Spectral line emission from the viewpoint of nonlocal electron kinetics

Yu. Golubovskii[1], D. Kalanov[1], S. Gorchakov[2], D. Uhrlandt[2]

[1]*Faculty of Physics, St. Petersburg State University, Ulyanovskaya 3, 198504 St. Petersburg, Russia*
[2]*INP Greifswald, Felix-Hausdorf-Str. 2, 17489 Greifswald, Germany*

Modern nonlocal electron kinetics theory predicts several interesting effects connected with spectral line emission from positive column in the range of low and medium pressures at varying discharge current. According to theoretical work [1] «paradoxical non-monotonic behaviour of excitation rate spatial profiles in bounded plasmas» could be expected. This hypothesis is examined in the present work through spatially resolved optical measurements of the line emission radiation from the positive column of a dc argon glow discharge. The effect of intensity radial profile broadening has been found in experiment, but intensity maximum shift predicted by theory [1] has not been found. Numerical model based on the solution of electron Boltzmann equation and a system of rate equations for excited argon atoms have been applied to study the influence of the radial potential form on the spatial distributions of excited atoms. The radial potential which satisfactory fulfils the ionization balance and describes well observed experimental data for spatial profiles of excited atoms in the whole considered pressure range was found. It is shown that the form of the potential is deciding for radial profiles of excitation and ionization rates.

1. Experimental method

Measurements of excited species densities in the positive column of an argon glow discharge were performed in the range of pressures 0.25-5 Torr and currents 1-20 mA. Discharge tube with a radius of 1.5 cm, monochromator with diffraction grid 1200 str/mm and a registration scheme with photomultiplier have been used. The used optical scheme with 15-fold image reduction and a lens corrected to aberrations provides the half-width of the instrumental function of about 0.66 mm. For systematic measurements two optically thin spectral lines have been chosen: 7504 Å (transition $2p_1 \rightarrow 1s_2$) and 8264 Å (transition $2p_2 \rightarrow 1s_2$).

Figure 1 presents examples of measured radial profiles of Ar ($2p_1$) density for different pressure and current. At the pressure of 0.25 Torr and currents below 5 mA the radial distribution has the smallest profile width. It can be assumed that at these experimental conditions the pronounced nonlocal behaviour of the electrons takes place and the so-called nonlocal approach (averaging over radial electron motions) is valid for the description of the plasma phenomena. In this case the population of the high-energy tail of the electron energy distribution function (EEDF) shows a valuable decrease in radial direction. Therefore, the zone where the excitation is significant is restricted to a narrow region near the axis. An increase of the pressure leads to broadening of the intensity and density profiles due to less pronounced nonlocal behaviour of EEDF caused by electron-electron collisions. For these higher pressures (1-5 Torr), where the energy relaxation length becomes comparable with the tube radius, profiles become close to a trapezoidal shape. Despite this fact no shift of intensity maximum has been found. Similar results have been obtained for Ar ($2p_2$) state.

2. Description of the model

For the detailed analysis of experimental results a numerical model has been developed. It was assumed that the plasma is in the steady state, the column is uniform in axial and azimuthal directions, and axially symmetric. The model comprises solution of an electron Boltzmann equation for the description of the properties of the electron component and a system of one-dimensional rate equation for the densities of the lowest excited argon atoms. The EEDF was calculated using the method presented in [2]. Electron kinetic equation was solved in the form of elliptic equation:

$$\frac{1}{r}\frac{\partial}{\partial r}\left[\frac{ru}{3N_0\sigma_\Sigma(u)}f_0\right]+\frac{\partial}{\partial\varepsilon}\left[\frac{u(eE_z)^2}{3N_0\sigma_\Sigma(u)}\frac{\partial}{\partial\varepsilon}f_0\right]$$

$$+\frac{\partial}{\partial\varepsilon}(2\frac{m_e}{M}u^2N_0\sigma_{el}(u)f_0)=$$

$$=uN_0(\sigma^*(u)+\sigma_{io}(u))f_0+S^*(r,\varepsilon,f_0),$$

$$\sigma_\Sigma(u)=\sigma_{el}(u)+\sigma^*(u)+\sigma_{io}(u),$$

$$S^*(r,\varepsilon,f_0)=(u+u_{ex})N\sigma^*(u+u_{ex})f_0(r,\varepsilon+u_{ex})$$

$$+4(2u+u_{io})N\sigma_{io}(u+u_{io})f_0(r,u+\varepsilon+u_{io})$$

where $\sigma_{el}(u),\sigma^*(u),\sigma_{io}(u)$ are cross-sections of elastic collisions, excitation and ionization (see

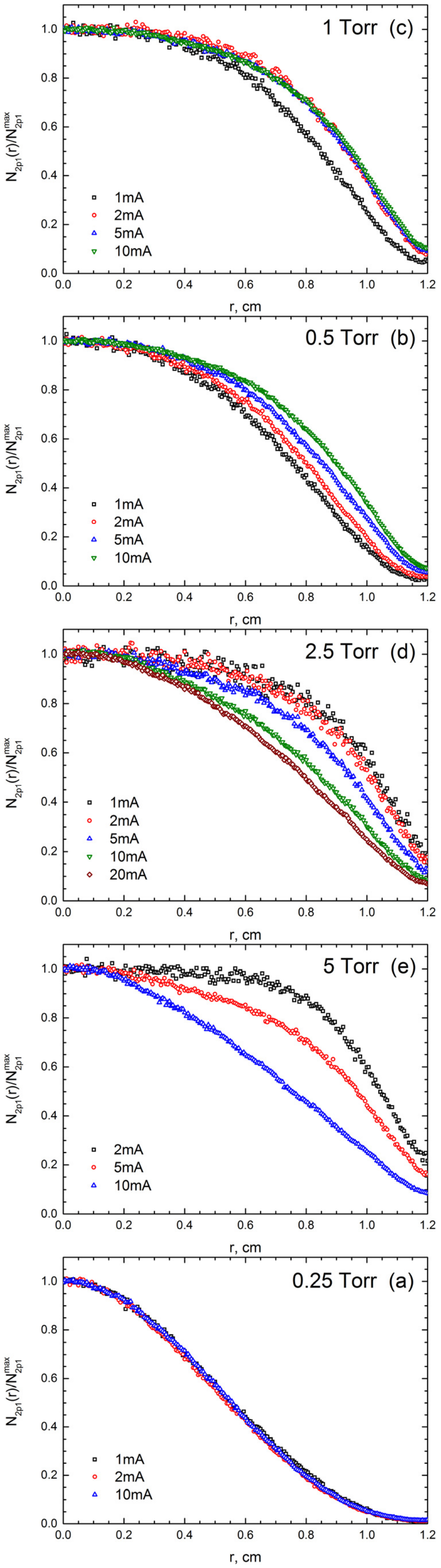

Fig.1. Radial profiles of Ar ($2p_1$) atom density for different current at the pressure of 0.25 Torr (*a*), 0.5 Torr (*b*), 1 Torr (*c*), 2.5 Torr (*d*), 5 Torr (*e*).

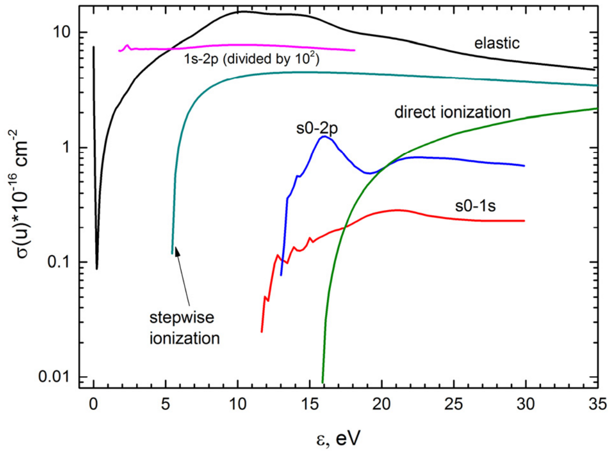

Fig.2. Collision cross-sections used in calculations. The cross-section of stepwise excitation ($1s \to 2p$) is divided by factor 100 for scaling purpose.

Figure 2) correspondingly, N_0 is the ground state density, E_z is the axial electric field strength, m_e, M are the masses of electron and neutral atom correspondingly. The excitation and ionization thresholds are denoted as u_{ex} and u_{io}. Equation was solved in the potential field of the following form:

$$\Phi(r,z) = -eE_z z + e\varphi(r)$$

The values of axial field E_z were taken from experimental data, while the radial potential $\varphi(r)$ was varied in order to describe experiment well and at the same time to provide satisfactory fulfilment of the ionization balance.

For the interpretation of obtained experimental results from the sight of nonlocal kinetics the line emission intensity has to be determined. A simplified level model of argon has been used. Radial distribution of line intensity in $2p \to 1s$ transitions can be obtained by the expression

$$I_{p,s} \sim N_p(r)A_{p,s} = N_0 z_{0,p}(r) + N_0\left[z_{0,s}(r) + z_{0,p}(r)\right] \times \frac{z_{s,p}(r)}{A_{eff} + A_{diff} + A_m + z_{s,i}(r)}$$

where N_0 denotes the ground state density; $z_{0,p}(r), z_{0,s}(r)$ are direct excitation frequencies of levels N_s and N_p; $z_{s,p}(r), z_{s,i}(r)$ are stepwise excitation and stepwise ionization frequencies; A_{eff}, A_{diff}, A_m are the probabilities of resonance emission, diffusion and chemoionization; $A_{p,s}$ is a probability of spontaneous emission in $2p \to 1s$ transitions.

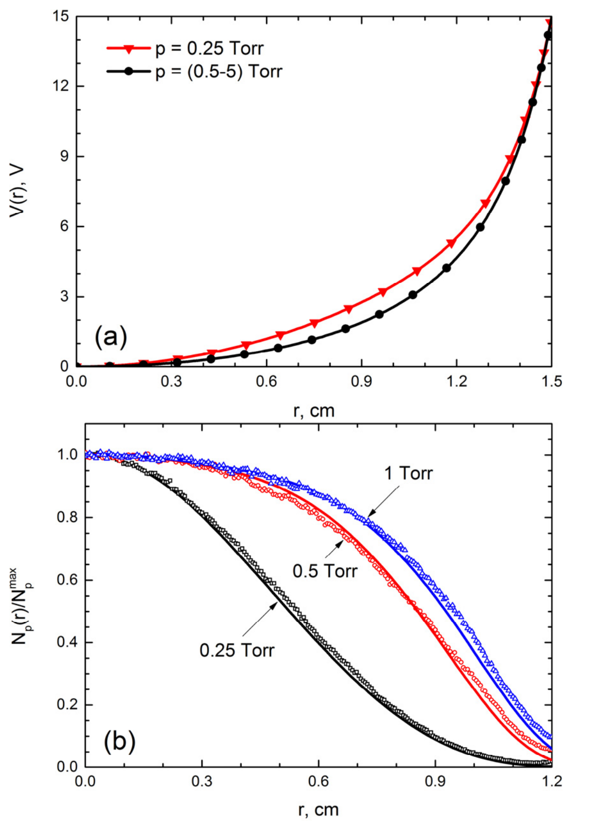

Fig.3. (a) Radial potential used in calculations. (b) Comparison of measured and calculated density profiles in Ar at current of 5 mA for different pressures. Points - experiment, solid lines – calculation.

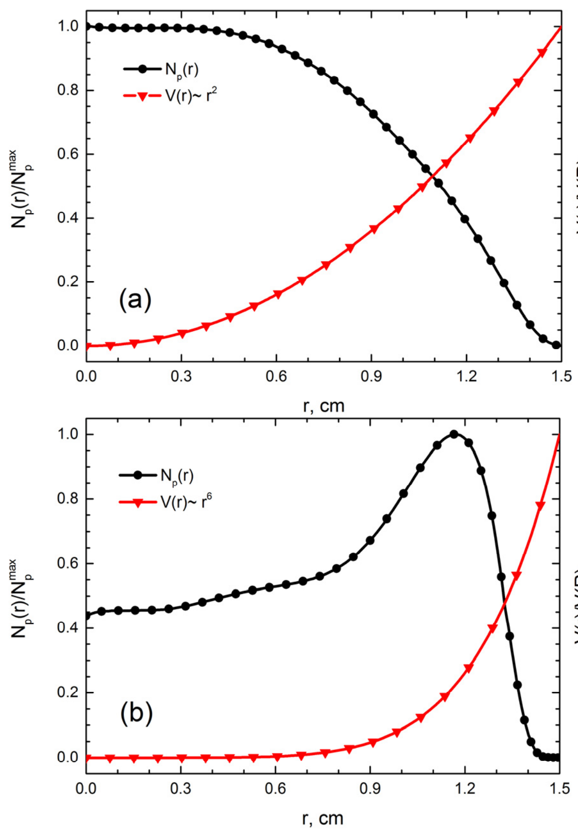

Fig.4. Normalized excitation rate profiles for $2p$ states in argon at 5 Torr calculated using different radial potential profiles (a - $V(r) \sim r^2$, b - $V(r) \sim r^6$).

3. Results and discussion

Calculations of the line emission profiles have been performed for the parameter range which corresponds to that of the experiment. Comparison of modelling results with measured data is presented in Figure 3. Chosen radial potentials provide a good agreement between predicted and measured data.

Detailed analysis of theoretical results shows that the nonlocal electron kinetics can certainly lead to a shift of the excitation rate maximum out of the column axis. But the appearance of this shift is closely connected with the form of the radial potential profile as it is illustrated in Figure 4. The off-axis maximum is clearly pronounced when the radial potential has a flat profile near the discharge axis (Fig.4b). If the radial potential increases fast near the discharge axis, then the number of fast electrons near the axis decreases (Fig.5a,c), and in the case of flat potential the density of fast electrons on periphery increases sufficiently for formation of non-monotonic profiles (Fig.5b,d).

Potentials used in calculations (Fig.3a) were selected to describe well experimental data for the line intensities. Chosen potentials also satisfactory fulfil the ionization balance equation:

$$\operatorname{div} j_i = \frac{1}{r}\frac{\partial}{\partial r} r b_i n_e(r) \frac{\partial V}{\partial r} = I(r)$$

where j_i is the ion flux in radial direction, b_i - ion mobility, and $I(r)$ denotes the ionization rate. When the ionization rate is known it is possible to restore the radial potential using following expression

$$V(r) = \int_0^r \frac{dr'}{r' b_i(r') n_e(r')} \int_0^{r'} I(r'') r'' dr'' .$$

Restored potential shows slightly different radial course in comparison with potentials chosen to describe line emission. However, the differences in particular near the axis are much smaller than the potential variations considered for the evaluation of the non-monotonic course of excitation rates (see Fig.4).

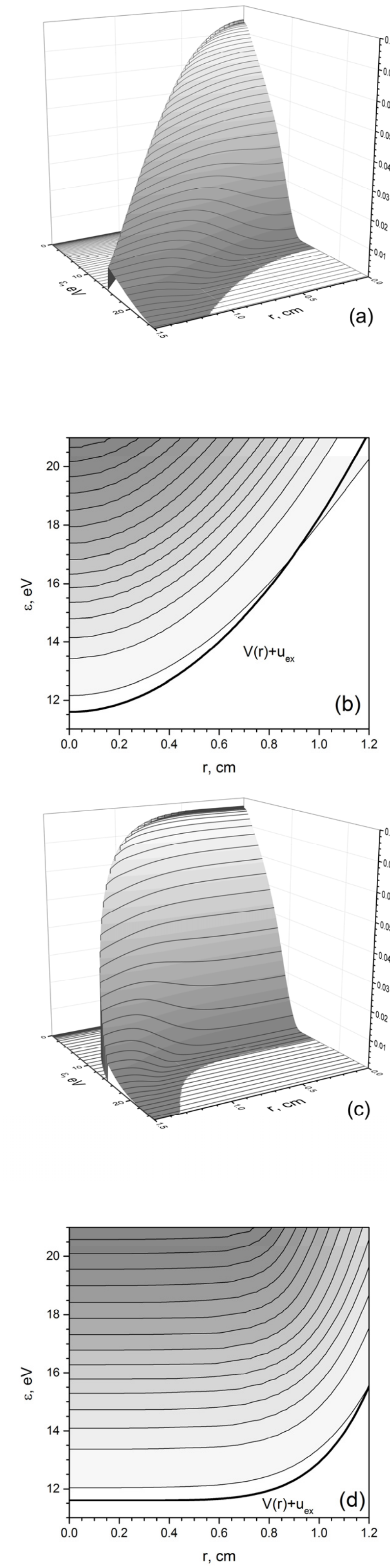

Fig.5. EEDF $f_0(\varepsilon, r)$ in argon at pressure of 5 Torr, calculated with model potential $V(r) \sim r^2$ (*a,b*) and $V(r) \sim r^6$ (*c,d*). *a,c* – linear scale, illustrating the area of low-energy electrons, *b,d* – contour plots in log scale, illustrating the changes of high-energy tail.

4. Summary

Experimental investigations and numerical modelling has been performed for a dc argon discharge in order to study the influence of the non-local electron behaviour on the spatial profiles of excited atoms. It has been shown that at realistic profiles of the radial potential a monotonous decrease of these densities occurs. Furthermore, consideration of the experimental results leads to the conclusion that radial potentials in the low-pressure glow discharge are formed in such a way that they provide monotonic radial profiles of excitation and ionization rates. Possible deviations from a monotonous behaviour, like e.g. shift of intensity maximum away from the discharge axis, occur only in the case of potential form which is far away from the reality.

5. Acknowledgements

Authors gratefully acknowledge Saint-Petersburg State University for the research grant 11.38.203.2014. A part of the research leading to these results has received funding from the European Union Seventh Framework Programme (FP7/2007-2013) under grant agreement №316216.

6. References

[1] L. Tsendin, E. Bogdanov, A. Kudryavtsev, *Phys. Rev. Letters* **94** (2005) 015001.

[2] D. Uhrlandt, R. Winkler, *Plasma Chem. Plasma Proc.* **16** (1996) 517.