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A. Brockhaus, R. Sauerbier, J. Engemann. Comparison of three excitation schemes for cylindrical dielectric barrier discharges. *European Physical Journal: Applied Physics*, 2009, 47 (2), pp.1-6. 10.1051/epjap/2009083 . hal-00480175

HAL Id: hal-00480175

<https://hal.science/hal-00480175>

Submitted on 3 May 2010

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COMPARISON OF THREE EXCITATION SCHEMES FOR CYLINDRICAL DIELECTRIC BARRIER DISCHARGES

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Abstract. Three dielectric barrier discharge jets have been analyzed mainly by spectroscopic methods. Power densities, electron concentrations, and heavy particle temperatures are in the same range for the jets. All three jets are excited by sinusoidal voltages with kilohertz frequencies. High speed imaging proves that for quite different excitation schemes the jet plasma consists of isolated, fast moving, light-emitting structures. These so-called plasma bullets are emitted at a certain phase of the excitation voltage and occur in one half-cycle only.

PACS: 52.80.-s, 52.80.Hc, 51.50.+v

1. INTRODUCTION

Non-thermal atmospheric pressure plasmas (APPs) are attractive both to industry and to science. Companies seek for cost reduction by replacing traditional low-pressure plasma applications based on vacuum technology with inexpensive APPs. In the scientific world these plasmas – although being known for more than 100 years – still present considerable challenges concerning modelling and diagnostics. This study is restricted to dielectric barrier discharges (DBDs) which offer an experimentally simple way to limit the discharge current and to keep the gas temperature low. However, the self-interrupting nature of DBDs, combined with difficulties due to the high collision frequencies, severely complicates diagnostic investigations.

We have compared three different types of cylindrical DBDs which are all excited by sinusoidal voltages in the kilohertz range. All three plasma sources can produce plasma jets, i.e. light emitting plasma zones extending at least to some centimeters from the source. This feature is of course essential for all kind of non-thermal plasma processing applications.

From very early measurements of current-voltage characteristics it became obvious that the plasma jets are non-stationary – in contrast to their visual appearance. With the aid of a fast intensified CCD camera we could prove that the jet consists of so-called plasma bullets, i.e. small light emitting structures which are emitted from the DBD source at a certain phase of the sinusoidal excitation voltage [1]. The bullets move with velocities of several kilometers per second. It was not clear if this phenomenon was a speciality of our cylindrical plasma source #1 or was of general importance. Therefore a number of experiments was set up to improve the understanding of plasma formation, its properties and plasma transport.

2. EXPERIMENTAL SET-UP

The set-up for all three jets is displayed in Figure 1. DBD jet #1 is a very simple device. It just consists of a quartz glass tube of 15 cm length, inner diameter 4 mm, outer diameter 6 mm, with two ring electrodes (silver or aluminium foil, $L = 50$ mm in length) attached to the outside surface. The distance between the two electrodes is $D = 35$ mm. This discharge has been operated in helium always.

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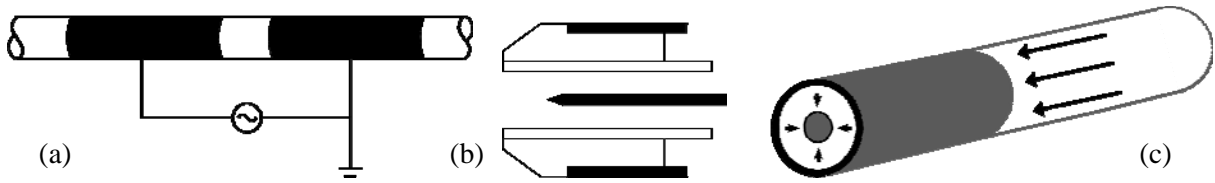


FIGURE 1. Set-up and working principles for all three plasma sources: (a) simple cylindrical DBD jet #1, (b) nozzle of jet #2 with internal needle electrode, (c) piezo-ceramic jet #3 converting thickness vibrational mode to longitudinal mode.

The simple design allows for an analytical solution of its electrical on-axis field. It is an exercise in Maxwell theory to prove that the electric field of a uniformly charged ring with charge q and radius R is given by

$$E_z(z) = \frac{q}{4\pi\epsilon_0} \frac{z}{(R^2 + z^2)^{3/2}} \quad (1)$$

One can integrate this solution to derive the axial field of a charged cylinder of length L :

$$E_{z,cyl}(z) = \frac{q}{4\pi\epsilon_0 L} \left\{ \left[R^2 + (z - L/2)^2 \right]^{-1/2} - \left[R^2 + (z + L/2)^2 \right]^{-1/2} \right\} \quad (2)$$

Finally, the on-axis field for our plasma jet is then obtained from a superposition of two cylindrical fields as

$$E_{z,jet}(z) = E_{z,cyl}(z - D/2 - L/2) - E_{z,cyl}(z + D/2 + L/2) \quad (3)$$

Here D denotes the distance between the two cylinders, i.e. the centers of the cylinders are located at $z_1 = D/2 + L/2$ and $z_2 = -D/2 - L/2$. The field is displayed in Figure 2 for our case $D = 35$ mm and $L = 50$ mm. The absolute field strength is maximum at the cylinder edges and is diminished between the cylinder electrodes. However, it is obvious that the field will be significantly modified by (i) surface charges on the dielectric and by (ii) free charges in the volume when the plasma has been ignited.

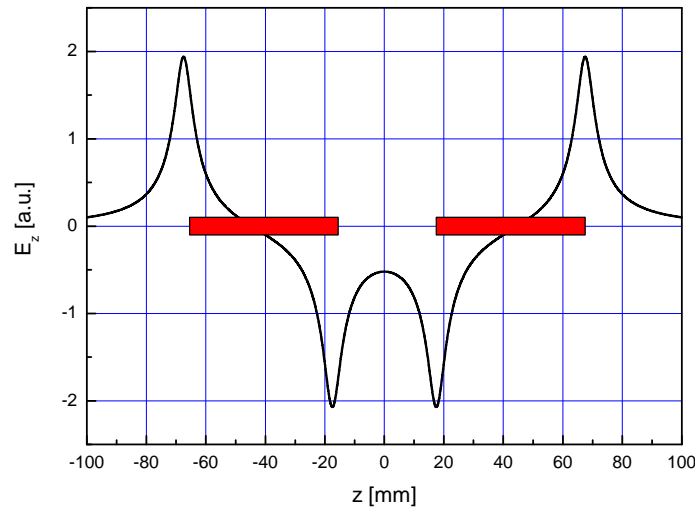


FIGURE 2. Axial electric field for the cylindrical DBD plasma source #1. The position of the electrodes is indicated by the two bars.

In source #2 one of the outer ring electrodes is replaced by a concentric metallic needle electrode within the tube. In contrast to source #1 this excitation scheme enables easy jet production in a variety of gases including air [2]. It could be thought of as a combination between a DBD and a corona-type discharge. It is striking that although the electric field configuration differs drastically from the one we discussed previously, jet #2 is also able to produce a kind of *bullet* phenomenon.

Finally, APP jet #3 is based on a piezo transformer principle. The scheme is displayed in Figure 1 (c). In jet #3 two piezoceramic tubes vibrating in different modes are mechanically coupled. The arrows in Figure 1 (c) indicate the polarization of the material. Details may be found elsewhere [3, 4]. By working at resonance frequencies around 100 kHz a low sinusoidal voltage applied to one of the piezo tubes is therefore able to generate a high electric field between the end points of the other ceramic which is sufficient for plasma ignition. This source does not require a high-voltage power supply and is electrically safe in the sense that there is no electrical shock if the ceramic is touched.

For all three sources the power density is in the range of some W/cm^3 and typical gas flows are 2-8 slm. We used mainly time-resolved imaging and optical emission spectroscopy to analyze the plasma and to compare the three sources.

3. RESULTS AND DISCUSSION

3.1. Electrical Measurements

With the aid of Rogowski coils we obtained time-resolved current measurements. For the “classical” DBD discharge #1 the current is dominated by displacement current with almost 90° phase difference between voltage and current. This is just the charging and discharging of the cylindrical capacitor with the electric field discussed in the previous section. From the voltage amplitude $V_0 = 6 \text{ kV}$ and the current amplitude $I_0 = 7 \text{ mA}$ at a frequency of 13 kHz we obtain a capacitive load of $C = 14 \text{ pF}$. The displacement current can be measured with and without a plasma. However, when the plasma is ignited, a small peak adds to the current waveform. As will be shown in the next section, this current peak corresponds to the so-called plasma bullet. The plasma is by no means stationary but exists only during a small fraction of the period. This part of the curve is displayed in Figure 3. From the “extra current” of around $750 \mu\text{A}$ lasting for approximately $2.8 \mu\text{s}$ we get a charge of $2 \times 10^{-9} \text{ As}$ which means around 1.3×10^{10} elementary charges.

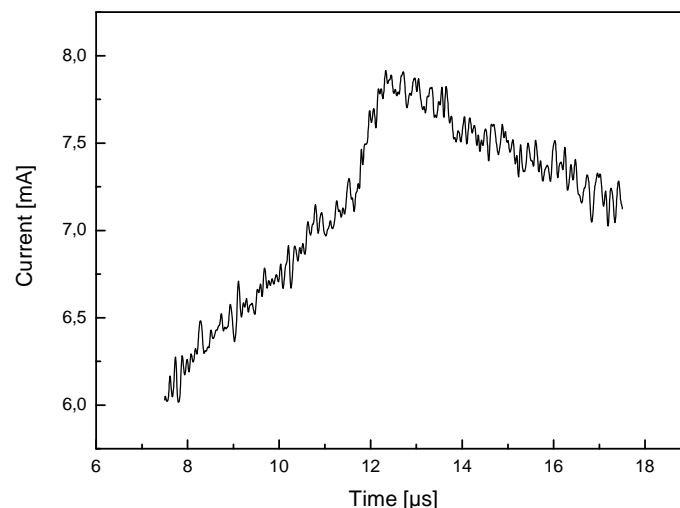


FIGURE 3. Part of the sinusoidal current curve. The peak after $12 \mu\text{s}$ corresponds to the observed plasma bullet.

3.2. Plasma Imaging

The visual appearance of all three jets gives the impression of a stationary plasma being “blown out” from the jet nozzle, see Figure 4. However, this is an illusion. With the aid of a high speed intensified CCD camera we could prove that the jet plasma consists of single, fast moving plasma bullets. The bullet velocity is in the range of several kilometers per second which is much faster than the gas flow velocity (some m/s). In fact the bullet phenomenon can be observed as well if the gas flow direction is reversed.

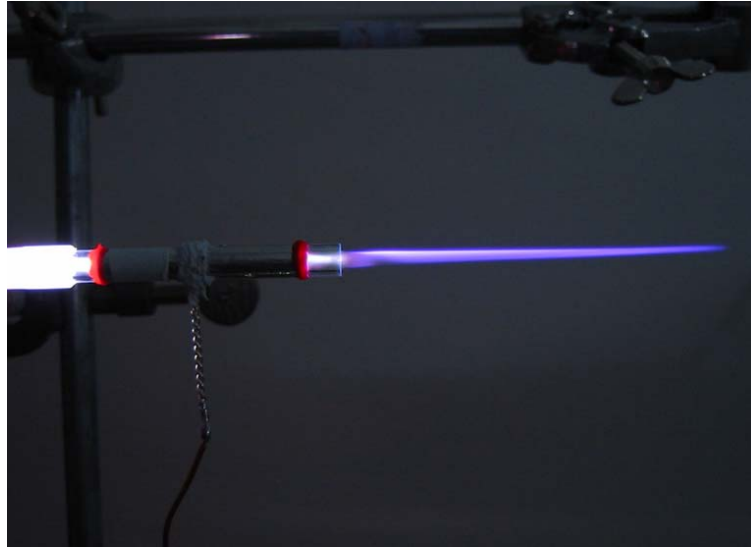


FIGURE 4. Photo of plasma jet #1 operating in pure helium. Plasma jet length downstream is ca. 10 cm.

Figure 5 displays two sequences of single CCD images obtained from jets #1 and #3. Exposure time is 50 ns with a delay of 100 ns between the photos. The bullet feature has first been reported from the simple version of jet #1 [1]. There is a striking similarity in the images although the piezo excitation of jet #3 differs considerably from the #1 DBD scheme.

Very recent measurements gave evidence that jet #2 may also produce plasma bullets. Since this device can also be operated in helium-free gases it can be used to settle the naturally arising question if the bullet phenomenon occurs in helium only. This is definitely not the case. However, the details are not very clear up to now. In helium the bullet looks quite similar to the images from jets #1 and #3.

In pure argon one sometimes observes a kind of instability. In the long-exposure photo a slight bending and twisting of the plasma jet is visible, see Figure 6. Even backward going parts of the plasma can be identified. If the same phenomenon is investigated with high temporal resolution, a very strange bifurcation can be seen: the clearly identifiable plasma head – the bullet – splits off into two separate parts which seem to diverge. It should be noted that the instability occurs apparently randomly and is a relatively rare phenomenon: only one instability can be observed about every 100 periods.

How can the bullet phenomenon in general be understood? It does not seem probable to assume that heavy particles i.e. neutrals or ions, move with km/s velocity since this would mean enormous heating at 1 bar. On the other hand the existence of excited light emitting species as well as electrical measurements prove that there is a charge transport associated with the bullets. One may therefore conclude that high energetic electrons are present causing both ionization and excitation in the helium gas.

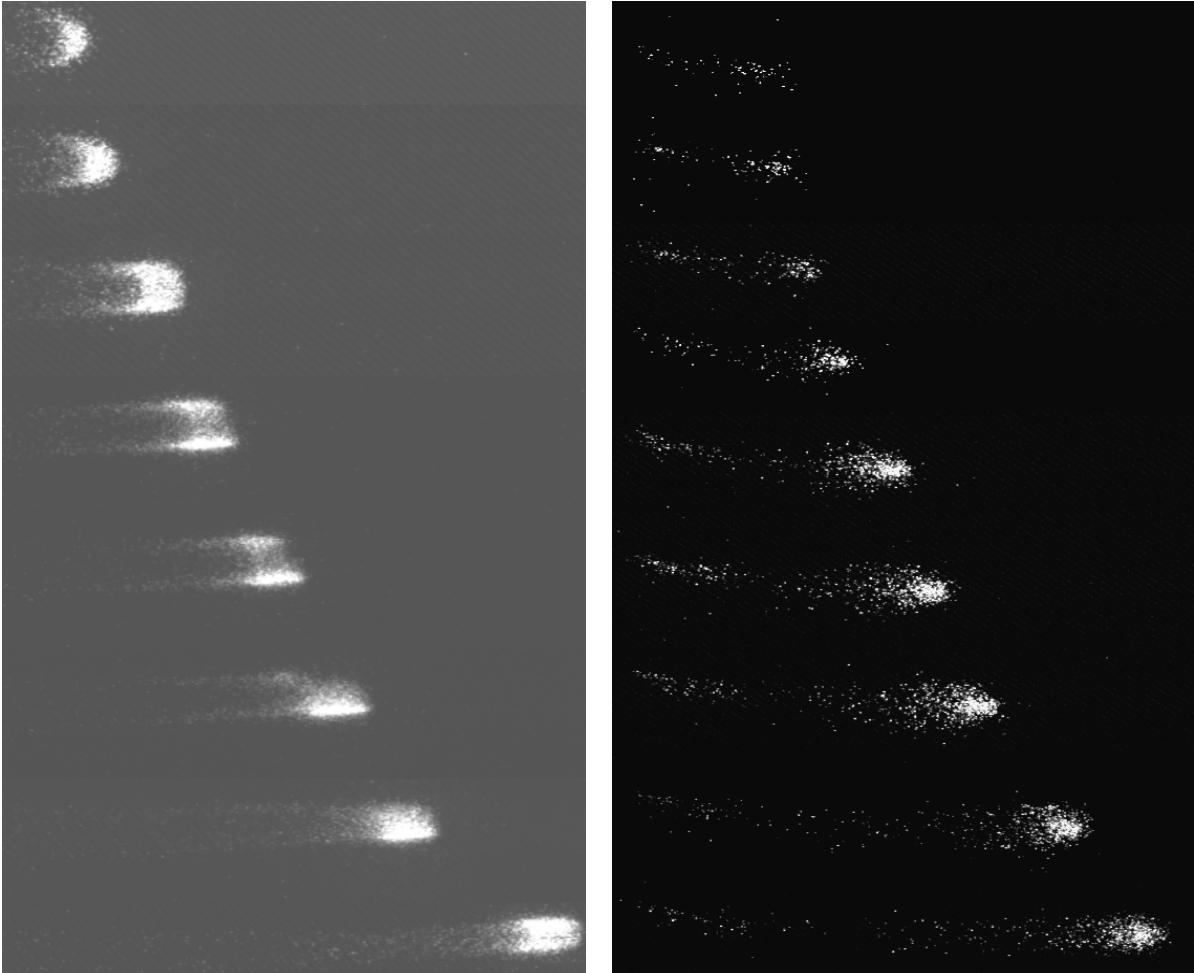


FIGURE 5. CCD images of plasma jet1 #1 (left) and #3 (right). Width of imaged area is 1.7 cm.

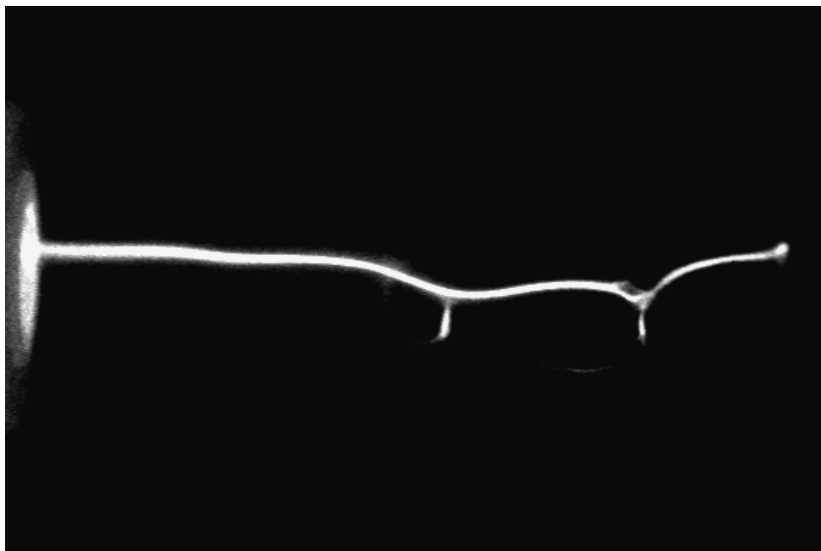


FIGURE 6. Time-integrated image (45 μ s, single shot) from jet #2 in pure argon.

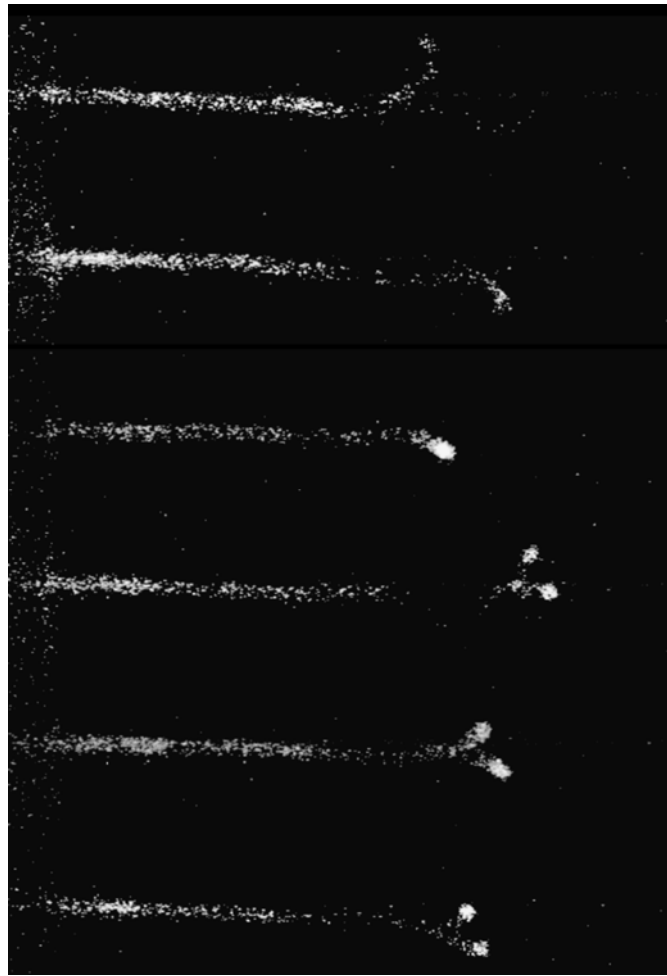


FIGURE 7. Bifurcation observed in jet #2 operating in pure argon. Exposure time is 30 ns for every image.

The bullets can be precisely triggered in the sense that there is a fixed phase relation between the sinusoidal excitation voltage and the bullet leaving the tube. It appears that the bullet starts from a surface charge cloud when the voltage sweeps from negative to increasing positive values. Therefore we conclude that the bullet is associated with a negative ionization front – essentially an anode-directed streamer which propagates with increasing velocity [5]. It is very likely that photoionization plays a significant role [6]. In this fast process highly energetic photons are produced at the streamer head.

One of the early streamer theories has been presented by Dawson and Winn [7] and has been applied to a discharge similar to our type #1 by Lu and Laroussi [8] and other authors [9]. This model requires a minimum number of charges of 3×10^9 , a condition which is clearly met by our discharges. The cylindrical geometry of our sources force the light emission to be predominantly axially. However, the physics are still not well understood. A modified fluid plasma model is currently under development to clarify the phenomenon.

3.3. Rotational Temperature

Plasma jet #2 can be operated in a large variety of gases including air or pure nitrogen. For the other sources helium is easiest to ignite although they allow for the addition of other gases in the percentage range. In principle it should be possible to use any gas but the excitation will then require much higher voltages. For example, recently we were able to generate a piezo-type plasma jet in pure argon but the plasma source design differs completely from the #3 geometry to achieve higher electric fields. These relative new developments will therefore be presented in a separate publication.

In any case it is difficult at atmospheric pressure conditions to avoid contamination e.g., by water from humid air. Downstream from the source outlet the plasma mixes with ambient air. It is therefore possible to observe typical nitrogen emission bands in all three sources.

We used the second positive system (C-B) of molecular nitrogen at 337 nm and the first negative system (B-X) of the molecular nitrogen ion N_2^+ at 391 nm to obtain rotational temperatures from Boltzmann plots.

Generally the results prove that all three jets are low-temperature discharges. Ion temperatures are slightly higher than the neutral molecule temperatures. Figure 8 displays the results for N_2^+ rotational temperatures at several observation points along the axis of jet #1. For these measurements a small slit was cut into the electrodes to get optical access to the plasma. Although plasma emission can be detected over the whole length of the dielectric tube it appears that the temperature is about 100 degrees higher at the powered electrode.

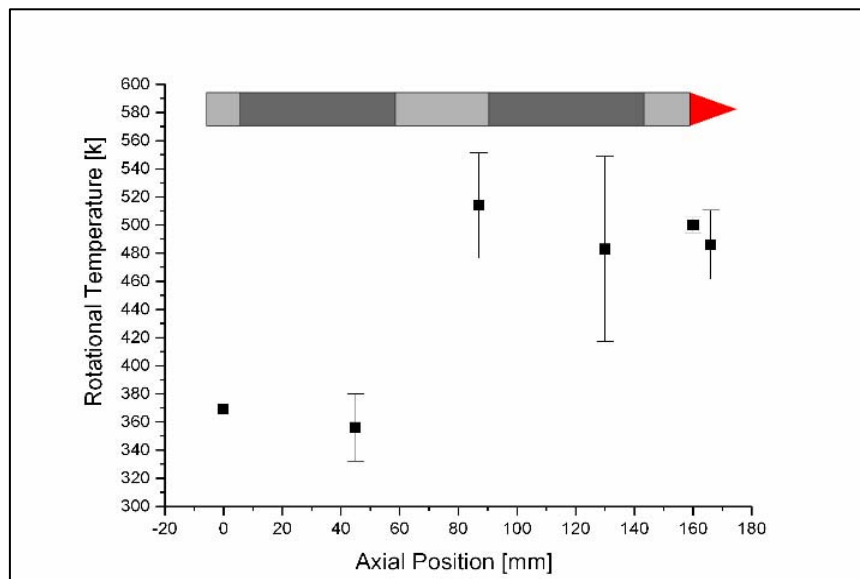


FIGURE 8. Rotational temperatures of N_2^+ at different plasma regions for jet #1.

3.4. Stark Broadening

As already mentioned at atmospheric pressure some contamination of the plasma gas with water is nearly unavoidable. This, however, has the advantage that several atomic hydrogen lines may usually be observed thus enabling diagnostic access. We analyzed the H_β line at 486 nm with a high resolution monochromator (1 m focal length with a 2400 lines/mm blazed grating). As an example a line shape, in this case from recording the optical emission of DBD jet #1, is displayed in Figure 9. The full line width at half maximum is typically between 0.05 and 0.08 nm, for all three jets. The best signal to noise ratio is obtained with jet #1. The piezo jet #3 gives the lowest emission intensities.

The main line broadening effects are i) van der Waals, ii) Stark, and iii) Doppler broadening, in this order, resulting in a Voigt line profile [10, 11]. However, only Stark broadening depends on the electron density. The other mechanisms depend on the gas temperature which is known from the rotational temperature measurements. Taking into account these data we obtain electron densities between 5×10^{12} and $2 \times 10^{13} \text{ cm}^{-3}$.

The results should be interpreted as gross estimations because van der Waals broadening is larger than Stark broadening by a factor between 2 and 10. Advanced techniques like Abel inversion to derive the radial density distribution are therefore not appropriate. The measurements result in averaged electron densities which are one order of magnitude higher than calculations based on the overall power balance and typical electron mobilities.

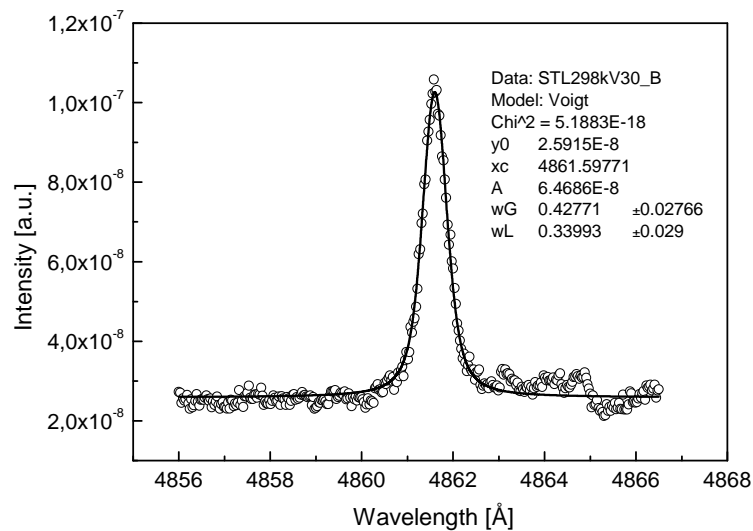


FIGURE 9. Hydrogen H β line from jet #1 with a Voigt profile fit (solid line).

4. CONCLUSIONS

Three different dielectric barrier discharge excitation schemes have been compared. All plasma sources exhibit jet behavior. Power densities, electron concentrations, and heavy particle temperatures are in the same range. However, only one jet (#2) is able to operate in gases without helium admixture. This is a clear advantage from an application-oriented point of view. It is due to a unique combination of DBD and corona-like excitation principles. On the other hand, this source requires a more sophisticated power supply since current reduction is mandatory for certain process gases to prevent a transition of the discharge to noisy brush- or spark-like modes.

The most striking result is that despite quite different excitation schemes the jet plasma consists of isolated, fast propagating plasma bullets. These structures have been observed in all three plasma sources. In contrast to early assumptions one can therefore not attribute the existence of plasma bullets to the specific electric field configuration of say, source #1. The bullet phenomenon is tentatively interpreted as an ionization front, i.e. a streamer-like feature with an important contribution from photoionization. In our view it is very likely that moving plasma bullets may also be present in other types of atmospheric pressure discharges.

ACKNOWLEDGEMENTS

Albrecht Brockhaus would like to thank the organizers of HAKONE XI for the enjoyable and fruitful meeting in Oleron Island, and U. Kogelschatz, G. Naidis, R. Brandenburg, S. Pancheshnyi, and many others for enlightening discussions during the conference.

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