2013) 82.

[22] Kuraica M. M. and Konjević N., *Appl. Phys. Lett.*, **70** (1997) 1521.

[23] Kuraica M. M., Konjević N. and Videnović I. R., *Spectrochim. Acta B*, **52** (1997) 45.

[24] Konjević N., Ivković M. and Sakan N., *Spectrochim. Acta B*, **76** (2012) 16.

[25] Ivković M., Konjević N. and Pavlović Z., *J. Quant. Spectrosc. Radiat. Transfer*, **154** (2015) 1.

[26] `https://physics.nist.gov/PhysRefData/ASD/lines_form.html`.

[27] Morel V., PÉRÈS B., Bultel A., Hideur A. and Grisolia C., *Phys. Scr.*, **T167** (2016) 014016.