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Characterization of highly transient EUV emitting discharges

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Abstract. The method of disturbed Bilateral Relations (dBR) is used to characterize highly transient plasmas that are used for the generation of Extreme Ultra Violet (EUV), i.e. radiation with a wavelength around 13.5 nm. This dBR method relates equilibrium disturbing to equilibrium restoring processes and follows the degree of equilibrium departure from the global down to the elementary plasma-level. The study gives global values of the electron density and electron temperature. Moreover, it gives a method to construct the atomic state distribution function (ASDF). This ASDF, which is responsible for the spectrum generated by the discharge, is found to be far from equilibrium. There are two reasons for this: first, systems with high charge numbers radiate strongly, second the highly transient behaviour makes that the distribution over the various ionization stages lags behind the temperature evolution.

1. Introduction

Nowadays we are used to the idea that every year a new computer can be bought for the same or lower price than that of the year before and that this new computer has a much better performance. This progress has been made quantitative by the empirical law, known as Moore's law, stating that the number of transistors on integrated circuits roughly doubles every year. This means that the structure sizes have to be reduced each year with a factor of about $\sqrt{2}$. The patterns on computer chips are obtained via lithography, which uses a radiation source to image structures. As the structures become smaller the wavelength of the radiation has to become smaller as well. At this moment lithography in commercial systems can realize structures of 90 nm, which is done by employing a radiation source generating UV at 193 nm. For finer structures and thus lower wavelengths we not only need to change the radiation sources but also the optics. The reason is that there is no material that is transparent for wavelength below 150 nm so that the lens system (refraction) has to be replaced by mirrors (reflection). But also reflective optics has its wavelength dependent limitations and an optimum is offered by multi-layer optics for a small band around 13.5 nm, a spectral region known as Extreme Ultra Violet (EUV).

This dictate, given by the optics, implies that we, plasma physicists, have to search for sources that can deliver radiation at 13.5 nm. The most promising sources are those of highly transient discharges in a Xe gas and Sn vapor. These elements have EUV transitions in the ion stages VIII – XIII that can be excited due to the dissipation of about 3 J during 100 ns in a volume of $3 \cdot 10^{-9} \text{ m}^3$. As 13.5 nm corresponds to an energy-gap of 92 eV, we can understand that these plasmas must be high in temperature. And in order to avoid large heat fluxes, the discharges have to operate in a pulsed mode. It is the aim of this paper to study these types of discharges. This characterization study will be guided by the method of disturbed Bilateral Relations [1] that is based on the competition between equilibrium disturbing and restoring processes. On the macroscopic level the equilibrium disturbance is related to the efflux of charged particles, radicals and photons out of the plasma and the competition is described by the energy and particles balances. But these effluxes have their roots on a more

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elementary level where the competition will distort the Atomic State Distribution Function (ASDF). This close relation between the macroscopic and elementary level is essential for the dBR method.

This study gives global values of the electron density and electron temperature. Moreover it provides a method to construct the atomic state distribution function (ASDF) that, being responsible for the generated spectrum, is found to be far from equilibrium. In this paper we will take the Xe-discharge as an example. However, sometimes results will be related to those found in Sn-discharges for which results of Thomson scattering are available.

2. Disturbed Bilateral Relations

The dBR method aims to find good estimates for the basic plasma properties, like the electron density n_e , the electron temperature T_e , the gas temperature T_h and the degree of equilibrium departure. With this information it is possible to get, for an important class of plasmas, insight in the ASDF and the electron energy distribution function EEDF. An important parameter for the determination of the degree of equilibrium departure is the Saha-normalized overpopulation

$$\delta b(p) \equiv b(p) - 1, \quad \text{where } b(p) \equiv n(p) / n^s(p) \quad (1)$$

equals the density of an excited level $n(p)$ with principal quantum number p divided by the value $n^s(p)$ as predicted by the Saha formula. The latter is given by

$$\eta^s(p) = (n_e/2) (n_+/g_+) [h^3/(2\pi n_e k T_e)^{3/2}] \exp(I_p/kT_e), \quad (2)$$

in which $\eta(p) = n(p)/g(p)$ is the number density of an atomic state, that is the number density $n(p)$ of a level 'p' divided by the statistical weight $g(p)$; n_+ and g_+ are the number density and the statistical weight of the ion ground-level respectively. Levels for which the density obeys Equation (2) are said to be in partial Local Saha Equilibrium (pLSE).

2.1 Steady state conditions. We start with a short outline of the dBR for the case of steady state (SS) conditions [1] by depicting the competition between equilibrium disturbing and restoring processes as follows:

$$\begin{array}{ccc} \alpha = in & & \beta = out \\ \phi_i \longrightarrow | \longrightarrow v_f & & v_b \longleftarrow | \longrightarrow \phi_i \quad \phi_i = N_\alpha v_f = N_\beta v_b \end{array} \quad (3)$$

showing that the efflux ϕ_i out of the plasma (part) is responsible for the inequality of the (internal) forward and backward processes. As explained in [1, 2] α and β can play various roles ranging from a couple of two atomic levels to, for example, the combination electrons $\{e\}$ and heavy particles $\{h\}$. The transport leak ϕ_i can be realized by the escape of photons, electron-ion pairs or the heat loss to the environment. In the case of equilibrium we have $\phi_i = \phi_i = 0$ and the diagram in Equation (3) becomes

$$\begin{array}{ccc} \alpha & & \beta \\ | \longrightarrow & & \longleftarrow | \\ N_\alpha^{eq} v_f = N_\beta^{eq} v_b; & & \end{array} \quad (4)$$

Writing Equation (3) as a balance equation for β : $N_\alpha v_f = N_\beta (v_b + v_i)$ and dividing this by Equation (4) gives

$$y(\alpha) = y(\beta)[1 + v_i/v_b] \quad (5)$$

where $\nu_i \tau_b = \nu_i \nu_b^{-1}$ is the escape per balance time ($\tau_b = \nu_b^{-1}$) while the dimensionless parameter $y = N/N^{eq}$ expresses the densities in units of the corresponding equilibrium values.

2.2. *Transient plasmas.* We will now adjust the method of dBR such that it can be applied to transient plasmas and consider ionizing plasmas, for which, due to surplus of energy input, the forward process exceeds the sum of the backward and efflux. This will lead to an *accumulation* of higher energy levels or stages, which in the following diagram is given by the vertical arrow.

$$\begin{array}{ccc} \alpha & & \beta \quad \nu_A \\ \longrightarrow & \longrightarrow \nu_f & \nu_b \leftarrow \uparrow \rightarrow \nu_i \end{array} \quad (7)$$

where ν_A refers to the accumulation frequency. In analogy with the reasoning given above we will find

$$y(\alpha) = y(\beta)[1 + \{\nu_i + \nu_A\} \tau_b], \quad (8)$$

which, just as in the SS situation, provides an criterion for the presence of equilibrium and a tool to construct distribution functions via the density ratio of the $\{\alpha, \beta\}$ system.

Moreover, we can define a criterion for the presence of Quasi Steady State (QSS): The $\{\alpha, \beta\}$ system is in QSS if $\nu_A \tau_b \ll (1 + \nu_i \tau_b)$.

3. Experimental

As an example we take a plasma created by the discharging a capacitor over a anode- cathode gap in a Xenon environment filled with a density of about $5 \cdot 10^{21} \text{ m}^{-3}$ [3]. The energy of 3 J, initially stored on the capacitor, is dissipated by the plasmas in a time period of about 100 ns. The hollow structure of the cathode is important for the very first moments of the plasma ignition but we will not consider this period here. Global optical observations reveal the following phases in plasma evolution 1) *plasma creation*, 2) the *pre-pinch*, 3) the *pinch*, and 4) *plasma decay*.

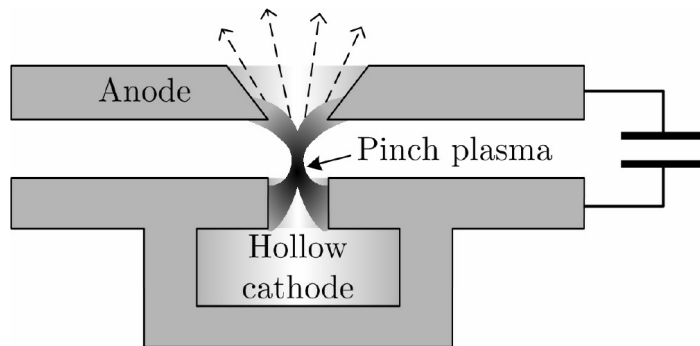


Figure 1. A sketch of the hollow cathode EUV source. The source is cylinder symmetrical.

In the first phase plasma is created with a prominent outer ring with a radius that is comparable to that of the hole in the anode which is 3 mm. In the transition to the pre-pinch phase the plasma is compressed towards a radius of about 1 mm. This implies that the density is increased with a factor of 10. During the course of phase 2) it is observed that different ionization stages are reached, even lines of Xe XIII ($Z = 13$) system are observed. The phase transition 2 -> 3 is a well-defined event in time. During a period as short as 20 ns a collapse takes place in which the radius of the plasmas is reduced

again in a factor of 3. It is believed that this collapse takes place due to radiative cooling which suddenly reduces the thermal pressure so that the pinch is free to enhance compression.

4. Global discharge model

The aim of global discharge models is to derive approximate values of *internal* plasma parameters like the electron temperature, electron density and the gas temperature out of *external* control parameters like the pressure, plasma size and power density. They are often used for SS plasmas of low degree of ionisation and the procedure is based on the approximate solution of the balances for the electron density, the electron energy and the heavy particle energy.

Here, we will follow a comparable approach that in certain sense is easier. We will concentrate on the later part of the phase 2), the pre-pinch phase.

The electron density can be obtained out of the global observations that during phase 1) the plasma is compressed by a factor 3 in radius (9 in area) and that at the end of phase 2) the mean ion charge more or less equals 10. Thus we get a good estimate of the n_e -value by multiplying the initial gas density with 100. This gives $n_e = 5 \cdot 10^{23} \text{ m}^{-3}$.

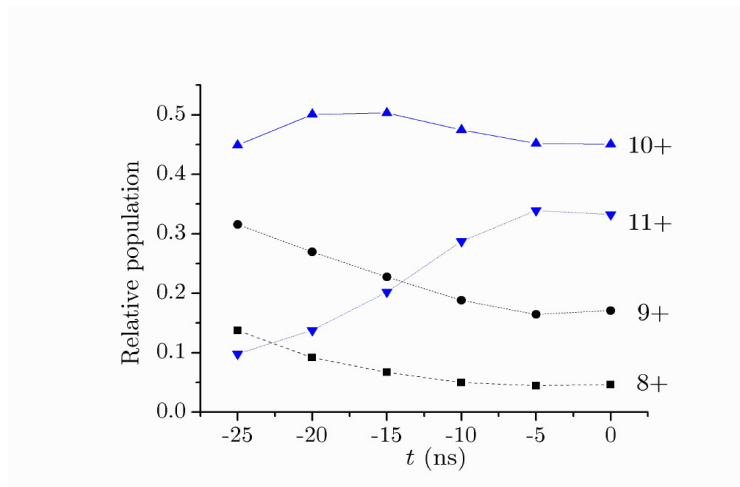


Figure 2. The evolution of the various ionization stages as a function of time; $t = 0$ is the moment of the maximum pinch. In Equation (9) we consider the $Z-1 \rightarrow Z$ transition with $Z = 10$; i.e. just before $t = -25$ ns.

Just as in the case of SS plasmas of low ionization degree we use the particle balance to derive the electron temperature. In the SS case the T_e is pinpointed by the demand that the number of electron-ion (e-i) pairs produced by (effective) ionization equals that of the (e-i) pairs that escape out of the active plasma zone. However, now for transient plasmas, the situation is different. Due to the high power density we get, before diffusive transport can take place, a rush over the ionisation stages $\text{XeI} \rightarrow \text{XeII} \rightarrow \text{XeIII} \rightarrow \text{etc.}$ Thus the charge number of the core, Z , rapidly changes in time. We will concentrate on the time interval of the $Z-1 \rightarrow Z$ shift. This can be described by the particle balance of the system Z :

$$\partial_t n_Z^+ + \nabla \cdot n_Z^+ w_Z^+ = n_e n_{Z-1} K_{ion}(Z-1; Z) - n_e^2 n_Z^+ K^{2e}(Z, Z-1) - n_e n_Z^+ K^{rad}(Z, Z-1) \quad (8)$$

where we neglected the processes $Z \rightarrow Z+1$ while $K_{ion}(Z-1; Z)$, $K^{2e}(Z, Z-1)$ and $K^{rad}(Z, Z-1)$ are the rate coefficients of ionisation, 2 electron recombination and radiative recombination. To get a T_e -value we first neglect diffusion, it is too slow and (partially) compensated by the pinch process; second we will neglect the recombination processes. Then, when the transition $n_Z = n_{Z-1}$ takes place we have

$$v_A = \partial_t n_Z^+ / n_Z = n_e K_{ion}(Z-1; Z) \quad (9)$$

For the ionization rate we can employ

$$K_{ion} = C_i(S) (8 R^2 / \pi m k T_e)^{1/2} \{4 \pi a_0^2\} (R/I^*) \exp(-I^*/kT_e) \quad (10)$$

where $R = 13.6$ eV while I^* is the effective ionisation potential. It is the excitation potential of the first level that is collisional [1]. By using the stage-dependent adjustment parameter $C_i(S)$ we can apply equation (10) for the ionization of any ion.

Before the collapse we see that a shift takes place from 9+ to 10+. So the stage Xe X with an ionisation potential of 205 eV makes place for Xe XI as the most dominant ionization stage. For $n_e = 3 \cdot 10^{23} \text{ m}^{-3}$ we get $I^* = 120$ eV. Taking $C(Xe, S) = 1$ and equating $n_e K_{ion}(Z-1; Z) = 10^8 \text{ s}^{-1}$ we find a temperature value of $\hat{T}_e = 37$ eV. This is in reasonable agreement with the results of Thomson scattering found for a Sn discharge [4].

5. The ASDF

The knowledge of the (evolution of the) ASDF is of crucial importance since it determines the radiation properties of these EUV plasmas. Two aspects have to be studied; first the distribution of the ions over subsequent ionisation stages, the so-called Z-distribution, and the distribution within each ionisation stage, the p -distribution, where p refers to the principle quantum number that gives the location of the level in the relevant ionic stage. It is defined as $p = Z \sqrt{R/I_p}$ where I_p is the ionisation potential of the ion in level p .

5.1. *The Z-distribution* can be derived via the particle balance Equation (8), which can be written as

$$n_e n_{Z-1} K_{ion}(Z-1; Z) = n_e^2 n_Z K^{2e}(Z; Z-1) + n_e n_Z K^{rad}(Z; Z-1) + \partial_t n_Z \quad (11)$$

This expression can in principle be refined by taking the traffic between Z and Z+1 into account (cf. [4]). It should be noted that the first two terms refer to the proper balance of ionization and 2e-recombination. Dividing by this Saha balance $n_e n^{eq}_{Z-1} K_{ion}(Z-1; Z) = n_e^2 n^{eq}_Z K^{2e}(Z; Z-1)$ we get

$$y(Z-1) = y(Z) \{1 + (v_{rad} + v_A)/v_b\} \quad \text{or} \quad b(Z-1) - I = (v_{rad} + v_A)/v_b \quad (12)$$

where we used the definition $y(Z-1)/y(Z) \equiv b(Z-1)$. The restoring frequency can be derived using the principle of detailed balancing giving $v_b = v^{2e} \equiv n_e^2 K^{2e}(Z; Z-1) = (n_e/n_+) n_+^3 K_{ion}(Z-1; Z)$. Thus

$$v^{2e} = n_e^2 / 2 g_+ C_i(S) [h^3 / (\pi m_e k T_e)^2] \{4 \pi a_0^2\} R (R/I^*) \exp(-I_2^*/kT_e) \quad (13)$$

For the transition Xe ($9+ \rightarrow 10+$) we get numerically $v^{2e} = 4 \cdot 10^3 \text{ s}^{-1}$ which is much smaller than v_A , the accumulation frequency, which according to figure 2 is on the order of 10^8 s^{-1} .

For the radiative recombination we use the expression of Seaton

$$K^{rad}(Z; Z-1) = 2 \cdot 10^{-20} \sqrt{(E_{ion}/kT_e)} \{0.429 + 0.5 \ln(E_{ion}/kT_e) + 0.469 \sqrt{(kT_e/E_{ion})}\} \quad (14)$$

which for the transition Xe ($9+ \leftarrow 10+$) implies that $v_{rad} = n_e K^{rad}(Z; Z-1) = 1.5 \cdot 10^6 \text{ s}^{-1}$ again much lower than v_A .

With these numerical values we find that $b(Z-1) - I = (v_{rad} + v_A)/v_b \approx v_A/v_b \approx 10^8/4 \cdot 10^3 = 2.5 \cdot 10^4$! Thus the transient behavior of the plasma leads to a severe overpopulation of lower stages. In fact this means that the distribution over the ionization stages lags behind the evolution of the temperature.

5.2. *The p-distribution.* An essential role in the determination of the ASDF is played by the collisional boundary level p_{cr} ; the level for which the number of radiative decay processes per collisional life $[v_{i,tot} \tau_{b,tot}]^B(p)$ equals unity. As explained in [1, 3] p_{cr} satisfies the condition

$$[v_{i,tot} \tau_{b,tot}]^B(p_{cr}) = A^*(p_{cr}) / \{n_e K(p_{cr})\} = 10^{23} p_{cr}^{-9} n_e^{-1} Z^6 = 1 \quad (15)$$

Levels $p < p_{cr}$ are so-called radiative levels and are determined by the corona balance (CB) whereas for $p > p_{cr}$ the levels are collisional. The position of p_{cr} strongly depends on the charge number Z . The reason is that for increasing Z the rate for radiative decay roughly increases with Z^4 whereas the collisional rate decreases with Z^2 . We will first concentrate on the CB. A level p in CB is populated by electron excitation ($l \rightarrow p$) and depopulation by means of radiative decay to lower levels ($p \rightarrow l$).

$$\begin{array}{ccc} \alpha = l & & \beta = p \\ \left| \begin{array}{c} n(1) n_e K(l,p) \\ \longrightarrow \end{array} \right. & n(2) n_e K(p,l) & \left| \begin{array}{c} \longleftarrow \\ \ll \approx \approx \end{array} \right. \\ & & \left| \begin{array}{c} n(p) A^*(p) \end{array} \right. \end{array} \quad (16)$$

As explained in [1] we can derive the CB part of the ASDF by applying the dBR method on a level in CB and the ground state; say $\{\alpha, \beta\} = \{l, p\}$ (cf. Equation (16)). In SS situations it was found that

$$y(l) = y(p) [1 + [v_i \tau_b]^B] \quad \text{with} \quad [v_i \tau_b]^B = A^*(p) / n_e K(p, l) \quad (17)$$

where $y = \eta / \eta^B$ and B refers to Boltzmann. In the case of a transient plasma we should replace $[v_i \tau_b]^B$ by $\{v_i + v_A\} \tau_b$. However, due to the Z^4 scaling we can understand that the typical hydrogen A-values of 10^8 s^{-1} will be on the order of 10^{12} s^{-1} for $Z = 10$. This means that $v_A \ll v_i$ so that the ASDF will change in a quasi steady way. Thus the ASDF in the CB domain of the ASDF will be ruled by $[v_i \tau_b]^B = A^*(p) / n_e K(p, l)$. This is a rather smooth function of p since both $A^*(p)$ and $K(p, l)$ scale with the oscillator strength of the transition.

The levels $p > p_{cr}$ are ruled by the Excitation Saturation Balance (ESB) which implies that the overpopulation ($y(p)/y(l)$) at p_{cr} with respect to Boltzmann, thus $A^*(p_{cr}) / n_e K(p_{cr}, l)$ will decrease for large p -values according to a power law $b-l \propto p^{-x}$ with x somewhere in between 5 and 6.5 [5].

6. Conclusion

The application of the method of disturbed bilateral relations gives apart from good estimates for global plasma parameters also tools to construct the ASDF of highly transient EUV emitting plasmas. Both aspects of the ASDF, the Z - and the p -distribution, are found to be far from equilibrium. However, the p -distribution evolves in a quasi-static way and can be described by a normal corona balance whereas the non-equilibrium nature of the Z -distribution is completely different. The shift of the ionization stages lags behind the temperature evolution of the plasma.

References:

- [1] Van der Mullen J J A M and Broks B H P submitted to *J. Phys. Conf. series*; this conference
- [2] Van der Mullen J J A M and Jonkers J 1999 *Spectrochim. Acta* **54B** 1017
- [3] Kieft E R, Garloff K, Van der Mullen J J A M and Banine V 2005 *Phys. Rev.* **E 71** 036402
- [4] Kieft E R, Van der Mullen J J A M and Banine V 2005 *Phys. Rev.* **E 71**
- [5] Van der Mullen J J A M 1990 *Phys. Rep.* **191** 109