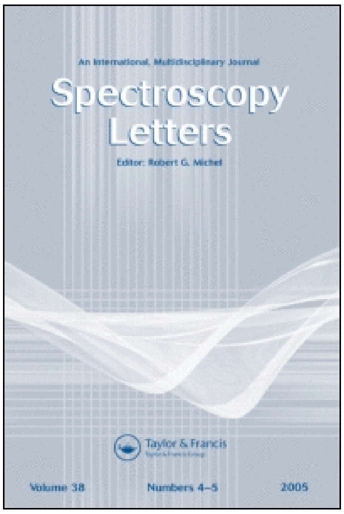

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Spatially Resolved Profile and Shift of the Spectral Line in a Hollow Cathode Discharge

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ABSTRACT Simultaneously recorded spatially-resolved profiles of He I 492.2 nm singlet line are used in hollow cathode discharge plasma diagnostic. A shift of the line center is observed. Both shift and lower pressure branch of Lorentz half-width depend equally on gas pressure and localization of the emitting plasma bulk along the radius. The observed effects are referred to the macroscopic electric field penetrating into negative glow. In this field the fast electrons of the electron energy distribution function keep their beam-like character in negative glow too.

KEYWORDS hollow cathode discharge, radial irregularities, shift of the spectral line, source function

INTRODUCTION

The profile of a spectral line emitted/absorbed in low-pressure gas discharge plasma including sputtering cells, informs on the atom- and ion-velocity distribution, the particle density, micro- and macroscopic electric field intensity, Van der Waals and resonance effects, etc.

The profiles have a significant influence on spectral interferences and on the slopes and shapes of atomic absorption and fluorescence analytical working curves.[1–3]

Analysis of the profile allows deriving information on spectral source, i.e., "*source function*" from the experimental data. The detailed information on this function also allows optimizing both the operation mode of the light source and the optical scheme geometry.[4]

Usually, the macroscopic electric field in a low-pressure gas discharge has been taken as negligible and no Stark effect in the profile taken in mind.[5] Some irregularities in the Lorentzian width of the profile under low gas pressure[6] evoked the present study.

In this work, a less studied aspect of the spectral line profile, i.e., its spatial irregularity, is analyzed. A method based on both spatially resolved and compared Fabry-Perot Interferograms (FPI) is proposed to display the radial behavior of the profile of the emission line in a hollow cathode discharge (HCD).

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The characteristic contact surfaces negative glow (NG)—cathode dark space (CDS)—cathode is a HCD attribute. Due to this peculiarity HCD is known to be a reservoir of sputtered atoms excited by the high energy electrons of the specific electron energy distribution function (EEDF). The unique sputtering-excitation properties have lately traced much interest to HCD within modern spectroscopic investigations. The present study gives evidence for a macroscopic electric field manifestation in the NG of a HCD.

EXPERIMENT

An Al cylinder of diameter $D = 18$ mm and length of 30 mm is used as HC. It is the typical HC geometry.[5] Based on the relation $D \times p =$ constant[7,8] this diameter predetermines stable HCD mode for operation under low enough critical gas pressure p. In our case the value 0.18 Torr is the critical one. In order to avoid the influence of the macroelectric field in CDS near the bottom the cylinder is bottomless.

The essence of the experiment is simultaneous recording of the profiles of one and the same line emitted from two plasma regions. It is realized by changing periodically the recording position on geometric axis using a reflecting mirror. Figure 1 illustrates the optical arrangement, which allows different regions R_i of the HCD along the radius R

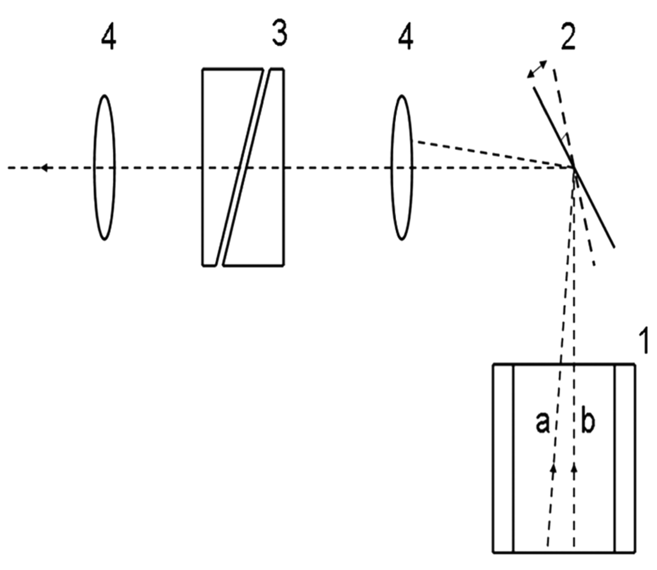

FIGURE 1 **Optical arrangement of the experiment, 1 – HCD tube; 2 – reflecting mirror; 3 – interferometer; 4 – lenses, a, *b*–positions of the variable optical axis.**

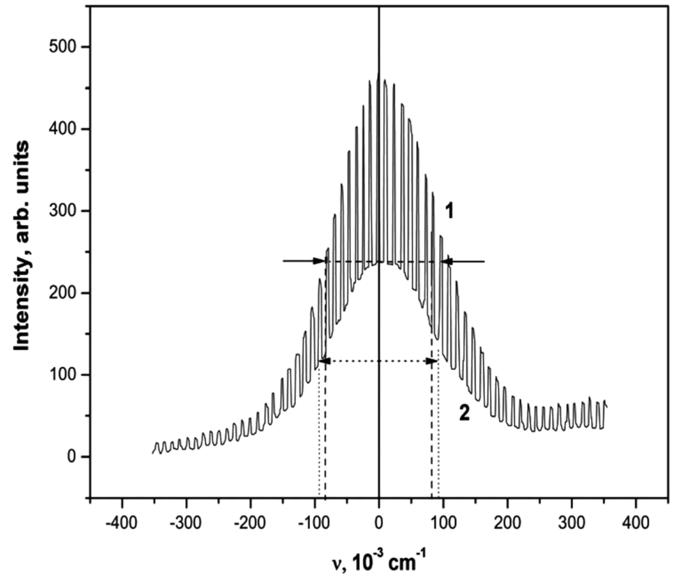

FIGURE 2 **Spatially resolved interferogram of the spectral line λ = 492.2 nm. Pressure 0.45 Torr; 1 – along the cathode axis, 2–5.0 mm off-axis.**

to be projected on the diaphragm of the monochromator entrance slit by the reflecting mirror 2 and time relay device. The latter fixes the optical axis periodically (at periods of $T = 45 \div 120$ s) along either *a* or *b* positions (Fig. 1), meaning that two profiles are recorded simultaneously (Fig. 2). It should be noted that the intensity of the He I spectral line selected, i.e., $1s4d^1D_2 \rightarrow 1s2p^1P^0_1$ ($\lambda = 492.2$ nm) attains a maximum along the cathode axis under working gas pressure $p_{He} = 0.2$–2.0 Torr. A scanning by air pressure variation Fabry-Perot interferometer and *Lock-in nanovoltmeter* type 232B ("Unipan") were used.

The exposure time T depends on the profile of lower intensity 2 (Fig. 2) and different time constant, i.e., 3, 10 and 30 s of the *Lock-in nanovoltmeter* were taken.

Then the profiles recorded 1 and 2 (Fig. 2) may be compared correctly in all parameters, including their center position. A shift of the line center becomes measurable, which is not a trivial task. The profiles of the line emitted along the geometric axis (R_o) and at distances of 5 mm (R_5) and 8 mm (R_8) have been registered at *dc* operating mode. Every FPI contains the line shape emitted from the plasma along the axis R_0—the axis of the symmetry in the cylindrical HC—as a standard and along another R_i. A cylinder with a base of 1 mm determines the spatially resolution, limited by both optical system and source geometry.

RESULTS AND DISCUSSION

The radial dependences of the emitted spectral line, i.e., its intensity $I(R_i)$, Lorentzian half-width $\Delta L(R_i)$ and center position are observed in FPI measurements. As for the radial dependence $I(R_i)$ under mentioned operating p_{He} region our experiments showed the dominant intensity along the geometric axis (at R_0) (Fig. 2), i.e., $I\,(R_0) \times I\,(R_i)^{-1} > 1$. Simultaneously, the relation $I\,(R_0) \times I\,(R_i)^{-1} \propto p_{He}^{-1}$ holds. The rest dependences are of particular interest:

Lorentzian half-width ΔL is obtained by using the deconvolution procedure in Zhechev et al.[9] The values of ΔL are found depending on both R_i and p_{He}. Figure 3 illustrates these dependences.

The inequality $\Delta L(R_8) > \Delta L(R_5) > \Delta L(R_0)$ takes place at any operating gas pressure value. Within these relations two contrary trends of $\Delta L(p_{He})$ are observed in plasma columns at R_5 and R_8, i.e., $\Delta L \propto R_i p_{He}^{-1}$ and $\Delta L \propto R_i\, p_{He}^{1}$. Obviously, the broadening $\Delta L \propto R_i p_{He}^{-1}$ at $p_{He} < 0.6$ Torr could not be related to any interactions between the particles. One can note that: the pressure 0.2 Torr is the lowest one for the stable HCD mode for operation.

Another limitation of the measurements comes from the weak line intensity under $R_i > R_8$, where the comparison with the line shape at R_0 is not accurate enough. Therefore, under low pressure $p_{He} < 0.6$ Torr Lorentzian is broadened by a factor dominating the collisional broadening and acting from CDS along the radius.

The simultaneous record of the two profiles allows the positions of their centers to be compared. The center of the line emitted along the geometrical axis (at R_0) is taken as a base. A shift $\delta\nu$ of the profile's center along the cathode radius was observed (Fig. 4). The shift is observed at pressure $p_{He} \leq 0.6$ Torr where satisfies the relation $\delta\nu \propto R_i$, p_{He}^{-1}. Table 1 illustrates the values of $\delta\nu$ vs. the combination $\{R_i, p_{He}\}$. No values of $\delta\nu \neq 0$ take place at $p > 1$ Torr. The largest shift is measured close to the CDS at $p_{He} = 0.2$ Torr.

One more special feature is related to the sign of shifting. The profile of $\lambda = 492.2$ nm line shifts to the red spectral region. In control measurements the spectral line $1s3p^1P_1^0 \rightarrow 1s2s^1S_0$ ($\lambda = 501.6$ nm) was found to shift to the blue, contrary to $\lambda = 492.2$ nm (Table 1). The same relation $\delta\nu \propto R_i$, p_{He}^{-1} takes place for $\lambda = 501.6$ nm.

The observed effects of broadening $\Delta L \propto R_i$. p_{He}^{-1} and shift $\delta\nu \propto R_i$, p_{He}^{-1} of the spectral line at low enough pressure draw attention to the macroscopic electric field in CDS. Generally, this field has been taken as localized in CDS only.

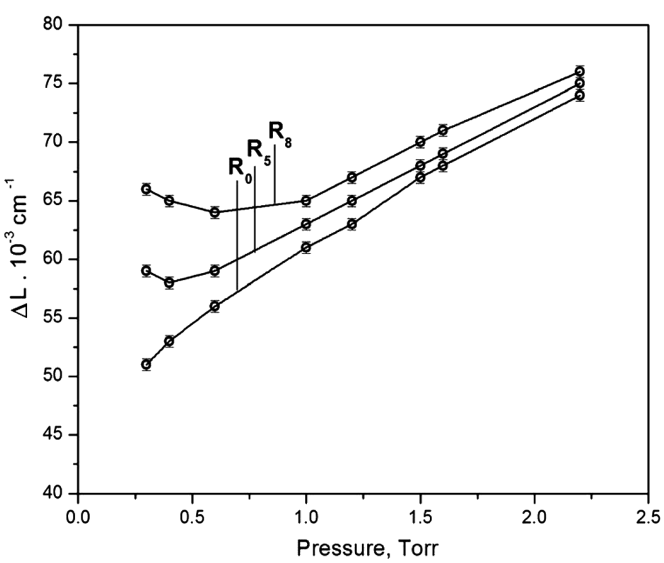

FIGURE 3 Lorentzian half-width ΔL as a function of pressure p and radius R ($\lambda = 492.2$ nm); R_0 – along the cathode axis, R_5 – distance of 5 mm from R_0, R_8 – distance of 8 mm from R_0. Discharge current 5 mA, the constant of FBI is 1 cm^{-1}.

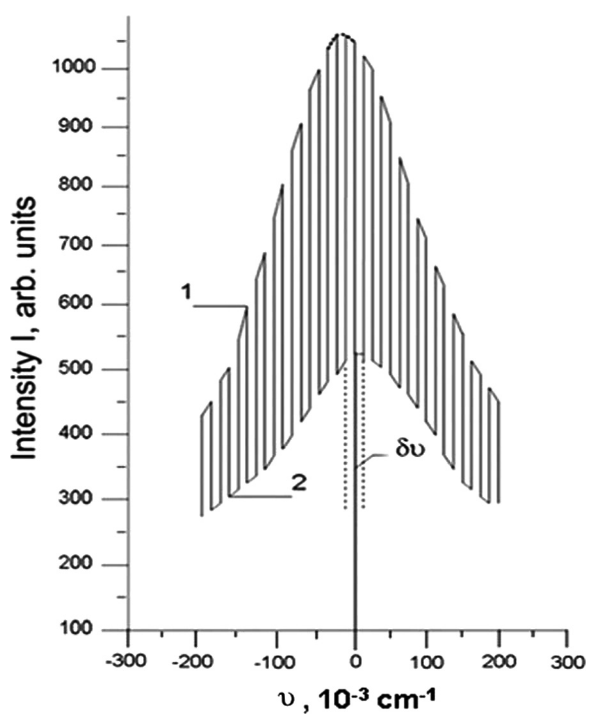

FIGURE 4 Center shift $\delta\nu$ of the spectral line $\lambda = 501.6$ nm. Emission from plasma region: 1 – along the cathode axis; 2–8 mm off-axis.

TABLE 1 Shift $\delta\nu$ of the Line Center Along the Cathode Radius R_i

He $\lambda = 492.2$ nm			He $\lambda = 501.6$ nm		
Pressure, Torr	R_i	$\delta\upsilon$, 10^{-3} cm^{-1}	Pressure, Torr	R_i	$\delta\upsilon$, 10^{-3} cm^{-1}
1.2	R_5	0	1.0	R_5	0
	R_8	0		R_8	0
0.6	R_5	0	0.5	R_5	0
	R_8	3.0 ± 2.0		R_8	3.3 ± 2.0
0.25	R_5	2.2 ± 2.0	0.2	R_5	2.4 ± 2.0
	R_8	4.3 ± 2.0		R_8	5.0 ± 2.0

In an additional experiment the enlarged optically image of NG – CDS – Cathode structure was examined closely under $p_{He} \in [0.2 \div 2.0]$ Torr. The borderline between NG and CDS turns out to be not clearly defined at $p_{He} < 0.8$ Torr due to CDS penetration to NG. This fact correlates with relation $\delta\nu \propto R_i$, p_{He}^{-1} (Table 1). Thus, the observed effects of ΔL and $\delta\nu$ are likely to be caused by the macroscopic electric field in NG penetrating from CDS.

The main argument for electric field influence is the red shift observed of $\lambda = 492.2$ nm line. The only reason is the singlet $^1F_3^0$-term (191447.24 cm^{-1}) of 4f-configuration. This singlet is close from above to the upper level $4d^1D_2$ (191440.71 cm^{-1}) of $\lambda =$ 492.2 nm. The closeness is a precondition for strong interaction between singlets. On the other hand the behavior of the upper level only, i.e., 4d $^1D_2$ determines the reaction of the dipole transition against an electric field. In our case 4d $^1D_2$ shifts to $2p^1P_1^0$ only because of the close 4f-configuration and as a result the line center shifts to the red spectral region.

The radial ΔL- and $\delta\nu$ irregularities take place in the plasma bulk between CDS and some negative glow -cylinder of $R_i \propto p_{He}$. It means that in EEDF the fast electrons, accelerated across the CDS keep their beam-like character in this CDS bulk too. Thus, at low enough values of buffer's density the beam-like electrons increase their relative contribution to the electron-atom excitation. The latter may be analyzed within the frames of the multipole representation. EEDF may be presented by spherical functions $Y_q^{(k)}$ Z(n) in velocity space[10] as: $f(\mathbf{v}) = \sum_{k=0}^{\infty} \sum_{q=-k}^{k} Y_q^{(k)}(n) f_q^{(k)}(v)$, where $n = v/|v|$ is the unit vector directed along the electron velocity vector v and $f_q^{(k)}(v)$ the multipole moments. The moment of zero order $f^{(0)}(v)$ describes the isotropic part of EEDF taking into account velocity modulus $|v|$, i.e., energy distribution; thus $f^{(0)}(v)$ characterizes the spectral properties (excitation and ionization including Penning one[11]) of plasma light emission. The moment $f^{(2)}(v)$ characterizes the electron pulse flow. This EEFD moment is specific for HCD due to the above mentioned beam-like electrons ($\approx 10^6$ cm^{-3}).[12] On the other hand the electron beam is known to generate the alignment in excited atom state,[13] i.e., coherence between the atom levels with difference of their magnetic quantum numbers equal to ± 2. This process within the frames of the discharge, i.e., without external excitation, is known as self-alignment. Coherence of type (self-) alignment manifests itself optically in the plane polarization P of the spontaneous emission I_{e_λ} from (self-) aligned states; the vector of polarization e_λ is parallel to electron velocity vector v.[14]

Thus, in addition to the other known characteristics, the self-alignment of the excited states may be found as one more inherent property of a HCD medium.

Indeed, partial polarization P of He I $\lambda = 501.6$ nm and $\lambda = 492.2$ nm lines, where $P \propto p_{He}^{-1}$ was observed. Magnetic depolarization $P = P(B)$ takes place in an external magnetic field $B \perp R$. According to Alexandrov et al.[13] and Nedelec[14] it means magnetic destruction of the self-alignment.[15]

CONCLUSIONS

A method is proposed to display the radial behavior of the profile of the emission line in a hollow cathode discharge.

The spatially resolved profiles of the emitted line $\lambda = 492.2$ nm allow for their correct comparison in all shape parameters. Moreover, their simultaneous record makes evident any relative shift $\delta\nu$ of the line center and this value may be measured.

Lorentzian half-width ΔL is found depending on the position of the emitting plasma along the radius R_i and the inequality $\Delta L(R_8) > \Delta L(R_5) > \Delta L(R_0)$ takes place at any operating gas pressure value.

Two contrary trends of $\Delta L(p_{He})$ are observed in plasma columns at R_5 and R_8, i.e., $\Delta L \propto R_i p_{He}^{1}$ and $\Delta L \propto R_i\, p_{He}^{-1}$. The latter takes place at low gas pressure $p_{He} < 0.6$ Torr. Under this pressure the lines $\lambda = 492.2$ nm and $\lambda = 501.6$ nm are observed to shift their centers along the radius R_i, i.e., $\lambda = 492.2$ nm to the red and $\lambda = 501.6$ nm to the blue. Here the shift turns out to be also $\delta\nu \propto R_i,\, p_{He}^{-1}$.

The values ΔL and $\delta\nu$ reach maximum close to the cathode dark space. Both Lorentzian broadening and shift of the line center and their dependence on the position are ascribed to the macroscopic electric field in CDS. The field penetrates into NG at low enough gas pressure.

The extended electric field is considered to stimulate the beam-like electrons in a HCD. It is a precondition for coherence of the type self-alignment of the excited atomic states.

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