SPECTROSCOPIC INVESTIGATIONS OF CORONA DISCHARGE IN HIGH PRESSURE HELIUM AT 300K

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Abstract. Gaseous Helium at 300K and pressure (0.1-3)MPa was excited using a corona discharge both for negative and positive high voltages. The light emitted from the ionization zone of the discharge was analyzed. Asymmetric shape of atomic lines 706 nm and 728 nm was recorded. Blue wings of the lines are more intensive than their red wings. The line shape is described as a convolution of Lorentz profile of the line center and a quasi-statistical profile of a blue wing of the line. Such analysis predicts no heating of the gas in the ionization zone for positive corona and considerable heating for negative corona.

1. Introduction

Emission spectroscopy is a powerful tool to obtain information about the important parameters that characterize non-equilibrium discharge plasma at both low and high pressures. Spectroscopic observations of the light emitted by ionization gases can be used to determine conditions surrounding the emitted atoms or molecules [1, 2]. Corona discharge is characterized by strong spatial inhomogeneity. The corona current is determined by mobility of electrons or positive ions in a low-field drift zone but the current density increases strongly with approaching to the region with high electric field near a tip electrode. This region (ionization zone) is a source of a light emitted by the corona. Excited atoms interact with environment and features of their spectra give information about density and temperature of a gas in the ionization zone.

The pressure broadening of spectral lines depends on the gas density. The “impact” interaction of radiator with surrounding atoms determines the Lorentzian profile of spectral lines emitted by a discharge in low pressure gases [3-6]. In this case the width $\Delta\lambda$ (Full Width Half Maximum) of a line and the line shift S relative its non-perturbed location is proportional to gas density. Collisional interactions between excited and ground state atoms lead to broadening and shift of emitted spectral lines. When radiating and perturbing atoms are the same species, there are two types of interaction that must be considered. If the emitted spectral line involves a transition between two states neither of which are optically connected to the ground state, then the interaction leads to both broaden and shift the spectral line. Such are all lines due to transitions between triplet levels of the ortohelium. The analysis of broadening and shift of such lines has been made in [7] for the Lennard-Jones potential of the dispersion interaction between excited atom and surrounded atoms in the ground state. The shift sign ("red shift" for the shift toward longer wavelengths and "blue shift" for the shift toward shorter wavelengths) depends on character of radiator-perturbator interaction. The blue shift corresponds to significant repulsion. Measurements [8-10] shows the blue shift of the line 706 nm (3^3S-2^3P transition) of the helium spectrum. The measurements were made using discharge in low pressure gas (< 10 Torr) and gave the symmetric Lorentzian profile of the line with shift and width are proportional to the gas density. If the experiment is carried out in a gas under fixed pressure and experimental conditions permits a heating of the gas by electric current, the density of the gas (and parameters of spectral line) is inverse proportional to the gas temperature. Moreover, the impact interaction depends on a relative velocity of colliding atoms. It leads to direct dependence of the parameters of gas temperature.

The other type of interaction arises when lower state involved in the transition is connected to the ground state by a resonance transition. The "resonant" interaction broadens but does not shift the spectral lines. The example of such lines is the line 728 nm (3^1S-2^1P transition) of the helium spectrum. The pressure width of the line has no direct dependence of temperature [11-13].

Growth of the density of a gas is accompanied by distortion of the "impact" Lorentz profile of lines. It has been shown [14] that the van der Waals attraction between a radiator and perturbators results in increasing of the intensity of a "red wing" of a line as compared with its "blue wing". The asymmetry of a spectral line shape was described using "quasi-static" approach where perturbators are assumed stationary [14]. The quasi-static profile of a line with van der Waals perturbation has non-zero intensity for wavelengths which are larger than the wavelength of non-perturbed line. It is due to attractive interaction between radiator and surround atoms.

The corona discharge in helium under pressures up to 5MPa allows us to observe lines of He I in these conditions. It was originally supposed to calculate temperature and density of a gas in ionization zone of corona, using the impact approximation for treatment of the width and the shift of spectral lines. But measurements have detected considerable asymmetry of observable lines. Their "blue" wings have more intensity than "red" wings. It has demanded to include both impact approach and quasi-static one in the analysis of the shape of spectral lines. The impact profile of a line depends on both the gas pressure and the temperature. The quasi-static profile depends on the density only. Ideal gas relation between the density and the temperature at fixed pressure is appended to analysis of the line shape based on impact approximation. The impact approximation describes the central part of the line while the quasi-static approximation describes its wing. The different density and temperature dependences of these mechanisms allow us to treat the experimental data more definitely.

The objective of the analysis is to predict the gas temperature in the ionization zone using the experimental data on spectral line emitted from the zone. It will be shown that the gas is heated in the negative corona but does not in the positive corona.

2. Experiment and results

Experiments have been carried out in gaseous He at the fixed temperature 300K of the gas and different pressures in the cell from 0.1MPa up to 3MPa. The corona discharge (ionization of gaseous He) occurred in a vicinity of a tip electrode under high voltage. The discharge domain (ionization zone) had a volume less than an inter-electrode space (drift zone). The corona current has been measured for different pressures in a space-charge-limited regime. This regime is characterized by an electric current as a quadratic function of the applied voltage V. Electrons with high mobility are the charge carriers in the drift region of the negative corona and it results in large negative corona current. Positive ions with low mobility are the charge carriers in the positive corona. Therefore the current of the positive corona is lower than that of the negative one.

The gaseous sample was produced from helium at the grade N 60 (Air Liquide) with an impurity concentration of about 0.1 ppm of oxygen. The gas was further purified by a series of traps that were filled with a mixture of molecular sieves (3-10 Å) and charcoal, activated under vacuum typically at 350°C for 3 days. The corona discharge cell included a point electrode and had a characteristic impedance of 50 Ohm and it could withstand pressures up to 10 MPa. Before filling the cell was pumped to about 10^{-4} Pa using a turbo-molecular pump. Tungsten tips with a radius of 2.5μm were prepared by electrolytic etching. The electrode spacing was 8 mm. All metallic electrodes were supported by Macor insulators. The high voltage from a stabilized dc power supply (Spellman RHSR/20PN60) was connected to the point electrode. In the cell the temperature of the gas could be adjusted to 300 K at a fixed pressure P for each series of measurements.

Light emitted from the region close to the point electrode was analyzed by a spectrograph through a sapphire window. The spectrograph (Acton Research Corporation of 300 mm focal length) was equipped with 3 plane gratings: one with 150 gr./mm and two with 1200 gr./mm that were blazed at 750 nm and 300 nm, respectively. The 2D-CCDTKB-UV/AR detector is located directly in the exit plane of the spectrograph. Its dimensions are 12.3x12.3 mm with 512x512 pixels of 24x24 μm for each pixel. In order to reduce the dark current, the detector was cooled to a temperature of 153 K (dark current <1 e/pixel/hour at 153 K). In our conditions, the instrumental broadening measured by recording profiles of argon lines from a low pressure discharge lamp is $\Delta\lambda_{instr}$=0.1 nm for a 1200 grooves/mm grating.

The light emitted from the corona region was collected and spectra in the range 500 - 1080 nm were recorded. Most of atomic lines and molecular bands were identified. These lines correspond to radiative transitions between excited states of He* atoms and He_2* excimer molecules. At low pressure the lines are sharp and their peak position match the atomic lines and molecular bands of helium from gas phase experiments, Table 1.

A strong background continuum from 490 to 1100 nm appears in spectra at high pressures above 4.0 MPa. Moreover, the width of the lines increases with pressure and their relative intensity decreases. The noise superimposes on the line shape at high pressures. Though, the qualitative characteristic of the line shape can be recorded for pressures up to 2.5 MPa. Blue shift of line maximum and more appreciable broadening of lines are observed. Atomic lines manifests asymmetric of their profile which increases with pressure. The skewness of the line shape is exhibited as larger intensity of its blue wing compare with a red wing. This effect is due to interaction of a radiator with surrounding atoms. The treatment of the phenomena is presented below and some properties of the corona ionization zone are predicted based on the treatment. We concentrate our attention on the analysis of shape of the atomic lines 706 nm and 728 nm. Figure 1 shows the Grotrian diagram of the atomic levels involved in these transitions. The line 706 nm interacts with surrounded atoms by dispersion forces and it is subjected of the pressure shift and broadening. The line 728 nm has lower level 2^1P which is connected with the ground state by the resonance line 58.4 nm and it is subjected of the resonant broadening without shift.

Our experiments have exhibited more complicated picture of the spectra of lines emitted by corona discharge in dense helium. Figures 2 and 3 show the 706 nm line (the triplet transition) being broadened and shifted with increasing pressure towards smaller wavelengths (blue shift). The shape of the line observed in high pressures gas is different strongly from symmetric Lorentzian shape of the spectral line of a low pressure gas discharge. The blue shift of the line indicates a significant repulsion between excited atoms and atoms in the ground state. The line profile is strongly asymmetric and its blue wing is more intensive than a red wing. Moreover, the spectrum measured in negative corona and one in positive corona are different. The asymmetry of the line is larger in the spectra of the positive corona. Both spectra exhibit the blue shift and more intensive blue wing. The figures present very interesting phenomenon of “satellites” observed at both 696nm and 715nm. Their intensity increases with pressure and the effect is more appreciable in the positive corona. In this paper we analyze the shape of the central line and give an explanation in the framework of the classic theory of pressure broadening of spectral lines.

The line 728 nm (the transition between singlet levels 3^1S-2^1P) was observed in the negative corona for different pressures. The spectra are presented in Figure 4. Both broadening of the line and blue shift have been recorded. The asymmetry of the line shape exhibits large intensity of its blue wing.

3. Theoretical treatment

The theory of "impact" broadening of spectral lines predicts symmetric Lorentzian profile of the line with shift and width being proportional to a gas density N [3 - 6]. The analysis of the line distortion due to model Lennard-Jones potential of the interaction between radiator and perturbators [7] showed that the shift of the line depends on the nature of the interaction. If the van der Waals attraction prevails over the short range repulsion, the shift is toward long-length side (the "red" shift). If the repulsion is prevailed, the "blue" shift is predicted. The blue shift has been observed in our experiments and this allows us to assume the repulsive interaction ($U(r) = \hbar C_{12} r^{-12}$) between a radiator and perturbators. Within the "impact" approximation for the repulsive interaction, the expressions for the line broadening $\Delta\lambda_L$ and the line shift S_L are given by:

$$\Delta\lambda_L = 6.44\left(\frac{\lambda^2}{2\pi c}\right) w^{9/11} C_{12}^{\ 2/11} N\,,$$

$$S_L = 0.922\left(\frac{\lambda^2}{2\pi c}\right) w^{9/11} C_{12}^{\ 2/11} N\,. \tag{1}$$

Here $\lambda = 706$ nm is the wavelength of the line, $w = 10^4 * T^{0.5}$ is the relative velocity in [cm/s] of the He atoms with T being the gas temperature in [K], C_{12} is the repulsive Lennard-Jones parameter in [cm^{12} s^{-1}] and N is the gas number density in [cm^{-3}]. The impact approximation predicts the symmetric Lorentz profile of the line with a ratio of the shift and the width of 0.143, which is indeed close to what we found in our experiments with He gas at 300K and low pressures. The theoretical calculations with more realistic description of the interaction than the L-J potential [15], gave for the 706 nm line broadening and shift at the temperature T

$$\Delta\lambda_L = 2.9\cdot 10^{-21}\cdot N\cdot (T/300)^{9/22}\,,$$

$$S_L = 0.384\cdot 10^{-21}\cdot N\cdot (T/300)^{9/22} \tag{2}$$

Here shift and FWHM are in nm, T is in K, and N is the gas number density in [cm^{-3}]. Using the theoretical value for the broadening (2) and its treatment with the repulsive potential (1) one can estimate the repulsion constant value C_{12}(706 nm) = $1.6*10^{-72}$ cm^{12}/s.

The resonance broadening of the line 728 nm is described in terms of the oscillation strength f = 0.276 of the resonance transition of the line 58.4 nm. The resonance broadening produces a temperature-independent width [5]

$$\Delta\lambda_{res} = 1.44\frac{\lambda^2 r_0 f \lambda_r}{8}\cdot N \tag{3}$$

Here $f = 0.276$, $\lambda_r = 58.4$ nm, $\lambda = 728$ nm, $r_0 = e^2/4\pi\varepsilon_0 mc^2 = 2.8\cdot 10^{-13}$ cm is the "classical radius of electron", and N is the density of atoms in the gas. The calculation gives for the resonance width of the lines 728 nm

$$\Delta\lambda_{res}(728nm) = 4.3\cdot 10^{-21}\cdot N, \tag{4}$$

It has been shown in [14] that the asymmetric line can be obtained in the quasi-static approximation in the frame of the pressure broadening theory. The approximation assumes zero velocity of atoms and it becomes significant for large density of a gas. The profile of a line $I(\Delta\omega)$ as a function of detuning $\Delta\omega$ from the line centre is described by the formula [16]

$$I(\Delta\omega) \propto \int_{-\infty}^{\infty} \exp[i\Delta\omega\rho - 4\pi N V(\rho)]d\rho \tag{5}$$

Here a red wing corresponds to $\Delta\omega<0$ and the blue wing is for $\Delta\omega>0$. The interaction between the radiator and perturbators $\Delta U(R)$ as a function of their spacing R is described by the term $V(\rho)$

$$V(\rho) = \int_0^{\infty}\left[1 - \exp\left(-i\rho\frac{\Delta U(R)}{\hbar}\right)\right]R^2 dR \tag{6}$$

The property $V(-\rho)=V^*(\rho)$ allows us to rewrite Eq.(5) in the form [14]

$$I(\Delta\omega) \propto \int_0^{\infty}\begin{pmatrix} \exp[-i\Delta\omega\rho - 4\pi N V^*(\rho)] + \\ + \exp[i\Delta\omega\rho - 4\pi N V(\rho)] \end{pmatrix} d\rho \tag{7}$$

Below we shall use the simple form of the repulsive potential $\Delta U(R) = C_{12}/R^{12}$ with one parameter C_{12}. It allows us to describe the line profile using dimensionless variables

$$I_{wing}(x) = \int_0^{\infty} \exp(-2.41y)\cdot\cos(x\cdot y^4 - y)y^3 dy \tag{8}$$

Here $x = \Delta\omega/(15.02\cdot N^4 C_{12})$. The integral in Eq.(8) differs from zero for $x>0$ (a blue wing of the line) due to positive sign of the repulse interaction. The quasi-static profile Eq.(8) has the blue shift and describes the high frequency wing of the line. The center of the line is described by the Lorentzian profile with the impact broadening

$$I_L(\lambda) = \left(\frac{\Delta\lambda_L}{2}\right)^2\left((\lambda - \lambda_0 + S_L)^2 + \left(\frac{\Delta\lambda_L}{2}\right)^2\right)^{-1} \tag{9}$$

with the shift S_L and the width $\Delta\lambda_L$. In order to combine the central profile Eq.(9) and the wing-profile Eq.(8), the convolution operation was used in a manner as it been done in [14, 17 - 19]

$$I(\lambda) = \int_0^{\lambda_0} I_{wing}(x) I_L(\lambda - x)dx \tag{10}$$

Here the integration variable x is the difference between the wing wavelength and the wavelength of the line center λ_0, $x = (\lambda_0 - \lambda) > 0$. The analytical expression has been used for the total shape of the line, Eq.(10)

$$I(\lambda) = \int_0^\infty \frac{z^{0.9}\exp(-0.1 \cdot z)}{\left((\Delta\lambda_w + \lambda \cdot z)^2 + \left(\frac{\Delta\lambda_L}{2}\right)^2 \cdot z^2\right)} dz \qquad (11)$$

Here λ is a wavelength from the shifted center of the line, $\lambda_0 - S_L$. The total profile depends on the impact broadening of line center (FWHM is $\Delta\lambda_L$, Eqs.(2) and (3)) and on the quasi-static parameter $\Delta\lambda_w$

$$\Delta\lambda_w = \frac{15.02\lambda_0{}^2 N^4 C_{12}}{2\pi c} = 2.48 \cdot 10^{-2} \cdot P^4 \qquad (12)$$

Here $\Delta\lambda_w$ is in nm and the pressure P is in MPa. These parameters have different density dependence. The parameter of impact width of the line center is proportional to gas density, while $\Delta\lambda_w$ has strong density dependence as N^4.

The theory developed was used to describe the shape of the lines observed in our experiments. The total profile of a line was calculated as a convolution both the Lorentzian impact profile dependent of temperature and density and the density dependent quasi-static profile described the blue wing of the line. The pressure P of a gas was fixed in our experiments. The density N and the temperature T of a gas in an emitting zone are connected by the ideal gas state relation $P = NT$. To simulate the profile of the line 706 nm the parameters of the impact profile $\Delta\lambda_L$ and S_L have been calculated using Eq.(2). For the line 728 nm the parameter $\Delta\lambda_{res}$ was calculated using Eq.(4). In the final analysis, the temperature was a fitting parameter which determines the total profile of a line for given pressure. The results of the simulation of the line 706 nm are presented in Figures 5 and 6 for different pressures for positive and negative coronas. The simulation shows that the more broadened lines observed in the positive corona represent the spectrum of an atom included in a dense gas under given high pressure and at the temperature of 300 K. Electric current of the positive corona is determined by drift of positive ions with low mobility. It seems that such low current is unable to heat the gas inside the emitting zone. The accordance between experiment and simulation shows that there is no heating of the gas in the positive corona, Fig. 5. The less broadened lines observed in a negative corona can be described tiking into account the gas heating. The best fitting between experimental and calculated profiles has been obtained on assumption that the temperature in the ionization zone is 500 K that is higher than in a bulk gas. The density of the gas in the zone is less than that in non-heating surrounding substance.

The electric current of the negative corona is determined by the drift of high mobility electrones. The magnitude of the negative coroa current is much more than the current of the positive corona. Therefore, the heating of the emitting gas in negative corona is more possible.

The quasi-static non-resonant interaction of a radiator with surrounding atoms leads to a shift and distortion of the singlet line 728 nm emitted from high pressure gas. The interaction was assumed as repulsion potential $U(r) = \hbar C_{12} r^{-12}$ with $C_{12} = 10^{-72}$ cm^{12}/s. We took the same potential as that for the case of 706 nm line because the repulsion is due to an exchange interaction between an excited electron of a radator and closed shell of an atom in the ground state. The total profile of the line was calculated as a convolution, Eq.(11), of the Lorentzian resonance profile with width calculated using Eq.(4) and the quasi-static profile, Eq.(8), which is produced by the non-resonant repulsion. The gas density was calculated from ideal gas state equation for the given pressure and the temperature was the fitting parameter of the simulation. The result of the simulation is presented in Figure 7. The most adequate agreement between the experimental line profiles and their simulation has been obtained for the temperatures 500 ÷ 560 K. This result correlates with that obtained by analysis of the line 706 nm spectrum. The temperature of the gas in the ionization zone of a negative corona is higher than the temperature of a bulk gas.

The calculations pedict more large width of the line 728 nm than that calculated using Eq.(4) for the resonance broadening and the blue shift, wich is non-linear function of the pressure. The data of the measurements are shown in Figures 8 and 9 as the width and shift observed in different pressures. We have assumed that the temperature of the gas in the emitting zone is 500 K and the same for different pressures. The result of the calculations of the width and shift of the line 728 nm in such conditions is shown in Figures 8 and 9 as lines. Note that the parameters of the line shape are non-linear functions of the pressure (or gas density for fixed temperature). The non-resonant repulsion gives strong dependence of the parameters on the gas density. The shift of the distorted line increases with density faster than a linear function.

4. Conclusion

The line observed in the negative corona has less broadening than predicted by its simulation for the gas density calculated using the pressure values and the temperature 300K in the ionization zone. The heating of a gas in ionization zone was assumed. The heating leads to decreasing of gas density under fixed pressure. The impact broadening of the line centre decreases proportional to the density and increases with the temperature. The more significant effect is attenuation of the asymmetry of the line shape due to weakening of the quasi-static parameter. The most adequate agreement between the experimental line profiles and their simulation occurs if the temperatures 500 K ÷ 560 K is assumed for the ionization zone of the negative corona. Indeed, the negative corona currant in the space-charge-limited conditions is larger than that in the positive corona, because higher electron mobility in the drift zone of the negative corona. The larger current results in possibility of considerable heating of the gas near a tip electrode.

5. Acknowledgements

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Table 1. Transitions observed in Helium corona discharge (300K, 0.1MPa).

Atomic lines	
λ(nm)	Upper-Lower
492,19	4d ^{1}D-2p ^{1}P
587,56	3d ^{3}D-2p ^{3}P
706,52*	3s ^{3}S-2p ^{3}P
728,13*	3s ^{1}S-2p ^{1}P
1083,02	2p ^{3}P-2s ^{3}S
Molecular bands	
λ (nm)	Upper-Lower
464,95	$e^3\Pi_g$- $a^3\Sigma_u^+$
573,49	$f^3\Delta_u$(v=0)-$b^3\Pi_g$(v=0)
575	$f^3\Delta_u$ (v=1)- $b^3\Pi_g$ (v=1)
577	$f^3\Delta_u$ (v=2)- $b^3\Pi_g$ (v=2)
639,6	$d^3\Sigma_u^+$- $b^3\Pi_g$
659,55	$D^1\Sigma_u^+$- $B^1\Pi_g$
913,61	$C^1\Sigma_g^+$- $A^1\Sigma_u^+$
918,3	$c^3\Sigma_g^+$- $a^3\Sigma_u^+$

* The lines are analyzed in the paper.

Captions

Figure 1. Grotrian diagram of the atomic levels involved in the transitions of the lines 706 nm and 728 nm.

Figure 2. Experimental spectra obtained for 706nm line in negative corona under different pressures.

Figure 3. Experimental spectra obtained for 706nm line in positive corona under different pressures.

Figure 4. Experimental spectra obtained for 728 nm line in negative corona under different pressures.

Figure. 5. Simulation of the 706nm line emitted by positive corona at 300K and 1.5 MPa and 2.3MPa – solid and dashed lines; dotted lines – Lorentzian profiles.

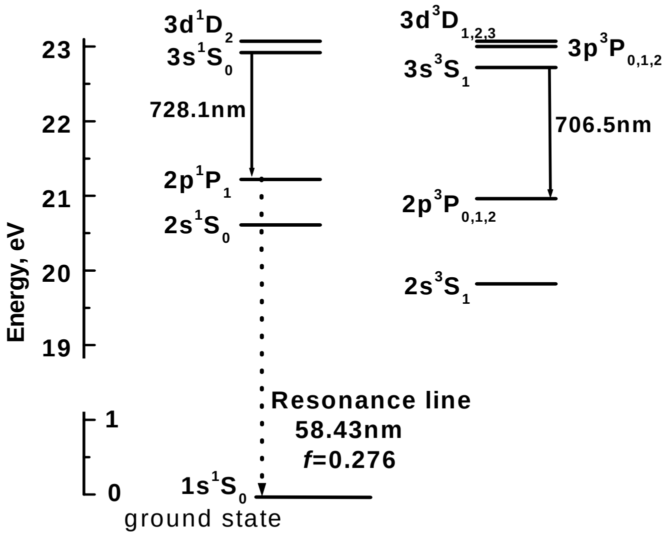
Figure 6. Simulation of the 706nm line emitted by negative corona at 500K and 1.5 MPa and 2.3MPa – solid and dashed lines

Figure 7. Line 728 nm emitted from negative corona in helium inder pressures 20 and 27 MPa. Points – experimental data; Solid lines – simulation; Deshed lines – Lorentz profile with resonance broadening in the same conditions

Figure 8. Width FWHM of 728 nm line in negative corona in helium under different pressures. Points – experiment; Line – result of simulation.

Figure 9. Blue shift of 728 nm line in negative corona in helium under different pressures. Points – experiment; Line – result of simulation.

Figures



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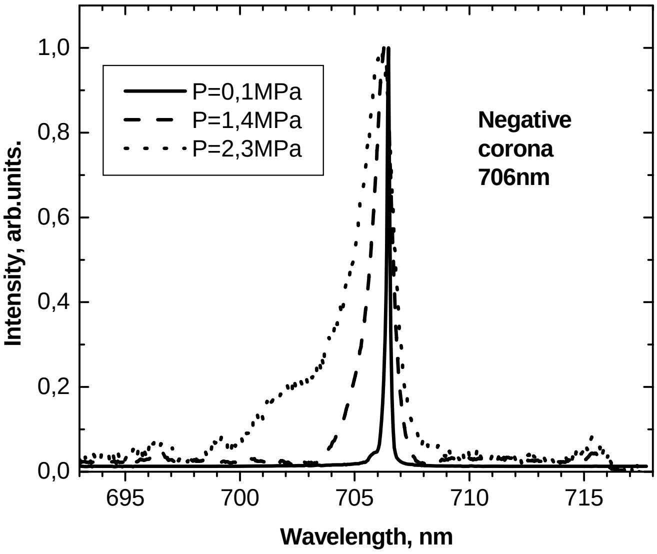


Figure 2. Experimental spectra obtained for 706nm line in negative corona under different pressures.

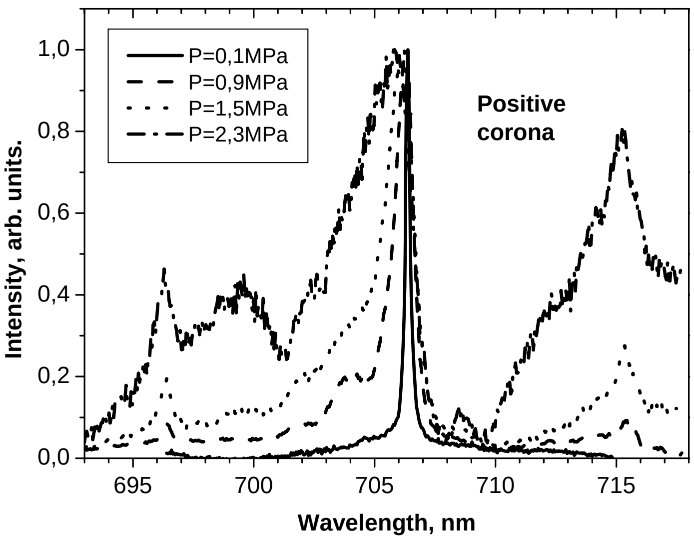


Figure 3. Experimental spectra obtained for 706nm line in positive corona under different pressures.

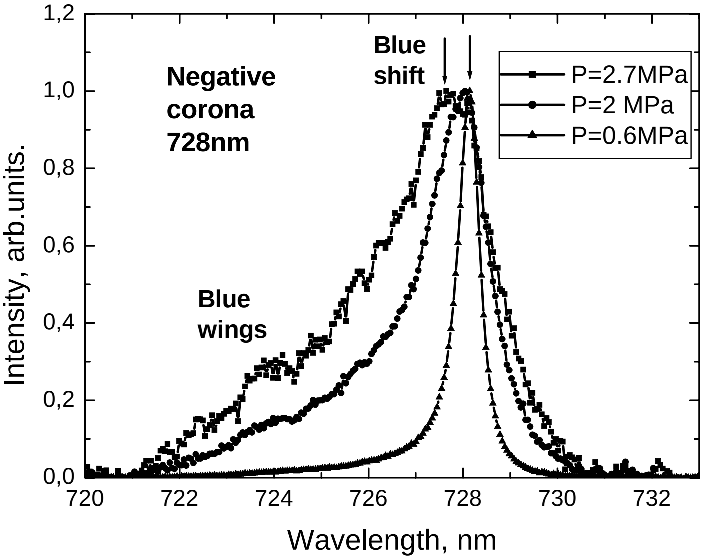


Figure 4. Experimental spectra obtained for 728 nm line in negative corona under different pressures.

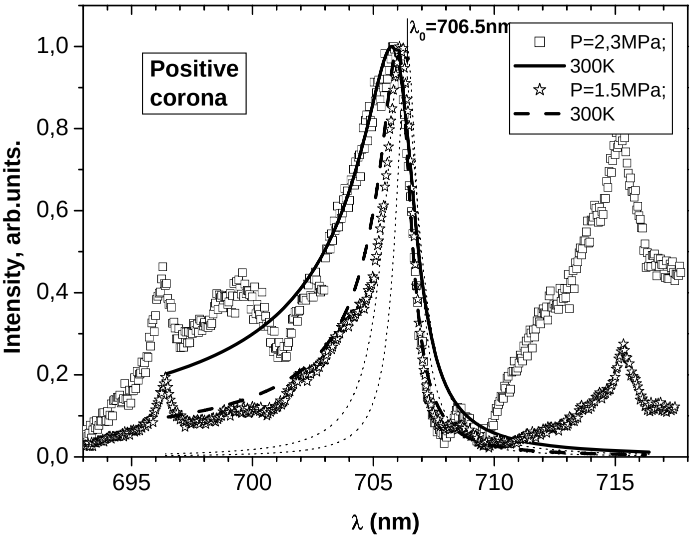


Figure. 5. Simulation of the 706nm line emitted by positive corona at 300K and 1.5 MPa and 2.3MPa – solid and dashed lines; dotted lines – Lorentzian profiles.

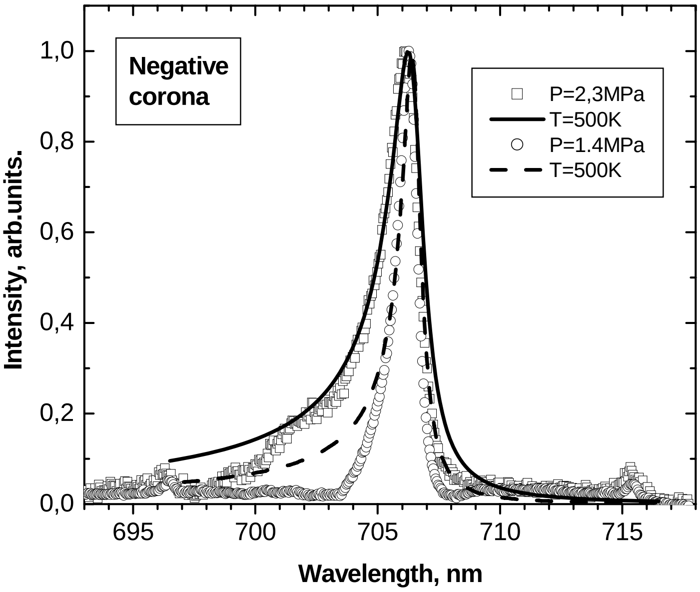


Figure 6. Simulation of the 706nm line emitted by negative corona at 500K and 1.5 MPa and 2.3MPa – solid and dashed lines

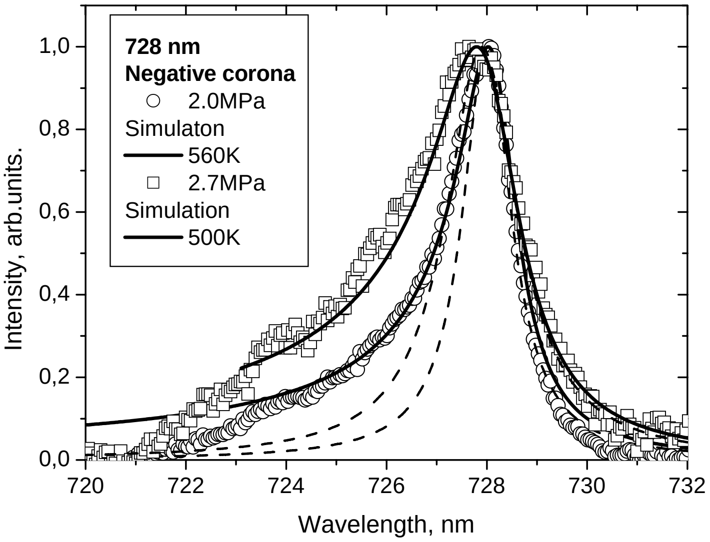
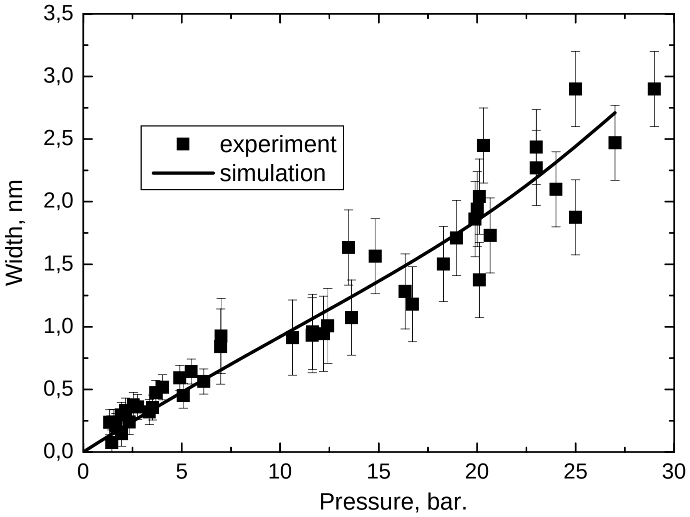


Figure 7. Line 728 nm emitted from negative corona in helium inder pressures 20 and 27 MPa. Points – experimental data; Solid lines – simulation; Deshed lines – Lorentz profile with resonance broadening in the same conditions



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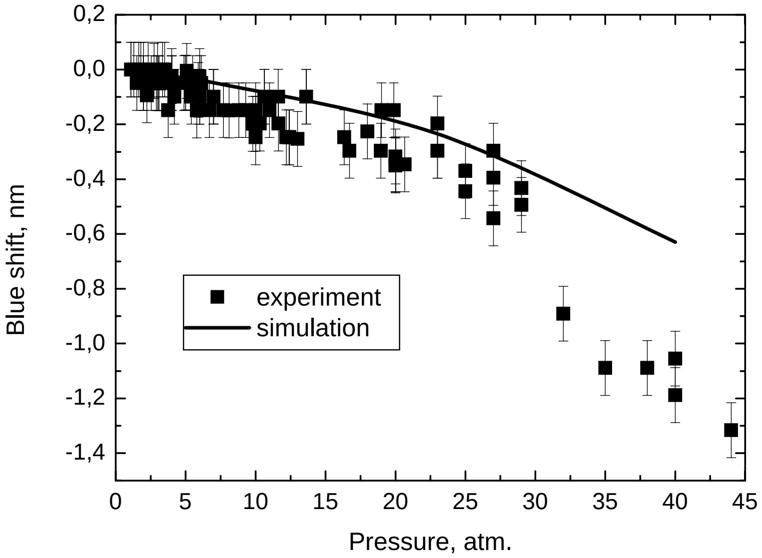

Figure 9. Blue shift of 728 nm line in negative corona in helium under different pressures. Points – experiment; Line – result of simulation.