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# On the velocity variation in atmospheric pressure plasma plumes driven by positive and negative pulses

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To better understand the variation in the “plasma bullet” velocity, the dynamics of an atmospheric pressure plasma plume driven by positive and negative pulses are investigated in detail. It is found that, before the plasma exits the nozzle, the plasma propagates at a speed of about 30 km/s for both positive and negative pulses. As soon as the plasma exits the nozzle, the plasma propagation speed increases dramatically for both cases. The peak velocity for the case of the positive pulse is much higher than that of the negative pulse, it is approximately 150 km/s and 70 km/s, respectively. According to the optical emission spectra, the acceleration behavior of the plasma bullet when it exits the nozzle is due to the increase in the  $N_2^+$  concentration. © 2010 American Institute of Physics. [doi:10.1063/1.3511448]

## I. INTRODUCTION

Nonequilibrium atmospheric pressure plasmas have recently been the subject of much attention due to their use in several novel applications, such as biological and chemical decontamination of media,<sup>1–26</sup> synthesis of nanomaterials,<sup>27,28</sup> as well as absorption and reflection of electromagnetic radiation.<sup>29,30</sup> Atmospheric pressure plasmas (excepting corona discharges) are traditionally generated between two electrodes. The discharge gap between the two electrodes usually ranges between a few millimeters to several centimeters, which substantially limits the size of the materials that may be treated directly. Fortunately, numerous types of cold atmospheric pressure plasma jet devices have recently been developed<sup>31–45</sup> which are able to generate cold plasmas in the surrounding air rather than in confined gaps. These new atmospheric pressure plasma jet devices present significant advantages over traditional discharges.

To develop suitable plasma jet devices for various applications, it is necessary to fully understand the mechanism of the atmospheric pressure plasma plume. Studies on the dynamics of the plasma plumes show that they are not a continuous volume of plasma, rather they are more like a bullet that travels from the exit aperture and terminates somewhere in the surrounding air. The speed of the “plasma bullet” varies from  $\sim 10^4$  to  $\sim 10^5$  m/s, which is several orders of magnitude higher than the gas velocities. Laroussi *et al.*, Lu *et al.*, Sands *et al.*, and Ye *et al.*<sup>31,36,46–53</sup> all found that the plasma bullet is electrically driven, which is quite similar to positive streamer discharges. Although closely analogous to typical positive streamers, the plasma bullets maintain unique features in the plasma initiation and formation.

Since the plasma bullet is electrically driven, it should behave differently when the plasma jet is driven by a negative pulse instead of a positive pulse. In this paper, to better understand the plasma bullet behavior, a high speed intensified charge coupled device (ICCD) camera with exposure

times of several nanoseconds is used to capture the temporal emission behavior of the plasma plume for both positive and negative pulses. It is found that the plasma bullets for both positive and negative pulses accelerate as soon as they enter into the surrounding air. To establish the mechanism of the acceleration behavior, optical emission spectra of the plasma plumes are captured before and after the plasma plumes enter into the surrounding air. It is found that the acceleration behavior of the plasma plumes can be explained by the photoionization based model proposed by Lu and Laroussi.<sup>36</sup>

The rest of the paper is organized as follows. The experimental setup is described in Sec. II. Details of the experimental results, including the current-voltage waveforms, high speed photograph of the plasma plume, and spatial resolved optical emission of the plasma plume for both positive and negative pulses are presented in Sec. III. Finally, Sec. IV provides a brief summary of this work.

## II. EXPERIMENTAL SETUP

Figure 1(a) is the schematic of the device. The high voltage (HV) wire electrode, which consists of a copper wire with a diameter of 2 mm, is inserted into a 4 cm long quartz

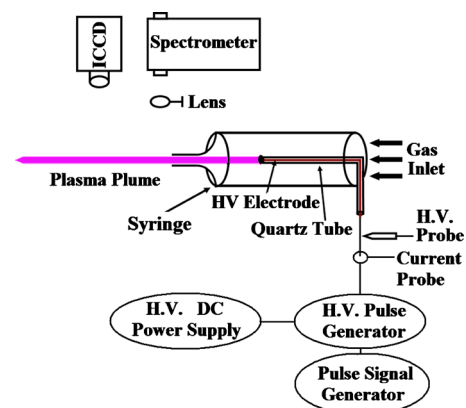


FIG. 1. (Color online) Schematic of the experimental setup.

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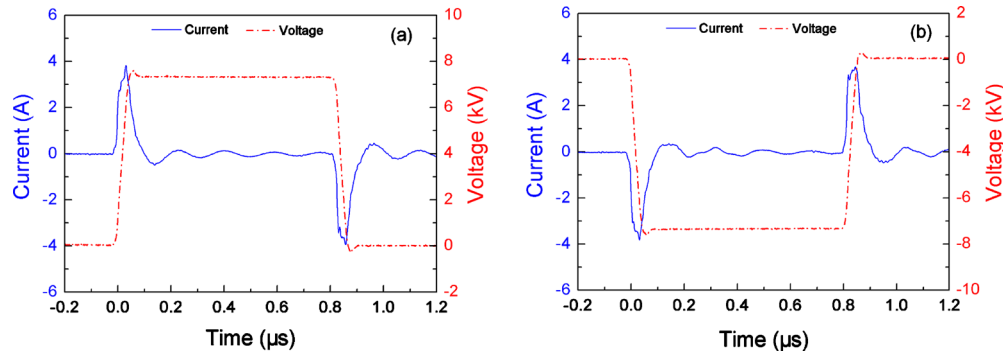


FIG. 2. (Color online) Current-voltage characteristics of the discharge for (a) positive and (b) negative pulses.

tube with one end closed. The inner and outer diameters of the quartz tube are 2 mm and 4 mm, respectively. The quartz tube along with the HV electrode is inserted into a hollow barrel of a syringe. The diameter of the hollow barrel is about 6 mm and the diameter of the syringe nozzle is approximately 1.2 mm. The distance between the tip of the HV electrode and the nozzle is 1 cm. When helium with a flow rate of 2 l/min is injected into the hollow barrel and the HV pulsed dc voltage (amplitudes up to  $\pm 10$  kV, repetition rate up to 10 kHz, and pulse width variable from 200 ns to dc) is applied to the HV electrodes, a homogeneous plasma is generated in front of the end of the quartz tube, along the nozzle, and in the surrounding air.

The applied voltages and the currents are measured by a P6015 Tektronix HV probe and a TCP202 Tektronix current probe, respectively. A fast ICCD camera (Princeton Instruments, Model: PIMAX2, exposure time down to 0.5 ns) is used to capture the dynamics of the discharge. The exposure time is set to 5 ns for all the photographs shown in this paper. A spectrometer (Princeton Instruments Acton SpectraHub 2500i) is used to measure the emission spectra of the plasma. The entrance and the exit slits of the spectrometer are fixed at 100  $\mu\text{m}$  for the experiment reported in this paper. A grating of 1200 grooves/mm is used for all the spectrum measurements.

### III. EXPERIMENTAL RESULTS AND DISCUSSIONS

Throughout this work, for both positive and negative pulses, the pulse widths, and pulse frequency are fixed at 800 ns and 8 kHz, respectively. The applied voltages are fixed at 8 kV and  $-8$  kV for the positive and the negative pulses, respectively. The helium flow rate was maintained at 2 l/min. The current-voltage characteristics of the discharge for the positive and the negative pulse are shown in Figs. 2(a) and 2(b), respectively. It should be pointed out that the current  $I$

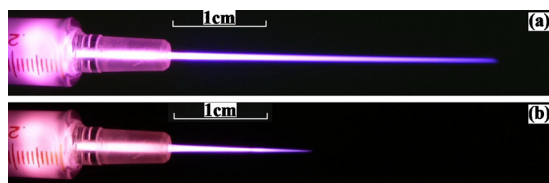


FIG. 3. (Color online) Photographs of the plasma for (a) positive and (b) negative pulses.

is the total current. It is the sum of the displacement current and the actual discharge current, where the actual discharge current is much smaller than the displacement current.<sup>39</sup>

Figures 3(a) and 3(b) are the photographs of the plasma plume generated by the positive and negative pulses, respectively. It can be seen that the plasma plume generated by the positive pulse is much longer than the plasma plume generated by the negative pulse. To further understand this phenomenon, a high speed ICCD camera is used to capture the dynamics of the plasma plumes. Figures 4 and 5 show the high speed photographs of the discharges taken at different delay times for the positive and the negative pulses, respectively. The time labeled on each photograph corresponds to the times in Fig. 2. As shown in Figs. 4 and 5, the plasma travels like a bullet before it exits the nozzle. For the case of the positive pulse, the plasma becomes much brighter as soon as it exits the nozzle. On the other hand, for the case of the negative pulse, the plasma also becomes brighter when it exits the nozzle but it is not as obvious as in the case of the positive pulse. It is interesting to note that the shape of plasma head for the case of the positive pulse is different to that of the negative pulse. It has a spherical shaped head for the case of the positive pulse, whereas it is more like the shape of a sword head for the case of the negative pulse. The reason behind the shape difference is not clear at the moment.

In order to have a more detailed discussion about the

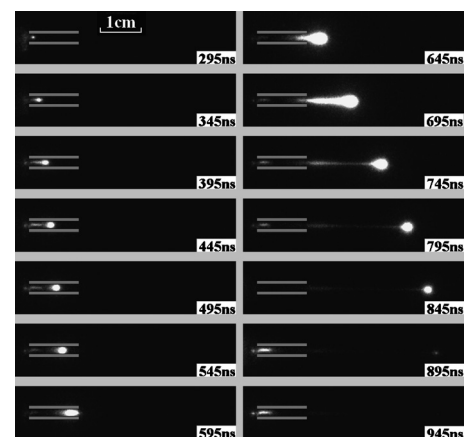


FIG. 4. High-speed photograph of the plasma plume for the positive pulse. The exposure time is fixed at 5 ns. The time labeled on each photograph corresponds to the time in Fig. 2(a).

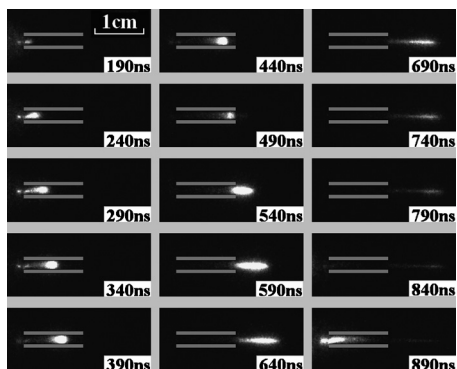


FIG. 5. High-speed photograph of the plasma plume for the negative pulse. The exposure time is fixed at 5 ns. The time labeled on each photograph corresponds to the time in Fig. 2(b).

plume dynamics, the plume velocity versus time is plotted in Fig. 6 and 7 for the cases of the positive and the negative pulses, respectively. As shown clearly in Figs. 6 and 7, the plasma bullets travel at a constant speed of about 30 km/s before they exit the nozzle for both cases. The plasma bullets accelerate as soon as they exit the nozzle for both cases too. But the peak velocity for the case of the positive pulse is much higher than that of the negative pulse and is about 150 km/s and 70 km/s, respectively.

Next, the optical emission spectrum is used to better understand the phenomenon mentioned above. For the cases of both positive and negative pulses, the optical emission spectra of the plasma bullets inside and outside the nozzle are measured. When the positive pulse is used, Figs. 8(a) and 8(c) show that the emission intensity of  $N_2^+$  is increased significantly (by more than one order of magnitude) when the plasma exits the nozzle. Thus, it is reasonable to assume that the  $N_2^+$  charge density increases when the plasma exits the nozzle. On the other hand, according to the model proposed by Lu and Laroussi,<sup>36</sup> the higher the  $N_2^+$  charge density, the stronger the local electric field induced by the plasma plume head, and the higher the plasma plume propagation velocity. This may explain why the propagation velocity of the plasma bullet increases when the plasma exits the nozzle. As to why the  $N_2^+$  charge density increases when the plasma exits the nozzle, it can be explained as follows. Before the plasma exits the nozzle, the concentration of  $N_2$  is very low inside the nozzle and most of the  $N_2$  is ionized. When the plasma propagates out of the nozzle, due to diffusion, the concentra-

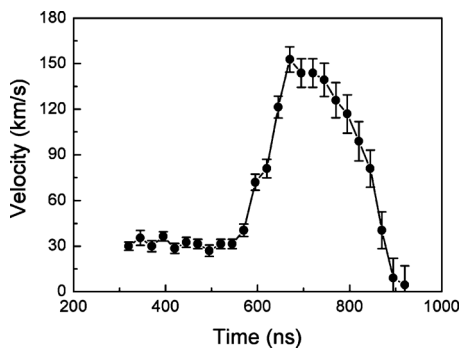


FIG. 6. Velocity of the plasma bullet vs time for the case of the positive pulse.

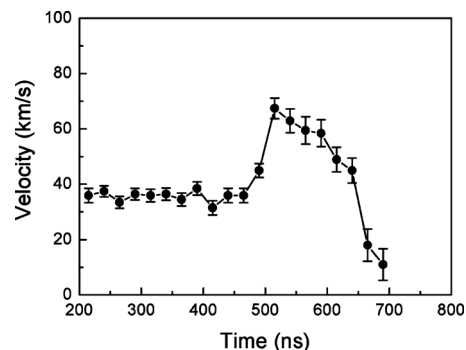


FIG. 7. Velocity of the plasma bullet vs time for the case of the negative pulse.

tion of  $N_2$  in the plasma plume increases significantly. Due to the high collision frequency, the metastable state He not only excites  $N_2$  but also ionizes  $N_2$  and then further excites it. As a result, the concentration of  $N_2^+$  increases significantly. This assumption is consistent with our following experimental observation. As can be seen from Fig. 8(a), before the plasma exits the nozzle, the emission intensities of both  $N_2$  and  $N_2^+$  are very low. The emission of  $N_2$  can only just be detected. Thus it is reasonable to say that most of the  $N_2$  is ionized. When the plasma exits the nozzle, Fig. 8(c) shows that the emission intensities of both the  $N_2$  and  $N_2^+$  increase dramatically. Therefore it is reasonable to say that the concentration of  $N_2^+$  increases when the plasma exits the nozzle.

When the negative pulse is applied, a similar behavior is observed as shown in Fig. 9. However, for the negative pulse, the peak velocity achieved is much lower than that for the positive pulse as mentioned above. This is similar to the streamer discharge. It can also be explained by the model proposed by Lu and Laroussi.<sup>36</sup> According to the model, the head of the plasma plume is like a plasma bullet containing  $N_2^+$ . Moreover, Lu *et al.*<sup>46</sup> also found that the electric field due to the external applied voltage plays an important role in the propagation of the plasma bullet. Therefore, when the positive pulse is used, the local electric field induced by the plasma bullet and the electric field from the external applied voltage have the same direction, so the total electric field is enhanced, which results in the high peak velocity of the plasma bullet. On the other hand, when the negative pulse is used, the two electric fields mentioned above have opposite directions, so the total electric field is weakened—which is the reason why the peak velocity achieved by the positive pulse is higher than that resulting from the negative pulse.

#### IV. CONCLUSION

To conclude, when the plasma exits the nozzle, the plasma experiences an acceleration process for both positive and negative pulses. This is explained as follows, due to diffusion, the concentration of  $N_2$  in the plasma plume increases significantly when the plasma propagates out the nozzle. The high concentration metastable state He can ionize  $N_2$  and further excite it, which results in the increase in the  $N_2^+$  concentration. This is confirmed by the optical emission spectra measurements.

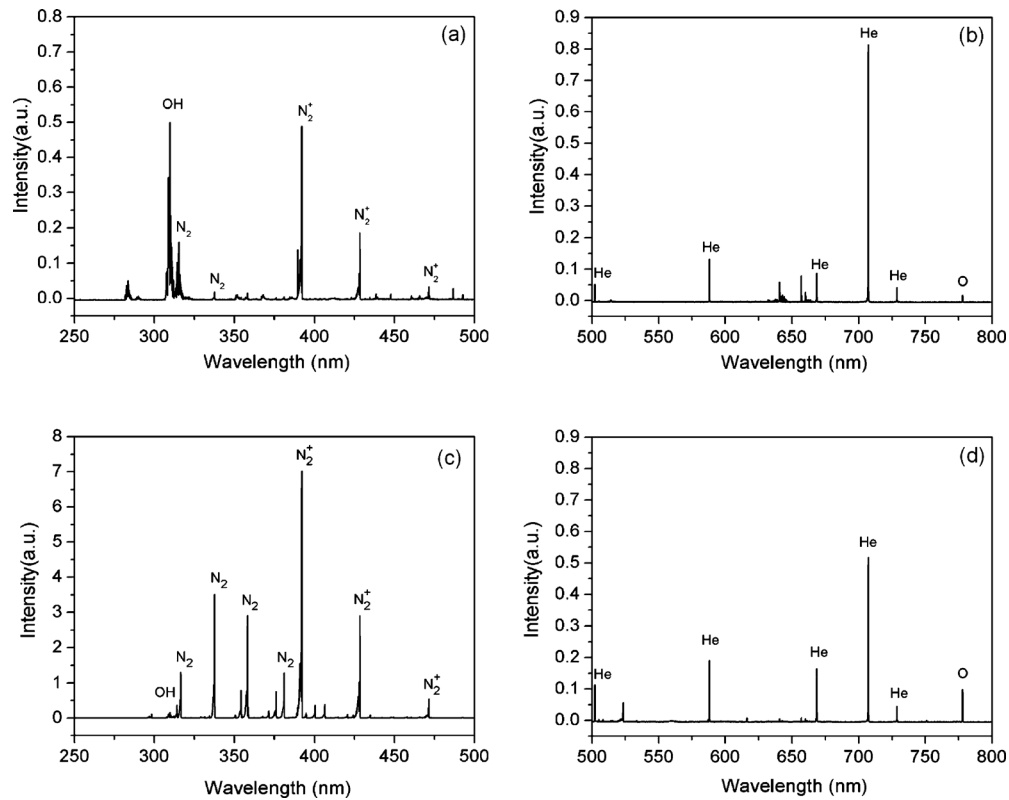


FIG. 8. Emission spectra of the plasma for the positive pulse. (a) and (b) are the emission of the plasma inside the nozzle, (c) and (d) are the emission of the plasma outside the nozzle (5 mm away from the nozzle).

Moreover, it is found that the peak velocity achieved for the positive pulse is much higher than that achieved for the negative pulse. This is because the plasma bullet propagates under two electric fields, i.e., the local electric field induced

by the plasma head that contains high concentration of N<sub>2</sub><sup>+</sup> and the electric field from the external applied voltage. When the positive pulse is used, the magnitude of the total electric field is the sum of magnitudes of the two electric fields, thus

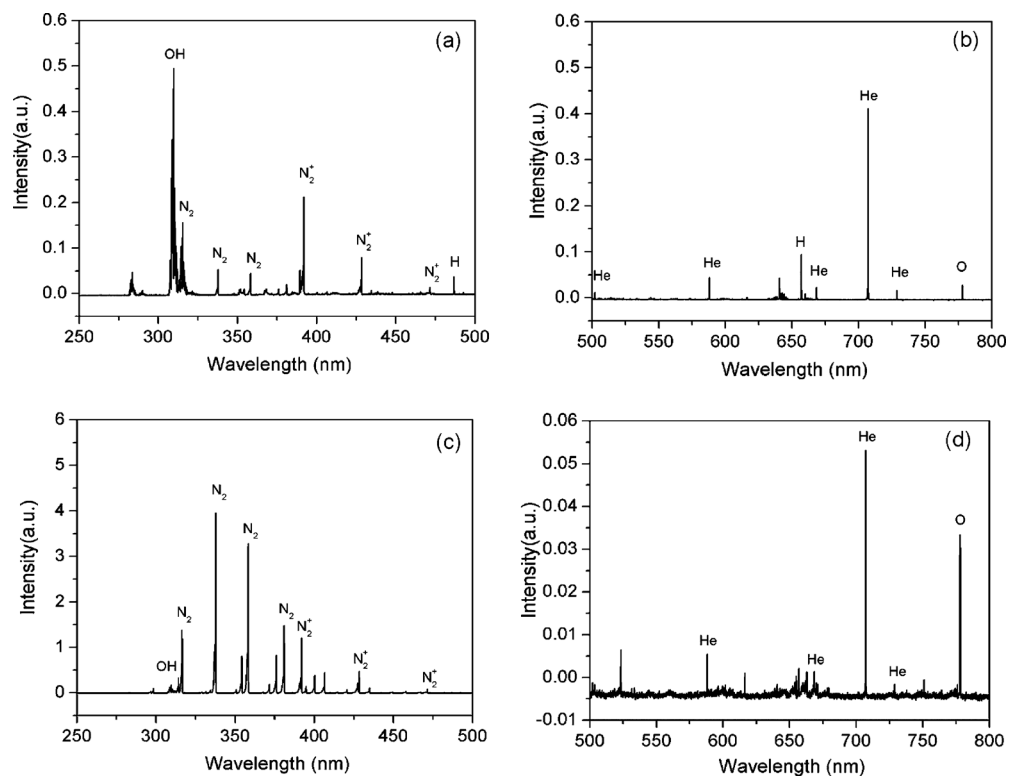


FIG. 9. Emission spectra of the plasma for the negative pulse. (a) and (b) are the emission of the plasma inside the nozzle, (c) and (d) are the emission of the plasma outside the nozzle (5 mm away from the nozzle).

the plasma velocity is higher. On the other hand, when the negative pulse is used, the magnitude of the total electric field is equal to the magnitude of electric field of the external applied voltage subtracted from the magnitude of local electric field induced by the plasma head. Therefore, the total electric field is weakened and the plasma velocity is lower.

## ACKNOWLEDGMENTS

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