Characterization of hollow cathode fall field strength measured by Doppler-free two-photon optogalvanic spectroscopy via Stark splitting of the 2S level of Hydrogen and Deuterium

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Abstract. Doppler-free two-photon optogalvanic spectroscopy has been applied to measure the strong electric field strength and the cathode fall characteristics of hollow cathode discharges operated in hydrogen and deuterium via the Stark splitting of the 2S level of atomic hydrogen isotopes. In this paper we show similarities and differences in the tendencies of the cathode fall characteristics of hydrogen and deuterium in a wide range of identical discharge parameters.

1. Introduction
The electric field strength (E-field) is one of the most relevant discharge parameters directly connected with charged particle fluxes, their energy distribution functions and charge densities. Spatial distribution of the E-field and its dependence on the plasma parameters are of basic importance for plasma discharge modelling and surface treatment technologies. Spectroscopic techniques for determining the E-field are based on the Stark effect of atomic or molecular spectra, i.e. splitting, shifting and intensity variations of spectral line components depend on the local E-field. Comparisons of the measured spectra with theoretical or calibrated values give the E-field. Laser spectroscopic techniques can provide high spectral, spatial and temporal resolution. They do not perturb the investigated plasma if nanosecond pulse duration and moderate irradiances are applied. Most of these techniques are based on the spectroscopic measurement of the Stark splitting, shifting or mixing of Rydberg states in different kinds of atoms and molecules using optogalvanic and/or fluorescence detection, see e.g. the overviews in references [1, 2]. Atomic hydrogen isotopes and helium are very good candidates for E-field measurements, because the Stark splitting of all their atomic levels is very well known, and atomic hydrogen also participates in many plasmas of industrial interest. In fact, different techniques for atomic hydrogen and helium have been published most of them included in [1, 2] and [3-10] respectively, and the references therein.

In this work we propose Doppler-free two-photon optogalvanic spectroscopy to measure the strong electric field strength (0.4 kV/cm to 4 kV/cm) in the cathode fall of hollow cathode discharges (HCD) via the Stark splitting of the 2S level of atomic hydrogen and its isotopes. This requires advanced pulsed tunable single longitudinal UV-laser spectrometers, which can provide spectral quality close to Fourier transform limit, i.e. about 300 MHz bandwidth at 243 nm with some nanosecond pulse duration. Pulse energies of about 1 mJ are sufficient for two-photon optogalvanic spectroscopy; hence the irradiance in the excitation region of the laser beams (diameter of about 200 μm) does not exceed...
1 GW/cm². These conditions lead, in a first step, to the two-photon excitation; the absorption of a third laser photon results in subsequent photo ionization. Hence, the charged particles created in the overlap volume of the laser beams are accelerated by the E-field and the corresponding variation of the discharge impedance can be measured (OG signal). This technique provides a reliable determination of the local E-field via the frequency shift of the (2P 1/2) and (2P 3/2) Stark components, while the integral of the E-field versus distance from the cathode surface gives the cathode fall voltage drop. We are going to compare the cathode fall behavior of a HCD operated with hydrogen and deuterium for various discharge conditions: currents from 50 to 200 mA and pressures from 270 to 1350 Pa.

2. Experimental set-up, measurements and E-field determination

The experimental set-up is shown in figure 1. Measurements are performed in the low density plasma generated in a home-made HCD of cylindrical symmetry. The cathode is placed in between two hollow anodes to allow end-on excitation. The discharge operates at a constant gas flow (corresponding to the discharge pressure) entering at one anode side and leaving at the opposite. This device provides stationary stable non-LTE plasma, with low density and low kinetic temperature of atoms and molecules. Experimental conditions in this study result in a very quite discharge regime without any tendency to anode or cathode sparking.

![Schematic view of the experimental set-up](image)

A comprehensive explanation of the whole experimental arrangement is given elsewhere [11, 12]. The laser system consists of a 10 Hz injection-seeded Q-switched Nd:YAG laser (Continuum, Powerlite) and a modified OPO-OPA system (Continuum, Mirage 500). This laser offers tunable radiation in the visible and near-IR range (426 nm to 2.12 μm). The required 243 nm radiation is obtained by sum-frequency generation (SFG) of 772 nm and the third harmonic of the Nd:YAG laser in a BBO crystal. This concept provides up to 10 mJ pulse energy in 2.5 ns and 300 MHz bandwidth [13]. The frequency detuning of the laser, i.e. the OPO signal-wave (772 nm), is controlled by a monitor etalon with free spectral range (FSR) of 7 GHz and a photodiode (PD). From this, we can derive that the regularity of the frequency detuning is typically about 1 % of the FSR of the etalon during a complete scan. An accurate frequency reference (ν₀) of the 1S – 2S transition for zero E-field is provided by the optogalvanic reference cell (in figure 1), which contains atomic deuterium or hydrogen, generated by thermal dissociation at a pressure of about 500 Pa.

A moderate amount of the laser energy is introduced in the experiment (figure 1). The 243 nm laser radiation is divided in two counter propagating circularly polarized laser beams providing Doppler-free two photon excitation for the 1S - 2S transition, according to the selection rules for this transition ΔL = 0. Each beam is focused with a 1 m lens into the plasma. In order to achieve the best spatial...
resolution, both beams cross in a horizontal plane parallel to the HCD axis, and the measurements are taken in the upper central part of the cathode fall region. The spatial resolution in the experiment is given by the overlap of the two laser beams. The overlap volume is about 30 mm long and has a focus diameter of 200 μm. Vertical displacements of the HCD permit performing measurements at various distances from the cathode surface. Taking into account the beam divergence (4:1000) and the focus diameter, the closest proximity to the cathode wall we can reach without irradiating the cathode surface is 150 μm.

A detailed description of the Stark effect in the 1S - 2S transition of atomic hydrogen isotopes can be found in [11] and theoretical calculations in [14]. The measured spectra of the 1S - 2S transition show clearly the theoretical evolution of the three underlying Stark components as a function of the electric field. The red shifted component (2P 1/2) starts growing and shifting in the wing of the central component (2S 1/2), while the blue one (2P 3/2) starts growing and shifting more slowly far off the central component which decreases and undergoes a very weak blue shift.

The comparison of the calculated Stark shifts with the experimental ones gives the corresponding E-field. In order to obtain the frequency shift of the three Stark-components with respect to the zero E-field frequency $\nu_0$, the measured spectra were fitted to lorentzian profiles. In the case of deuterium we fitted to three lorentzians. For the Stark splitting of atomic hydrogen one also has to account for the hyperfine structure underlying each Stark component, this means six lorentzians for the fit.

3. Results

After having determined the local E-field for all the spectra measured at various distances from the cathode surface, we can plot the E-field variation in the cathode fall region for the corresponding discharge parameters. In figures 2 and 3 we show the E-field vs. radial distance from the cathode surface for a fixed current of 100 mA, and several pressures: 270, 400, 600, 900 and 1350 Pa in deuterium and hydrogen. For 1350 Pa pressure a linear decay of the E-field is observed and with decreasing pressure the maximum field strength decreases, the dependence is more parabolic and the cathode sheath thickness is expanding. Only for the lowest pressure of 270 Pa the tendency of the sheath thickness behaves differently and contracts again. As it can be seen in the figures similar behaviour is observed for deuterium and hydrogen, although the E-fields at the cathode surface are larger for deuterium.

![Deuterium E-field vs Radial Distance](image)

**Figure 2.** E-field vs. radial distance from the cathode surface for a fixed current of 100 mA and several pressures, in deuterium

![Hydrogen E-field vs Radial Distance](image)

**Figure 3.** E-field vs. radial distance from the cathode surface for a fixed current of 100 mA and several pressures, in hydrogen
Figures 4 and 5 show a complement to the former figures, i.e. for both isotopes the measured variation of the E-field in the cathode fall region versus the radial distance from the cathode surface, for a fixed pressure of 900 Pa, and several currents: 50 (or 60), 100, 150, and 200 mA. These figures confirm, once more, a similar behaviour observed in deuterium and hydrogen; this means as the current increases the maximum E-field increases while the cathode fall region becomes more compressed. Similar results are found for a pressure of 400 Pa and the same discharge currents. Absolute values for the E-field are bigger in deuterium in all cases. The E-field is always larger in deuterium although the thickness of the cathode fall region is quite similar compared to hydrogen.

References

[1] Gravilenko VP 2006 Instruments and Experimental Techniques, 49 149
[2] Muraoka K, Maeda M 2001 Laser-Aided Diagnostics of Plasmas and Gases, (Institute of Physics Series in Plasma Physics IOP)
[3] Barbeau C, Jolly J 1991 Appl. Phys. Lett. 58 237
[4] Booth JP, Fadlallah M, Derouard J, Sadeghi N 1994 Appl. Phys. Lett. 65 819
[5] Booth JP, Derouard J, Fadlallah M, Cabaret L, Pinard L 1996 Optics Communications 132 363
[6] Czarnetzki U, Luggenhölscher D, Döbele HF 1998 Phys. Rev. Lett. 81 4592
[7] Czarnetzki U, Luggenhölscher D, Döbele HF 1999 Plasma Sources Sci. Technol. 8 230
[8] Czarnetzki U, Luggenhölscher D, Döbele HF 2001 Appl. Phys. A 72 509
[9] Cherkasova EK, Gavrilenko VP, Zhuzhunashvili AI 2006 J. Phys.: Appl. Phys. 39 477
[10] Gemisic Adamov M, Steiger A, Grützmacher K, Seidel J 2007 Phys. Rev. A 75 013409
[11] de la Rosa MI, Pérez C, Grützmacher K, Gonzalo AB, Steiger A 2006 Plasma Sources Sci. Technol. 15 105
[12] de la Rosa MI, Pérez C, Grützmacher K, Fuentes LM 2009 Plasma Sources Sci. Technol. 18 015012
[13] Grützmacher K, de la Rosa MI, Gonzalo AB, Steiger M, Steiger A 2003 Appl. Phys. B 76 775
[14] Seidel J 2000 private communication (unpublished)