Formation of UV-Radiating Strongly Non-Equilibrium Plasma with Multiply Charged Ions in the Expanding High-Pressure Gas Jet

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Abstract. In the present paper we target the physics of transition from subsonic to supersonic regimes in the expanding plasma flow. Originated form ECR ion sources, this problem is generalized to describe the stationary highly-localized plasma discharge formed under resonant microwave heating in a gas jet freely expanding after a high-pressure nozzle. The peculiar feature of such a discharge is formation of a strongly non-equilibrium plasma flow with multiply charged ions and the electron temperature much greater than the temperature of ions. We study possibilities for conjugating the slow movement of initially neutral dense gas and the supersonic flow of accelerated plasma, or, equivalently, a smooth transition from ion-acoustic barrier in the expanding flow with a varying charge state distribution. The proposed model is used to understand and optimize the recent experiments performed at IAP, Nizhny Novgorod, and aimed at demonstration of a new possibility of development of a point source of extreme ultraviolet radiation for the projective lithography. In these experiments, a line emission of multiply charged ions of noble gases has been investigated in the non-equilibrium discharge supported by high-power sub-millimeter waves in the expanding gas flow.

INTRODUCTION

Now the only practical source of extreme ultra-violet (XUV) radiation for high resolution projection lithography is line radiation of multiply charged ions of certain elements such as stannum or xenon [1]. Encouraging experiments on a development of a point-like UV radiation source have been performed at IAP [2]. These experiments demonstrate localized discharge supported by submillimeter radiation in the expanding gas jet. Essential feature of the experiments is formation of strongly non-equilibrium (T_e~100 eV and T_i~1 eV) plasma with multiply charged ions in which initial microwave power is deposited into the electron component. Despite the resonant nature of the power deposition, a high electron thermal conductivity acts against localization of a discharge in space. In these conditions, the size of XUV source is more likely governed by the fly-time of an ion in a particular ionization/excitation state rather than by the electrodynamics of resonant absorption of microwaves.

To support the experiments a stationary quasi-one-dimensional fluid model is studied in the present paper. It takes into account geometric flow expansion, electron impact ionization and line radiation of ions. The key point of the model is that physically feasible solutions should meet a transition from sub- to supersonic flow at the ion-acoustic singularity, which depends on the ion charge distribution, which in turn varies along the flow. As a result even location of the sound singularity is governed by nontrivial dynamics of the preceding flow. Solutions of the fluid equations in the vicinity of the ion-acoustic singularity are investigated and classified.

BASIC MODEL

We consider stationary fluid equations [3] for multi-component plasma produced by step-by-step electron-impact ionization. The particle balance and quasi-neutrality condition would result in

$$\partial_r(\sigma u n_i) = \sigma n_e (k_{i-1} n_{i-1} - k_i n_i)\,,\quad n_e = \sum_{i=1}^{Z_{\max}} i n_i\,, \tag{1}$$

Here r is the coordinate along the flow, n_i is the density of a plasma fraction with charge $Z=i$, $i=0,1,\ldots Z_{\max}$, n_e is the electron density, u is the flow velocity assumed equal for all fractions, k_i is the ionization constant for the i-th fraction assumed to be known function of the electron temperature, $k_{-1} = k_{Z\max} = 0$. The geometrical factor $\sigma(r)$ defines the jet expansion and is assumed to be known. In the one-fluid approximation momentum balance reads:

$$\partial_r(\sigma m_i n u^2) + \sigma\,\partial_r(n_0 T_0 + n_e T_e) = 0\,,\quad n = \sum_{0}^{Z_{\max}} n_i\,. \tag{2}$$

The ion pressure is neglected ($T_e >> T_i$). The set of fluid equations is closed assuming the constant electron temperature justified by high electron thermal conductivity typical for XUV sources.

Fluid equations (1)-(2) become singular at the critical point where the flow velocity approaches a local ion-acoustic velocity. This point may be found from

$$\partial_r(n_e/\sigma) = 0,\quad u = c\,. \tag{3}$$

Having in mind that in many practical applications the discharge is maintained in the vicinity of this critical point, we propose a technique that allows isolating the critical point in explicit way. By introducing a new variable τ such that $n_e\,dr = u\,d\tau$, the particle balance (1) is transformed to the universal form

$$\partial_\tau \gamma_i = k_{i-1}\gamma_{i-1} - k_i\gamma_i,\quad \gamma_i = \sigma u n_i/\Gamma\,, \tag{4}$$

where γ_i represents the specific flux of i-th component normalized over the total flux $\Gamma = \sigma n u = \text{const}$. Solution of these equations with constant k_i is trivial and may be expressed even in analytical form. Note that the solution is completely independent of the momentum balance. Once this solution is found, one can define the average charge and the local acoustic velocity as functions of τ:

$$\overline{Z}(\tau) = \sum_{1}^{Z_{\max}} i\,\gamma_i,\quad c^2(\tau) = \gamma_0 T_0/m_i + \overline{Z}\,T_e/m_i\,. \tag{5}$$

Thus, for the isothermal approximation the average charge unambiguously defines the charge-state distribution of multiply charged ions as well as the ion-acoustic velocity, $\overline{Z} \to \tau \to \gamma_i(\tau), c(\tau)$. It is interesting to note that for many cases the average charge may be approximated as $\overline{Z}(\tau) = \ln(1+\alpha\,\tau)$ [4].

With these definitions, the momentum balance equation (2) is transformed to

$$\begin{cases}(1 - c^2(\tau)/u^2)\;\partial_r u = -\,(\sigma/u)\;\partial_r(c^2(\tau)/\sigma)\\ \partial_r\tau = \Gamma\,\overline{Z}(\tau)/(\sigma u^2)\end{cases}. \tag{6}$$

Thus, velocity $u(r)$ of the plasma flow is reconstructed simultaneously with $\tau(r)$; then all other parameters, such as $n_e(r)$, $\overline{Z}(r)$ etc., are recovered. As expected, Eqs. (6) have singularity at $u = c$, that is the ion-acoustic transition.

CLASSIFICATION OF SOLUTIONS NEAR SOUND TRANSITION

Combining Eqs. (3) and (6), one finds the critical gradient for the ion-acoustic transition as

$$\partial_r\sigma = (\Gamma m_i/T_e)\;\partial_\tau \ln\overline{Z}\,. \tag{7}$$

This equality allows to link the coordinate r_c of the critical point with the average ion charge $Z_c = \overline{Z}(\tau_c)$ at that point, which may be considered as a free parameter at this stage. Expansion of Eqs. (6) in the vicinity of the critical gradient (7) results in the following equations for variations of Mach number and the coordinate along the flow [5]:

$$\delta M\;\partial_r\,\delta M = \alpha\,(r - r_c) + \beta\,\delta M \quad\Leftrightarrow\quad \begin{cases}\partial_\xi\,\delta M = \alpha\,\delta r + \beta\,\delta M\\ \partial_\xi\,\delta r = \delta M\end{cases}, \tag{8}$$

where $\delta M = u/c - 1$, $\delta r = r - r_c$, constant coefficients $\alpha = \frac{1}{2}\partial^2{}_r\sigma/\sigma + \frac{1}{2}\left(\partial_r\sigma/\sigma\right)^2$ and $\beta = \frac{1}{2}\partial_r\sigma/\sigma$ are calculated at the point $r = r_c$. By introducing an effective time ξ we transform the momentum balance equations to the linear form. Eigen values and typical solutions of this system for different expansion laws of the flux are shown in Fig. 1.

The most relevant to practical applications case corresponds to a saddle structures shown in the right bottom panel in Fig.1. For fast enough flow expansion ($n > 1/2$) there are subsonic and supersonic regimes, separated by separatrix along which the smooth transition from sub- to supersonic movement is only possible. Detailed analysis of such regimes, including non-linear stages far from the critical point, is done in [5]. In particular, it is possible to determine the initial conditions for the flow such that non-linear evolution of Eqs. (6) would match a particular trajectory of Eqs. (8) in the vicinity of the ion-acoustic singularity. Thus, the linearized equations may be used to classify all possible solutions of the non-linear problem.

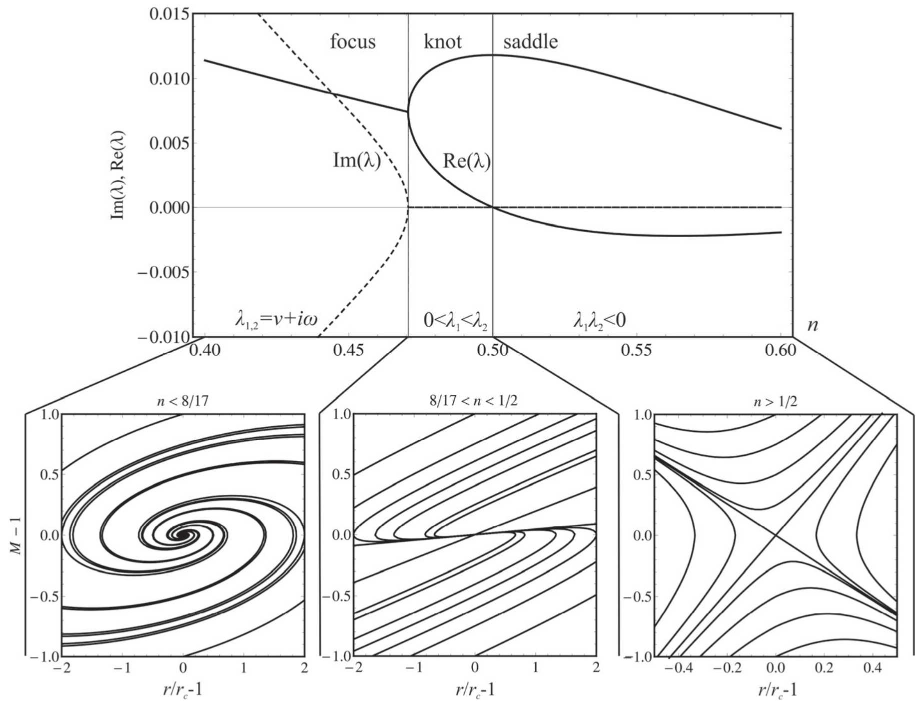

FIGURE 1. Eigen values and solutions of Eqs. (8) for different flow expanding indexes *n*, and $\sigma(r) \propto (r/r_c)^n$.

In limiting cases of strongly subsonic or supersonic flow solutions may be found analytically [4,5]. The subsonic regime is characterized by fast ionization at constant pressure, $T_e n_e + T_0 n_0 \approx \text{const}$, and spatial locality limited by variation of the flux cross-section $\Delta\sigma/\sigma < 0.2/\bar{Z}^{1/2}$. The important and unexpected feature of the supersonic regime is that due the plasma expansion the step-by-step ionization may stop far before the maximum ionization (defined by a number of electrons in a particular atom) is reached. For example, for the supersonic separatrix that starts exactly from the sound singularity the maximum increase of the average charge may be found as

$$\frac{d\bar{Z}(\tau_c)}{d\tau}\int_{\tau_c}^{\tau_\infty}\frac{d\tau}{\bar{Z}(\tau)} = \int_1^\infty \xi^{-5/2}(\xi^2-1)\exp\left(-\tfrac{1}{4}\left(\xi^2-1\right)\right)d\xi \approx 0.46 \quad\Rightarrow\quad \bar{Z}_\infty \approx 1.41\ \bar{Z}_c^{9/10} \qquad (9)$$

Far from the singularity the average charge is almost constant $\bar{Z} \approx \bar{Z}_\infty$, the flow velocity accelerates logarithmically slow $u \approx 1.44\, c_c \ln^{1/2}(\sigma/\sigma_c)$, and the electron density is governed mainly by the flux expansion $n_e \approx Z_\infty \Gamma/\sigma u$.

APPLICATION TO XUV SOURCE

As was already mentioned, in applications to experiments on XUV radiation source [2], one should inevitably consider the ion-acoustic transition inside a discharge just because there is a need for matching the subsonic flow near the gas nozzle to the supersonic plasma flow as the only possible regime for the rapidly lowering electron density. In reported experiments no oscillatory motion was resolved, so one may assume a stationary regime at least as a first approximation. Taking this into account we see that the only possible stationary solution within our model is the saddle separatrix corresponding to the positive eigenvalue (see Fig. 1, $n > 1/2$). We assume that once the physical system shows the stationary behavior, it self-organizes such that the solution follows the separatrix.

Although physics of such self-organization lies outside the scope of the studied model, we are able to construct the resultant steady-state just continuing the solution of Eqs. (6) in both sides from the saddle point. In such regimes, being of particular interest for applications, an average ion charge is consistently increased along the plasma flow, simultaneously the fraction of power losses due to line emission of highly charged ions is growing, and the emission spectrum is shifted to the shorter UV wave range. This modeling technique is developed in Ref. [5].

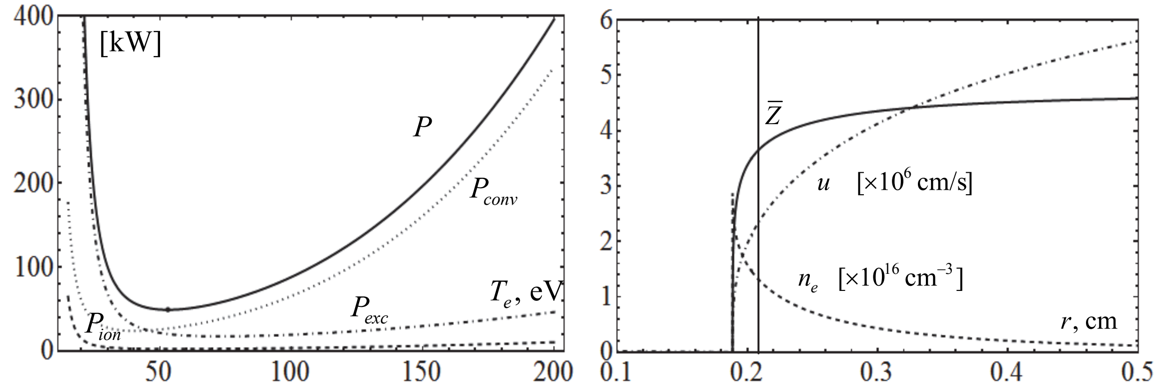

FIGURE 2. Modelling of discharge in Argon jet: the power losses versus the electron temperature (left) and the average charge, the fluid velocity and the electron density versus the coordinate along flux (right). Flux expansion is approximated by $\sigma(r) = \sigma_0 + \Omega r^2$. Total power is 50 kW, neutral gas pressure is 2.5 atm, and gas nozzle radius is 75 µm.

An example of calculations for parameters close to the experiment [2] is shown in Fig. 2. Important feature of such modeling is that fitting Eqs. (6) to the experimental data still leaves one free parameter. Namely, the position r_c of the ion-acoustic transition remains uncertain. However it may be defined from the stability criteria of the stationary discharge. Total power losses from the discharge may be estimated as a sum of the volumetric losses due to ionization and line excitation and convective losses due to plasma escape to the chamber wall,

$$P = P_V + P_C\,, \quad P_V = \int\left(\sum_{i=0}^{Z_{\max}-1} k_i E_i + \sum_{i=0}^{Z_{\max}} \sum_{\text{lines}} k_i^* E_i^*\, n_i n_e\right) n_i n_e\, \sigma\, dr\,, \quad P_C = \sigma u n\left(\tfrac{1}{2} m_i u_{wall}^2 + A Z_\infty T_e\right). \tag{10}$$

Here E_i, k_i, and E_i^*, k_i^* are ionization and excitation energies and constants, u_{wall} is flow velocity at the wall, and $A \approx 2.5 - 3.5$ slowly depends on gas composition. The convective losses are growing with the electron temperature T_e, and the volumetric losses are decreasing, both resulting in a minimum of total power over T_e. As shown in [4], both increasing and decreasing brunches are unstable, so the only possible stable solution correspond for the minimum of P over T_e. Together with the power balance, the stability condition allows to determine the remaining free parameter r_c for a given power Q deposited into the discharge from $P(T_e, r_c) = Q$ and $\partial P(T_e, r_c)/\partial T_e = 0$.

In our experiments, the developed theoretical approach allows to optimize the parameters of the high-frequency discharge for the most favorable spatial distribution of ions with different degrees of ionization, providing maximum plasma emission in a given spectral range [6].

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