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CHARACTERISATION OF PLASMAS PRODUCED BY THE "TORCHE À INJECTION AXIALE"

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ABSTRACT: Two different kinds of plasmas created by the microwave driven *Torche à Injection Axiale* (TIA) are investigated: one with helium and the other with argon as the main gas. Using absolute line intensity measurements the densities of the excited states are determined. Applying the ideal gas law gives the ground state density. It is found that both plasmas are ionising and that the excitation temperatures range from 3000 to 11000 K. The electron temperature and the electron density are determined using Thomson scattering. In the plasma with helium as the main gas, average densities between 0.64 and $5.1 \times 10^{20} \text{ m}^{-3}$ and temperatures around 25000 K are found. In an argon plasma the electron temperature is lower and the electron density is higher: 17000 K and around 10^{21} m^{-3} respectively. Radial profiles of the electron density, obtained by focusing the laser beam, appear to have a donut-like shape.

1. THE TORCHE À INJECTION AXIALE

The microwave torch called "*Torche à Injection Axiale*" (TIA), i.e. torch with axial gas injection, was developed by the group of Moisan in 1993 [1]. It consists of a coaxial structure perpendicular to a rectangular waveguide and some elements to ensure optimal power transfer to the plasma, cf. Figure 1. The TIA can, depending on the geometry of the nozzle, excite many kinds of gases or mixtures, such as air, CO_2 and noble gases. Here, we will discuss two plasmas: one with helium and the other with argon as the main gas.

Figure 1: The TIA-structure as proposed by Moisan *et al.* [1] (not on scale).

The gas flows through the inner conductor of the coaxial waveguide and the plasma is created just above the nozzle. Note that no discharge tube is used, the plasma expands in the open air. The plasma has the shape of a needle: it is approximately 2 mm in diameter and 15 to 50 mm long depending on the carrier gas and the applied power. The spectrum below 470 nm is dominated by molecular bands which arise from (association products of) molecules from the surrounding air (like the N_2^+ First Negative system, the C_2 Swan system and the CN Violet system). This indicates towards a strong interaction between plasma and the surrounding air [2].

2. ABSOLUTE LINE INTENSITIES

A helium plasma is created using an input power of 300 W and a flow of 5.0 slm. The length of this plasma is approximately 15 mm of which the turbulent upper 3 mm being is not taken into account in this paper. Only five helium lines can be measured accurately. The other lines are either too weak, or outside the sensitive range of our detection system (400 .. 800 nm), or superimposed on molecular bands. Since the plasma is very thin, it is not straightforward to measure radial distributions. Using the data listed in Reference 3 the absolute densities of the five states can be determined. The result is depicted in Figure 2a. It looks as if the atomic state distribution obeys a Boltzmann relation which results in a temperature $T_{\text{spec}} = 3800 \pm 200$ K. This spectroscopically determined temperature is slightly higher than the temperature found by Quintero *et al.* using relative line intensities: 3100 ± 150 K [4].

Figure 2: The measured ASDF of the helium plasma created by the TIA (measured at 5 mm above the nozzle). The state occupations depicted in figure a seem to obey a Saha-Boltzmann relation. In figure b the ground state density based on the ideal gas law (hollow circle) is added to the ASDF: strong deviations from Saha-Boltzmann can be observed.

Since absolute densities were measured, it is possible to include the density of the ground state using the ideal gas law. The gas temperature is unknown, but it can be roughly estimated by the rotational temperature of the N_2^+ molecule: 3000 ± 1500 K [4,5,6]. A large uncertainty margin is taken, since it is not known whether the rotational temperature is a good indication of the gas kinetic temperature. This yields a ground state density equal to $n_1 \approx (2.4 \pm 1.2) \times 10^{24} \text{ m}^{-3}$. In Figure 2b the value of the ground state density is combined with results obtained from the spectroscopic measurements. The temperature deduced from the ratio of the densities of the lowest measured excited state (i.e. the third level) and the ground state is found to be $T_{13} = 11400 \pm 300$ K. Its accuracy is purely determined by the unknown gas temperature.

Since at least two temperatures are necessary to describe the Atomic State Distribution Function (ASDF), it does not obey a Saha-Boltzmann relation. The ASDF depicted in Figure 2b is characteristic for an ionising plasma [7,8]. It can be shown that in this case the obtained excitation temperatures are underestimations for the electron temperatures. Moreover, measuring T_{spec} in a strongly ionising plasma using these levels will always yield a temperature around 3800 K [8].

In Figure 3 the temperatures describing the ASDF are depicted as a function of height above the nozzle (AN), i.e. the axial position. At every point, two

Figure 3: The temperatures describing the ASDF (see Figure 2) versus the height above the nozzle (AN). The error bars of T_{spec} correspond to the standard deviations in the fitted slopes; those in T_{13} stem from the uncertainty in the gas temperature.

temperatures are needed to describe the ASDF. Therefore it can be concluded that at every measured position the plasma is not in Local Saha Equilibrium (LSE).

Also the ASDF in argon plasmas is described by two temperatures: one for the states determined by spectroscopy $T_{spec} = 4860 \pm 150$ K and one for the ratio between the ground state and the first measured excited state (i.e. the third level): $T_{13} = 8700 \pm 300$ K (at 1 mm AN using 330 W input power and a flow of 3.0 slm).

3. THOMSON SCATTERING

In contrast with absolute line intensities, Thomson scattering [9] is not influenced by the absence of equilibrium. Therefore this technique was used to obtain the electron density and temperature. The setup that was used is treated in Reference 9.

In Figure 4a the electron temperature in the helium plasma is depicted as a function of the height above the nozzle (AN). The temperature is more or less stable around 25000 K and drops to 19100 K at 5 mm AN. The accuracy of the fits is approximately 300 K. As can be seen in Figure 4b, it appears that the electron density decreases from (5.1 ± 0.8) at 1 mm to $(0.64 \pm 0.10) \times 10^{20} \text{ m}^{-3}$ at 5 mm AN. Rodero *et al.* [5,6,10] used the Stark broadening of H_β to determine n_e in this helium plasma. Under similar conditions (3.0 slm, 300 W) they found a slightly lower density: $(4.2 \pm 0.2) \times 10^{20} \text{ m}^{-3}$ just above the nozzle.

Figure 4: The electron temperatures and densities in the helium plasma.

In the argon plasma the electron temperature remains stable around 17000 K, except for a strange dip between 5 and 6 mm AN (cf. Figure 5a). On the other hand the electron density decreases from (22 ± 3) to $(3.5 \pm 0.5) \times 10^{20} m^{-3}$, as can be seen in Figure 5b.

As expected in both plasmas the electron temperatures determined by Thomson scattering are much higher than the excitation temperatures presented in the previous section ($T \leq 11900$ K and $T \leq 8700$ K for helium and argon respectively). Again this indicates that the plasmas produced by the TIA are far from equilibrium as will be discussed more quantitatively in the next section.

Figure 5: The electron temperatures and densities in the argon plasma.

4. PARTICLE BALANCE

The electron temperatures found in the previous section for the helium plasma (19100 to 25000 K) are much higher than the temperatures found by Rodero *et al.* using a modified 2λ method: 11000 to 12500 K [5,6]. In this section we will compare the production of free electrons with their losses due to recombination, diffusion and convection. It will be shown that a high electron temperature is required to sustain the plasma.

The particle balance for charged particles is given by

$$n_e n_i S_{CR} - n_e n_+ \alpha_{CR} = \nabla \cdot (D_a \nabla n_+) + \nabla \cdot (n_+ \mathbf{w}_p) \quad (1)$$

In this equation the terms on the right hand side represent the efflux due to ambipolar diffusion (dominantly in radial direction) and to convection respectively and the terms on the left hand side the net production. The ionisation (S_{CR}) and recombination (α_{CR}) coefficients can be estimated using a collisional radiative model [7]. For helium we use the model of Drawin and Emard [11] and for argon the one by Benoy *et al.* [12]. The losses of free electrons due to convection, i.e. the second term at the right hand side of (1), can be estimated (assuming no multiple ionised particles) by

$$\nabla \cdot (n_+ \mathbf{w}_p) \approx \frac{n_e}{h_p} w_p \quad (2)$$

In which h_p is the height of the plasma. The plasma flow velocity w_p is estimated by the gas flow, the cross section of the nozzle and the heating of the gas by the plasma. The other loss term, i.e. the efflux due to diffusion can be written as

$$\nabla \cdot (D_a \nabla n_+) \approx \frac{n_e}{d^2} D_a \quad (3)$$

The ambipolar diffusion coefficient D_a is given by [13]. We assume that the characteristic diffusion length d equals the radius of the plasma. Using these approximations and T_e and n_e determined by Thomson scattering, the magnitude of the terms of the particle balance can be estimated. These results are listed in Table 1.

	helium	argon
ionisation	2×10^{26}	3×10^{27}
recombination	3×10^{21}	7×10^{22}
convection	6×10^{24}	1×10^{25}
diffusion		3×10^{24}

Table 1: Typical values of the separate terms of the particle balance (in $m^{-3}s^{-1}$), using T_e and n_e of Section 3. The diffusion loss rate is obtained using a gradient length of 1 mm.

Figure 6: The radial electron temperature and density profiles in the argon plasma at 2 mm AN, obtained by Thomson scattering. The spatial resolution is determined by the entrance slit of the monochromator (0.3 mm). The density profile seems to have a donut like shape, which enhances the losses due to diffusion.

much shorter than 1 mm. Note that the spatial resolution is 0.3 mm, so that the actual diffusion length is probably shorter. This means that the diffusion losses become of the same order of magnitude as the production by ionisation, see Table 1.

Although Rodero *et al.* found similar electron densities in the helium plasma, they found electron temperatures around 12000 K using a modified 2λ method [5,6]. As is discussed in section 0 using passive spectroscopy to determine T_e in these plasmas is questionable. Moreover, for this temperature the ionisation rate becomes $2 \times 10^{22} m^{-3} s^{-1}$ [11], which is even too small to balance the losses due to convection ($6 \times 10^{24} m^{-3} s^{-1}$, cf. Table 1). So a much higher T_e is needed to sustain this plasma.

5. CONCLUSIONS

The plasmas created by the *Torche à Injection Axiale* are far from equilibrium. The electron temperature is much higher than the gas temperature. Moreover, its small effective size causes huge losses of charged particles due to diffusion (typical residence time of a free electron is in the order of 1 μs). In order to sustain the plasma these losses have to be balanced by a strong ionisation rate. This flow through the excitation space gives the atomic state distribution function its typical shape for an ionising plasma, which causes that the measured excitation temperatures are much lower than the electron temperature.

Comparing the ionisation with the loss terms it seems that the production outranges the destruction by two orders. However, the actual losses can be higher due to:

1. mixing with the surrounding air,
2. formation of molecular argon ions,
3. a smaller gradient length.

ad 1

As is discussed by Timmermans *et al.* [2] extra destruction channels for the free electrons are created if the surrounding air is mixed with the plasma. For instance, for argon and nitrogen this mechanism is charge transfer followed by dissociative recombination. The charge transfer reaction is resonant since the ionisation energies of argon and molecular nitrogen are almost equal (respectively 15.68 and 15.58 eV). The second reaction is fast due to the Coulomb interaction between the positively charged molecule and the electron. The presence of this mechanism is supported by the fact that in the argon plasma the First Negative system of N_2^+ and many atomic nitrogen lines can be observed [2,4,5,6].

ad 2

A second way of losing charged particles is the formation of rare gas molecular ions followed by their destruction via dissociative recombination. Normally, the formation of the molecular ion is the limiting process, which rate is around $10^{27} \text{ m}^{-3}\text{s}^{-1}$ in the argon plasma (neutral density equal to $2.4 \times 10^{24} \text{ m}^{-3}$). However, since the formation of molecular ions is accompanied by its inverse process, the effective destruction rate is much lower [14].

ad 3

An other possible explanation for the large discrepancy between the production and the destruction of ions and free electrons might be that the actual radial gradient length is smaller than the value of 1 mm, as used for estimating the diffusion losses in Table 1. An indication of the gradient length is obtained by the skin effect: the penetration of the microwaves into the plasma is limited, since they are absorbed by the free electrons. According to Jackson [15] the skin depth is given by:

$$\delta = \sqrt{\frac{2}{\mu_0 \omega \sigma}} \quad (4)$$

For an argon plasma at our conditions the relative magnetic permeability μ_r equals 1 and its conductivity σ approximately $10^4 \text{ } \Omega^{-1}\text{m}^{-1}$ [16]. Due to the high frequency of the microwaves (2.45 GHz) the skin depth is only 0.1 mm. In order to measure the radial gradient length Thomson scattering measurements are performed using a focused laser beam. It is found (cf. Figure 6) that the radial gradient length is indeed

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