Influence of radiation transport on discharge characteristics of an atmospheric pressure plasma jet in Argon

S. Valin¹, Yu. Golubovskii¹, S. Gortschakov² and F. Sigeneger²

¹Saint Petersburg State University, Saint Petersburg, Russia
²Leibniz Institute for Plasma Science and Technology, Greifswald, Germany

Introduction

Long-living excited atoms play an important role in gas discharge plasma [1]. Those atoms are involved in the numerous processes e.g. stepwise excitation and ionization, chemoionization, associative ionization, etc. The formation of spatiotemporal distributions of metastable atoms is mainly controlled by diffusion and is described by diffusion equation. In contrast to this, the population of resonant atoms is established due to numerous events of photon absorption and re-emission. The integro-differential Holstein–Biberman (H-B) equation is required for description of this process. Numerous papers are devoted to the solution of the H-B equation. Most common method is to use the effective lifetime approximation, which is accurate enough only if the spatial distribution of the excitation rate is close to a fundamental eigenfunction of H-B operator.

One of the most promising and accurate solution methods for the H-B equation consists in the transformation of the integral operator to a matrix. That leads to a reduction of the H-B equation to a system of linear equations. Efficiency of such method (the matrix method), demonstrated on the examples of a constricted dc discharge [2] and free-burning arc discharge [3], embodies the basic motivation of the current study related to a non-thermal atmospheric pressure plasma jet.

Plasma jets at atmospheric pressure have become one of the perspective objects to investigate during the last decades [4]. A huge variety of different types of these sources of plasmas, which differ with respect to geometry, applying power, discharge conditions have been studied. The field of applications of these devices is extremely wide: from local surface cleaning and activating (the application of LTE jets) to the treatment of biological targets and the deposition of thin functional coatings (non-LTE jets).

The matrix method

Resonance radiation transport is governed by Holstein-Biberman integral equation:
\[ AN_r(r) - A \int_{\Omega} K(r,r') N_r(r') d^3 r' = S(r) - D(r) = W(r), \] (1)

where \( S \) and \( D \) are rates of excitation and de-excitation of the resonance level, \( N_r \) is the resonance atoms density, \( A \) is the probability of spontaneous radiation, and kernel has the form:

\[ K(r,r') = \frac{1}{4\pi (r-r')^2} \int_0^\infty \varepsilon_v k_v \exp(-k_v |r-r'|) dv. \] (2)

Here \( \varepsilon_v \) and \( k_v \) are the profiles of emission and absorption lines.

The matrix method consists in division of the entire volume of interest into a number of small cells and calculation the coupling coefficients for all pairs of those cells. Additionally, it is assumed that resonant atoms density remains constant within one cell, so that we can rewrite integral in Eq. (1) in the following way:

\[ \int_{\Omega} K(r,r') N_r(r') d^3 r' = \sum_{\alpha} \int \int \int \frac{K(\rho, \rho')}{\Delta V_{\alpha}} \rho^2 \sin \vartheta d\rho' d\vartheta d\varphi. \] (3)

The use of spherical coordinates \( (x = \rho \sin \vartheta \cos \psi, y = \rho \sin \vartheta \sin \psi, z = \rho \cos \vartheta) \) helps to eliminate the divergence of the integral in Eq. (1) when \( r = r' \) due to Jacobian determinant and thus simplify the process of integration. Integration of the kernel in certain geometry gives us a matrix \( a_{m,n,j,i} \), such that we can rewrite Eq. (1) in the following form:

\[ \sum_{j=0}^{M_j-1} \sum_{i=0}^{M_i-1} a_{m,n,j,i} N_r(r_j,z_i) = W(r_m,z_n). \] (4)

Here, \( a_{m,n,j,i} = A(\delta_{i,j} \delta_{m,n} - B_{m,n,j,i}) \) is four dimensional coefficient matrix, \( \delta \) is Kroneker symbol, and \( B_{m,n,j,i} \) can be calculated in accordance to schematic formula:

\[ B_{m,n,j,i} = \int_0^{\psi_2} \int_0^{\theta_2(\psi,m,n,i,j)} \int_0^{\rho_2(\theta,\psi,m,n,i,j)} K(\rho, \rho') \rho^2 d\rho' d\theta d\psi. \] (5)

After the transformation of the H-B operator in Eq. (1) into the matrix, the number density of the excited atoms \( N \) can be easily found: \( N_{ji} = (a_{mni}^{-1}) W_{mn} \).

**Plasma jet model**

The plasma jet device and simulation process are described in details in the work [5]. We present here some brief explanations, which are necessary for results understanding.

Axially symmetric geometry of the device is schematically shown in Fig. 1. It can be naturally divided into two parts. The first one contains inner capillary and refers to the active plasma generation zone (Fig. 1, label “1”). The second type is a solid cylinder domain where species come in due to the gas flow (Fig. 1, label “2”).
The reaction kinetics includes 40 reactions between argon species and 11 reactions between argon species and the precursor. The reaction under investigation is radiative decay of resonant atoms: $Ar^r \rightarrow Ar + h\nu$. The trapping of resonant radiation was approximately taken into account in the original investigations [5] by an effective lifetime according to Holstein theory for a cylindrical plasma.

In order to show the influence of proper consideration of the radiation trapping effect on the spatial distribution of argon excited species, it is necessary to reconsider balance equations for species. Then, the balance equation for excited state $Ar^r$ at the position $r = (r_j, z_i)$ may be written in the form:

$$\sum_{r'=(r_m,z_n)} Ar^r (r') [A^r_{eff} a_{mnji} + \sum_{q\neq exc} Z_q^-(r') + S^{ion}(r')] = W(r) + \sum_{q\neq exc} N_q(r) Z_q^+(r).$$

Here, $A^r_{eff}$ is effective probability of spontaneous radiation, $Z_q^+$ and $Z_q^-$ are the rates of resonant level excitation and de-excitation, $S^{ion}$ is term of losses due to ionization, $W$ is gain due to excitation from ground state and recombination.

**Results**

The calculations consist of two steps. At first step, the matrix is calculated for a cylinder with inner capillary; parameters are $r_{in} = 0.9 \text{ mm}$, $R = 2.0 \text{ mm}$ and $L = 24 \text{ mm}$. Then the matrix is included into the balance equations and number densities of excited species is obtained. Results for Argon resonant atoms are shown in Fig. 2.

It is important to note, that the height of both the inner capillary and the whole domain are assumed the same. The inner capillary is not shown in the figure.

As can be seen from Fig. 2, there is well pronounced broadening of the profiles in radial direction: the resonant atoms are delivered from the center ($r = 1.45 \text{ mm}$) to the walls of capillaries in the plasma generation region.
Effect that is more important is that in the axial direction. Increasing of $Ar^r$ number density in the effluent region ($-6 \text{ mm} \leq z \leq 1 \text{ mm}$) is up to $10^5 - 10^6$ orders of magnitude. Such a strong effect may influence reaction rates with the precursor molecules coming from the inner capillary. That, in its turn, may affect the process of target treatment due to increasing the active species number density. Other excited species are less affected by the radiation trapping effect.

**Summary and outlook**

Investigations of the radiation transport influence on excited atoms spatial distribution were performed in case of finite two-dimensional axisymmetric volume. New technique of transformation of the integral operator to a four dimensional matrix was developed and implemented. The matrix method was applied to the non-LTE plasma jet. Redistribution of excited atoms shows that radiation transport plays important role in such miniaturized device and should be properly considered.

In order to complete the correct consideration of radiation transport, it is necessary to investigate the evolution of the obtained spatial distributions in time domain and to obtain a self-consistent solution. Efforts in this direction will be part of future work.

**Acknowledgements**

The work is supported by German-Russian Interdisciplinary Science Center (G-RISC). Projects no. P-2017b-22, P-2018a-18, P-2019a-20.

**References**


