Diagnostics of Atmospheric Pressure Plasmas Utilizing Ultrafast Lasers

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Diagnostics play a key role in assessing our understanding of processes that occur in low-temperature plasmas by benchmarking predictive capabilities as well as through discovering otherwise unexpected behaviors. As the perceived landscape of low-temperature plasma science evolve and challenges become more complex (high densities, shorter lifetimes, more reaction pathways), a broad range of diagnostic capabilities are needed to provide a sufficiently complete picture of the plasma. Therefore, new methods need to be developed and made available to facilitate research efforts of the low-temperature plasma community. In this highlight, we described continued efforts to further the state-of-the-art in plasma diagnostics.

To further the development of the laser-collision induced fluorescence (LCIF) method [1] for use in such plasmas, a 640 Torr helium discharge, in a point-to-point configuration (Figure 1a) is being studied. A key and potentially transformative element of the ongoing effort is the utilization of a short pulse (~100 fs) laser to perform the initiation of the LCIF process (Figure 1a). For the data presented here, LCIF is observed for 10 ns, starting ~ 1 ns before laser excitation.

To demonstrate the ability of the LCIF method to interrogate spatial and temporal behavior of a plasma, the evolution of a 640 Torr helium afterglow plasma in response to a 250 ns high-voltage excitation event (Figure 1b) was studied. It was observed that two excitation fronts are present during the formation of the plasma channel and that behind these fronts resides regions of higher electron density. As the electron density builds, the velocity of the front launched from the cathode (lower electrode) accelerates due to increased localized electric field. The successful development of the LCIF method in atmospheric pressure plasma environments will be presented in an upcoming fast-track communication [2].

References
Continued development and implementation of plasma diagnostics for high pressure plasmas utilizing ultrafast, short-pulse lasers

- Diagnostic method: Laser-collision induced fluorescence (LCIF) capable of assessing electron densities, excited states and electric fields.
- Utilized method: Short-pulse laser excitation (< 100 fs).
- Application space: Quantify formation of nanosecond driven plasma discharge in highly collisional atmospheric pressure (640 Torr) environment.

LCIF scheme

- 640 Torr He filament
- Plasma generation measured with LCIF

DOE Plasma Science Center
Control of Plasma Kinetics
Time-Resolved Emission from the VUV to near IR of Atmospheric Pressure He Discharges into Open Air

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Time and space-resolved investigations of an rf-powered (200 kHz) He plasma jet, propagating through open air and impinging on dielectric substrates (quartz or MgF₂), have yielded insights into emission excitation mechanisms. VUV to near-IR emissions fall into two classes. Reactions that can produce excited electronic states with a pathway for releasing excess energy are primarily excited by collisions with He metastables (He*). Those states with no easy means of releasing this energy are excited by electron impact. For example, \( \text{N}_2(C^3\Pi_u \rightarrow B^3\Pi_g) \) emission is excited by electron-impact, since the 9.6 eV excess energy left after excitation of \( \text{N}_2 \) by collisions with He* cannot easily be released.

Evidence for e-impact excitation is found in time-resolved emission along the discharge axis. Optical emissions from \( \text{N}_2 \) and He at 706 nm are shown in Fig. 1a. For He, the only energetically possible excitation reaction is by electron impact. Hence He emission (dashed-dotted red line) peaks at peak electron density and energy at maximum (+) and (-) voltage and falls to zero near 0 V when there are no high-energy electrons. Time dependent \( \text{N}_2 \) emission is similar to He near the peak (+)V, when the plasma exits the tube and extends to the surface. No \( \text{N}_2 \) emission is observed at negative voltages, when the plasma is confined to the tube, since \( \text{N}_2 \) is not present in the He flowing through the tube.

Conversely, emission from \( \text{N}_2^+ \) is observed along the discharge axis for the entire period, and is only slightly smaller at 0 V as at peak (-)V. This observation can only be explained by excitation of \( \text{N}_2^+ \) by long-lived He* that is present throughout the rf cycle and reaches relatively high levels near the exit of the discharge tube. Penning ionization dominates because of the small exothermicity (1.06 eV) and facile partitioning of energy into the recoiling electron. Isolated \( \text{N}_2^+ \) emission near the surface is strongly modulated because the much higher \( \text{N}_2 \) and \( \text{O}_2 \) concentrations along the axis of He Laminar flow quench He*.

Lack of emission by \( \text{N}_2^+ \) near the surface at negative voltage rules out photoionization by He emission near 50 nm.

Figure 1 - Time-resolved emission along the axis of the discharge (red squares, 0°) and near the flat surface (<1 mm) of a prism (green squares, 40°). a) \( \text{N}_2(C^3\Pi_u \rightarrow B^3\Pi_g) \) at 337 nm and He at 706 nm (dashed-dotted lines). b) Time-resolved \( \text{N}_2^+ (B^2\Sigma_u^+ \rightarrow X^2\Sigma_g^+) \) emission at 391 nm.
MECHANISMS FOR OPTICAL EMISSION IN 1 ATM HE DISCHARGES INTO OPEN AIR

- VUV to near-IR emissions fall into two classes:
  1) Reactions that produce excited states with ways to release excess energy, primarily excited by collisions with He metastables (He*)
  2) Reactions that cannot release this energy are excited by e- impact.

- Case (1) leads to small modulation of emission. Examples include Penning ionization of N$_2$, and dissociation of H$_2$O to produce OH and H emission

- Case (2) leads to complete modulation during rf period. Examples include He and N$_2$ (C$\rightarrow$B) emissions.

![Graphs of OH and H emissions with voltage and time axes]