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Experimental study of the influence of the cathode in the characteristics of the cathode region

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Abstract. Doppler free two photon optogalvanic measurements of the Stark splitting of the 2S level of hydrogen are used to determine the local electric field strength (E-field) in the cathode fall region of a hollow cathode discharge operated in pure hydrogen. The aim of these measurements is to study how the cathode fall characteristic depends on cathode material (stainless steel and tungsten) and cathode diameter (10 and 15 mm). The measurements revealed that the cathode diameter has a minor influence whereas the cathode fall characteristics obtained for stainless steel cathodes are remarkably modified due to sputtering.

1. Introduction

The local electric field strength (E-field) in the cathode fall region is an important parameter for low density discharges of technological interest, because it controls electron and ions fluxes and their energy distributions in front of the cathode. Precise E-field measurements are demanded therefore to test plasma modelling and for better understanding of plasma surface interactions. Furthermore, atomic hydrogen plays an important role in many discharges of industrial relevance because of its high reactivity. Among different experimental techniques for plasma diagnostics, laser-aided spectroscopic techniques can provide outstanding high temporal and spatial resolution, see for instance [1-6].

Our group at the Valladolid University has been developing for a long time high resolution Doppler-free laser-spectroscopy techniques for plasma diagnostic purposes based on the 1S – 2S transition of hydrogen isotopes. Here we report on E-field measurements conducted in a hydrogen hollow cathode discharge (HCD) in the abnormal glow discharge regime. The E-field is determined via Stark splitting of the 2S level of hydrogen; Doppler-free two-photon absorption is provided by two counter-propagating circularly polarized beams of opposite directions followed by optogalvanic detection. The main aim is to study for the first time systematically the influence of cathode material and cathode diameter on the cathode fall characteristics in a wide range of discharge parameters.

2. Experimental arrangement

The whole experimental arrangement is explained elsewhere [7, 8], therefore only some important details will be explained here. The plasma is generated in a homemade hollow cathode discharge (HCD) that provides an improved quiet discharge (without any anode or cathode sparking) with excellent long and short term stability and reproducibility in a fairly wide range of discharge parameters. To achieve this, the discharge is operated symmetrically, i.e. the cathode is placed between two peaked stainless steel anodes (see Fig 1 in Ref [7]) and a continuous gas flow of about $10 \text{ cm}^3 \cdot \text{s}^{-1}$ enters at one anode side and leaves at the opposite one. Different exchangeable cathodes



allow for studying the influence of different cathode diameters (10 and 15 mm) and cathode materials (stainless steel and tungsten) on the cathode fall characteristics. The radiation required for the spectroscopic measurements is provided by a complex OPO-OPA laser system pumped by a 10 Hz injection-seeded Q-switched Nd:YAG laser. Finally the 243 nm radiation is generated by sum-frequency generation in a BBO crystal. This laser spectrometer delivers high power single longitudinal mode (SLM) 243 nm radiation, with pulse duration of 2.5 ns and 300 MHz bandwidth. The SLM condition is controlled by an etalon of 7 GHz free spectral range and a monitor.

The high resolution Doppler-free measurements are achieved with two counter propagating laser beams, circularly polarized in opposite senses according to the selection rules ($\Delta L = 0$) of the 1S-2S hydrogen transition. Both beams are focused, with two lenses of 1 m of focal length, in the upper central part of the HCD in a tiny overlapping volume, parallel to the cathode surface. Two linear translators, horizontal and vertical, ensure the best alignment and the possibility to take measurements at different distances from the cathode surface.

Optogalvanic detection consists of measuring the short time variation of the entire discharge impedance on a 50 ns timescale related to the increase in the number of electrical charges in the tiny measurement volume (about 100 μm in diameter and 10 mm length) due to resonant two-photon excitation and subsequent photo-ionization taking place during the laser pulse. The E-field strength determination is achieved by measuring the distances between the components ($2P_{1/2}$ and $2P_{3/2}$) of the 2S level of hydrogen due to the Stark effect and comparing it with the theoretical values [9]. The detection limit of this technique is about 0.5 kV/cm.

The laser pulse energy is a critical parameter and has to be controlled carefully to ensure the best spatial resolution. It has been demonstrated [10] that 80 % of the optogalvanic signal comes from the overlap volume of only 100 μm in diameter and 10 mm length if the laser pulse energy is kept below (45 μJ) i.e. irradiance of 150 MW/cm^2 in the overlapping volume. If the irradiance exceeds this value, the measured Stark components will suffer an additional broadening and shifting. In the case of tungsten one observes only a small additional broadening which is related to saturation due to the depletion of atoms in the fundamental level and the life time reduction of the 2S level due to photo ionization [11], hence the E-field determination presents negligible errors. On the other hand, this irradiance limit has been found to be critical for stainless steel cathodes. With irradiances above the limit, the Stark components suffer an increasing broadening and also a considerable shifting, that leads to an unacceptable overestimation of the E-field. Neither of the two effects can be related to saturation.

3. Results

The cathode fall characteristic, i.e. variation of the E-field with distance from the cathode surface was determined in a wide range of discharge conditions using cathodes of tungsten and stainless steel with inner diameters of 10 and 15 mm. Discharge currents varied from 50 mA to 300 mA at discharge pressures of 400, 600 and 900 Pa. The E-field reaches its maximum at the cathode surface, and decays mostly parabolically with increasing distance from the cathode.

The largest E-field values (about 4 kV/cm) were obtained for the largest discharge currents of 300 mA, quite independent from the discharge pressure. The slowest decay of the E-field was observed for 400 Pa and the lowest discharge current (50 or 75 mA). Compared with previous works, measurements down to the estimated detection limit given by the Stark splitting of 2S level, i.e. E-field strength of 0.5 kV/cm could be performed.

The variation of the cathode fall characteristics can be expressed quite well by three parameters:

- The maximum E-field at the cathode surface
- The extension d_{cf} of the cathode fall defined by zero E-field given by a parabolic or lineal fit to all measured values.
- The entire cathode fall voltage V_c , given by the integral of the E-field versus the distance from cathode surface.

Careful analysis of the cathode fall characteristics with respect to cathode material and cathode diameter revealed very clear trends that depends on the discharge parameters which are explained in detail in the following sections.

3.1 Influence of the cathode inner diameter.

For this study, the discharge is operated for both inner diameters with the same current densities at the cathode surface: e.g. 50 mA for 10 mm corresponds to 75 mA for 15 mm, both leading to a current density of 3.18 mA/cm². The slopes of the cathode falls are quite similar for both cathode materials and for both diameters [7]. Table 1 shows the summary of the results obtained for the three pressures and the three current densities in tungsten cathodes, within the corresponding ratios of the entire cathode fall voltage for 15 and 10 mm (V_{c15}/V_{c10}). This ratio is almost 1 within the measurement uncertainty. More pronounced differences were found for extreme discharge conditions: high pressure and low current and the opposite, low pressure and high current ($V_{c15}/V_{c10}=1.24$ for the highest current density in 400 Pa). However, the total mean value for all ratios is 1.05 with a standard deviation of 10%.

This comparison shows that the cathode fall characteristics do not depend on the cathode diameter for most of the experimental conditions; hence they are independent of discharge geometry. Therefore, they are well suited to be compared with other discharges geometries, especially with plane-parallel electrode designs. This conclusion is furthermore confirmed by the agreement of our results with measurements of other authors - mostly using plane parallel discharges - covering a wide experimental range (see [12, 13] and the references therein). However, the comparison done with different cathode materials shows that the entire cathode fall voltage is always smaller for stainless steel than for tungsten due to sputtering [8], see following section.

Table 1: Cathode fall voltages drop in tungsten cathodes for the two diameters and equal current density j , and the corresponding ratio.

j (mA cm ⁻²)	400 Pa			600 Pa			900 Pa		
	V_{c10}	V_{c15}	V_{c15}/V_{c10}	V_{c10}	V_{c15}	V_{c15}/V_{c10}	V_{c10}	V_{c15}	V_{c15}/V_{c10}
3.18	269	280	1.04	268	239	0.89	213	224	1.05
6.37	303	350	1.16	273	271	0.99	242	244	1.01
9.55	346	428	1.24	304	345	1.14	266	252	0.95
Mean Ratio	1.15			1.00			1.00		
Total Mean: 1.05, 10 % Standard deviation									

3.2 Influence of cathode material.

Similar to the preceding section we are comparing now the cathode fall characteristics for the two cathode materials: tungsten and stainless steel. In general, for stainless steel cathodes of 10 mm inner diameter, reliable optogalvanic measurements were not possible for the same wide range of discharge conditions as for tungsten, because the background noise is much larger. However, stainless steel cathode, 15 mm diameter, provides more stable conditions, and measurements could be performed for the same discharge conditions as for tungsten, except for 75 mA at 900 Pa.

The comparison shows very clearly some general trends. The E-field strength is always higher for tungsten than for stainless steel, and the cathode fall length d_{cf} decreases with pressure and current. These trends can be analysed by the entire cathode fall voltage V_c . In table 2, the V_c values for the three discharge pressures and four discharge currents in the range from 75 to 300 mA are presented. For 400 and 600 Pa the E-field slopes for the two cathode materials become remarkably separated with increasing current. That means that the ratio $V_c(T)/V_c(S)$ increases with increasing discharge current, being more pronounced for 400 Pa. Nevertheless, for 900 Pa the cathode falls are quite similar for

both cathode materials and all discharge currents; the ratio is fairly independent of the discharge current, having a mean value of 1.13 ± 0.04 .

Table 2. Cathode fall voltages (V_c) for stainless steel (S) and tungsten (T) cathodes in 15 mm and its ratio.

j (mA cm ⁻²)	400 Pa			600 Pa			900 Pa		
	$V_c(T)$	$V_c(S)$	$V_c(T)/V_c(S)$	$V_c(T)$	$V_c(S)$	$V_c(T)/V_c(S)$	$V_c(T)$	$V_c(S)$	$V_c(T)/V_c(S)$
3.18	280	232	1.21	239	195	1.23	224	-	-
6.37	350	248	1.41	271	205	1.32	244	213	1.15
9.55	428	262	1.63	345	229	1.51	252	233	1.08
12.73	444	261	1.70	386	242	1.60	304	268	1.13

A more detailed analysis of these trends is given in a recent study [8] coming to the conclusion that the differences between the cathode fall characteristics for tungsten and stainless steel can be explained by sputtering of the stainless steel cathode. Compared to tungsten, sputtering of stainless steel is about 10^4 times larger and its diffusion into the plasma is around 3.3 times faster for iron, because of the large difference of atomic mass: being 56 for iron and 184 for tungsten. On the other hand, the ionization energy of both materials is only about the half that for atomic or molecular hydrogen. As a consequence of these properties, the presence of sputtering is larger and its effect is much more pronounced in stainless steel. This contamination of the hydrogen plasma modifies the whole cathode fall characteristics. Due to the presence of this sputtering, we can conclude that the plasmas generated in stainless steel cathodes cannot be considered as pure hydrogen discharges; meanwhile those generated in tungsten cathodes can provide nearly pure hydrogen plasmas.

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