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Formation of spatially periodic fronts of high-energy electrons in a radio-frequency driven surface microdischarge

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The generation of spatially periodic fronts of high-energy electrons (>13.48 eV) has been investigated in a radio-frequency surface microdischarge in atmospheric-pressure argon. Optical emission spectroscopy is used to study the Ar I $2p_1 - 1s_2$ transition surrounding a filamentary microdischarge, both spatially and with respect to the phase of the applied voltage. The formation of excitation fronts, which remain at a constant propagation distance throughout the RF cycle and for the duration of the pulse, may be explained by a localized increase in the electric field at the tip of surface-charge layers that are deposited during the extension phase. © 2013 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4789371>]

Non-thermal microdischarges offer a means to generate low-temperature (about 300 K) ionized gases at atmospheric pressure.¹ Their non-equilibrium populations of ions and electrons facilitate the generation of reactive plasma chemistry with relatively small power requirements. These useful properties have resulted in their recent application to biomedicine.²

In asymmetric surface barrier discharges (ASBDs), the reactive species propagate over the surface of a dielectric material. A low-profile design enables the discharge to be readily accessible to the target, while the dielectric barrier between the electrodes mitigates arcing.³ Sample access is a significant challenge in atmospheric-pressure plasmas and can be solved by employing a plasma jet.⁴ However, this is only suitable for applications requiring targeted treatment and ASBDs have the advantage of not requiring a continuous gas feed. Therefore, they may be particularly suited to large-area applications such as sterilization,⁵ aerodynamic flow control,⁶ and the modification of material surfaces.⁷

Over the past decade, the majority of ASBD research has been undertaken in the dielectric-barrier-discharge frequency band (DBD: up to 500 kHz (Ref. 8)). The elevated rate of electron oscillation in RF discharges (several MHz) enables distinctive discharge behaviour including enhanced repeatability due to a strong memory effect and a lower breakdown voltage.⁹ These useful properties are coupled with elevated optical-emission intensity,¹⁰ which being a function of the electron energy distribution function and the plasma density, is a potentially significant advantage.

The deposition of plasma species, e.g., ions and electrons, on the dielectric as surface charge is critical to the performance of ASBDs.¹¹ In this study, the propagation of relatively high-energy electrons, i.e., with an energy significantly greater than the bulk at 1–2 eV (Ref. 12) and hence play a key role in excitation and ionization processes, is investigated for a 13.56 MHz ASBD with particular interest in the formation of surface-charge structures.

The reactor consists of two identical 70 μ m thick, 10 mm wide, and 30 mm long copper tape electrodes with rounded ends. These are horizontally offset and affixed to either side of a 150 μ m layer of Kapton tape as shown in Figure 1 (electrode length is measured into the page). The grounded electrode is encapsulated in Kapton so that ionization only occurs between the powered electrode and the dielectric. The propagation distance of the discharge is measured along the longitudinal axis of one microdischarge and this is perpendicular to the edge of the powered electrode as shown by the downwards arrow in Figure 1.

A vacuum chamber houses the reactor in an environment of atmospheric-pressure argon. This is achieved by evacuating the chamber with a rotary pump and then introducing argon gas until atmospheric pressure is reached. RF power is coupled to the exposed, powered electrode using a broadband amplifier and the power level is regulated using a

FIG. 1. Schematic of the reactor showing the RF power coupling system, direction in which the propagation distance is measured and the orientation of the camera with respect to the discharge.

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2-channel arbitrary waveform generator to ensure that the RF pulses are phase locked with the camera trigger.

The discharge is pulsed to minimize dielectric heating and a mode comprising 300 μm long filaments is generated using pulses with a duration of 15 RF cycles (1.1 μs) and a period of 30 μs .¹³ The region-of-interest for imaging is the area surrounding one microdischarge, for which optical emission is visible up to 6 mm past the maximum propagation distance of the filament.

For optimum optical access, the reactor is installed directly in front of a window in the vacuum chamber and positioned such that the plane of the dielectric layer, which is the surface over which the discharge propagates, is on the same horizontal plane as the chamber window and camera aperture. An intensified charge-coupled device camera (ICCD: 1024 $\times$ 1024 13 μm^2 pixels) is used to detect the optical emission from the discharge. A band-pass filter is fitted to the camera aperture and has a central wavelength of 750 nm and a full-half-width-maximum (FWHM) of 10 nm to facilitate the study of the Ar I $2p_1 - 1s_2$ transition at 750.4 nm.

The ICCD is triggered to acquire an image of the discharge once per pulse of RF power. The acquisition interval is constant at 4 ns and this corresponds to 18.5 images per RF cycle at 13.56 MHz (74 ns). The optical emission from each time-step is measured and then accumulated over thousands of pulses to achieve a sufficient signal-to-noise level.

The voltage measured at the powered electrode, which is defined here as the applied voltage, is obtained using a high-voltage probe. To accurately reconcile the phase of the applied voltage at the powered electrode with the optical emission detected by the ICCD, the signal delays introduced by the apparatus are measured.

The applied voltage and corresponding optical emission from the discharge are shown in Figure 2. The voltage envelope (a) increases steadily to approach 550 V by the end of the

pulse. The intensity of the optical emission (b), as measured along a line-of-sight in the direction perpendicular to the edge of the powered electrode (see Figure 1) illustrates a very high degree of repeatability, i.e., the discharge forms in exactly the same location for each successive cycle of the applied voltage.

Clearly evident in Figure 2(b) is the separation of the discharge into two distinct bands as it propagates out from the edge of the powered electrode. This may be due to the presence of a population of positive space charge (argon ions) that is created by strong ionization at the discharge tip during each RF cycle. Due to the approximately linear increase in the applied voltage throughout the pulse, the propagation distance of the discharge also increases at a constant rate¹⁴ and this may result in the separation of the discharge on either side of the space-charge region.

Horizontal bands of increased optical-emission intensity are also observed in Figure 2(b) and occur in approximately 700 μm intervals at the maximum extent of propagation of the discharge during each RF cycle. These “fronts” are residual over the course of the pulse and remain at a constant propagation distance despite the time-varying amplitude of the applied voltage within each RF cycle. The formation of these spatially periodic structures is studied by observing the behaviour of relatively high-energy electrons, which play a key role in excitation and ionization processes. For a weakly ionized discharge at atmospheric pressure, the $2p_1$ energy level is mostly populated by electron-impact excitation from the ground state.¹⁵ By monitoring the Ar I $2p_1 - 1s_2$ transition at 750.4 nm, which requires electrons with an energy greater than 13.48 eV, the behaviour of energetic electrons can be tracked by calculating the variation in the electron-impact excitation from the electronic ground state as a function of time $E_i(t)$ using the method of Gans *et al.*¹⁶

$$E_i(t) = \frac{1}{n_0 A_{ik}} \left(\frac{d\dot{n}_{\text{ph},i}(t)}{dt} + A_i \dot{n}_{\text{ph},i}(t) \right), \quad (1)$$

where n_0 is the ground state density, A_{ik} is the transition probability from state i to state k , $\dot{n}_{\text{ph},i}(t)$ is the number of photons detected per unit volume per time, and A_i is the effective decay rate. Since the FWHM of the filter is 10 nm, the $2p_5$ emission at 751.5 nm is also collected. However, this diagnostic configuration has previously been found to be sufficient for the observation of strong trends in the $2p_1$ emission.¹⁷

$E_i(t)$ was calculated for one RF cycle, as defined by the arrows in Figure 2(b), and is shown in Figure 3. The corresponding measurements of the applied voltage (arbitrary units of magnitude) are included at the bottom to give an indication of the phase and three distinct intervals (each 4 ns) of excitation activity are identified A-C.

In the first quarter of the RF cycle, the rate-of-change of the applied voltage is at a maximum and the discharge propagates out from the powered electrode as has been observed previously in RF and DBD surface discharges at atmospheric pressure.^{18,19} However, as the voltage approaches its maximum value in the cycle (1168.5 ns), which is here defined as the extension phase (A), the maximum excitation is observed in a separated front region (see Figure 2).

The front in (A) is observed to propagate significantly slower at 4×10^4 m/s than the tail at 3×10^5 m/s. Although

FIG. 2. Temporal variation of the (a) applied voltage and (b) optical emission detected through the 750 nm filter with respect to time for pulses of 15 RF oscillations (1.1 μs) with a 30 μs period. In (b), the edge of the powered electrode is located at a propagation distance of 0 mm and the arrows indicate the interval over which the electron-impact excitation is shown in Figure 3.

FIG. 3. Variation in the electron-impact excitation from the electronic ground state $E_i(t)$ over the course of one RF cycle. The interval corresponds to the arrows in Figure 2. The edge of the powered electrode is shown by the straight solid line. The applied voltage (arbitrary units of magnitude) is shown by open circles and a fitted curve gives an indication of the phase.

the velocity of the tail is in agreement with previous results for atmospheric-pressure surface discharges in argon,²⁰ this decrease in velocity with propagation distance may be attributed to the decreasing rate-of-change of the applied voltage as it approaches its maximum value within the RF cycle.

As the applied voltage approaches its minimum value (B), the discharge exhibits enhanced excitation activity close to the edge of the powered electrode. This becomes pronounced when the applied voltage exceeds 500 V (1000 ns in Figure 2) and is suggestive of a transition to the gamma mode.

When the rate-of-change of the applied voltage reverses again (C), the free electrons that were produced during (A) may be accelerated on the other side of the region of positive space charge, although with significantly weaker excitation.

Images corresponding to the E_i time-steps A-C are shown in Figure 4. Radially expanding excitation fronts,

FIG. 4. Images of the electron-impact excitation from the electronic ground state E_i at intervals A-C as defined in Figure 3. The edge of the powered electrode is shown by the solid line. The lines-of-sight over which the electron-impact excitation has been calculated for Figure 3 is shown by the dashed lines. The intensity of the excitation has been normalized with respect to the global maximum (enhanced online) [URL: <http://dx.doi.org/10.1063/1.4789371.1>].

FIG. 5. Propagation of the surface charge fronts during the extension phase (A in Figures 3 and 4) over sequential RF periods numbered 1-3 for which the amplitude of the applied voltage increases at a constant rate. Regions of increased electric-field strength at the tip of the surface-charge fronts and where these fronts overlap are shown by thick dashed lines. The region of positive space charge is not included for clarity.

which are similar with those previously observed for the impact of a discharge with a dielectric layer,²¹ appear in intervals of approximately $700 \mu\text{m}$ and this distance correlates very closely with the stripes of elevated optical-emission intensity in Figure 2(b) and excitation in Figure 3. It is interesting to note that the excitation fronts remain clearly visible in each image, even though the voltage amplitude and rate-of-change are different in each case. This suggests that there exists a localized increase in the electric field at these locations, which is not strongly dependent upon the phase of the applied voltage.

It is proposed that at the end of the extension phase (A in Figures 3 and 4), a portion of the secondary electrons generated by strong ionization within the front are deposited on the dielectric as surface charge. These remain on the surface for the remainder of the pulse and may result in a localized increase in the electric field at the tip of the surface-charge front. Due to an increase in the applied voltage throughout the pulse, as shown in Figure 2(a), the discharge propagates further and further away from the powered electrode with each RF cycle. Therefore, the localized electric field generated by the presence of surface-charge will also be enhanced at the places where each extension front overlaps that which preceded it. This process is depicted in Figure 5.

If the enhanced excitation intensity evident in these localized regions is caused by the presence of surface charge rather than the amplitude of the applied voltage, it can be expected that they are not strongly dependent upon the voltage phase and hence remain visible throughout the RF cycle.

In conclusion, optical emission spectroscopy has been used to determine the dynamics of relatively high-energy electrons, both spatially and with respect to the phase of the applied voltage, in a pulsed RF surface microdischarge in atmospheric-pressure argon. Spatially periodic excitation structures are observed to be highly repeatable throughout the pulse and may be explained by the generation of a localized electric field at the location where electrons have been deposited by previous propagation fronts.

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- ¹F. Iza, G. J. Kim, S. M. Lee, J. K. Lee, J. L. Walsh, Y. T. Zhang, and M. G. Kong, *Plasma Processes Polym.* **5**, 322 (2008).
- ²G. Y. Park, S. J. Park, M. Y. Choi, I. G. Koo, J. H. Byun, J. W. Hong, J. Y. Sim, G. J. Collins, and J. K. Lee, *Plasma Sources Sci. Technol.* **21**, 043001 (2012).
- ³F. Massines, N. Gherardi, N. Naudé, and P. Ségur, *Eur. Phys. J.: Appl. Phys.* **47**, 22805 (2009).
- ⁴D. O'Connell, L. J. Cox, W. B. Hyland, S. J. McMahon, S. Reuter, W. G. Graham, T. Gans, and F. J. Currell, *Appl. Phys. Lett.* **98**, 043701 (2011).
- ⁵D. Wang, D. Zhao, K. Feng, X. Zhang, D. Liu, and S. Yang, *Appl. Phys. Lett.* **98**, 161501 (2011).
- ⁶E. Moreau, *J. Phys. D: Appl. Phys.* **40**, 605 (2007).
- ⁷M. Šimor, Y. Creghton, A. Wypkema, and J. Zemek, *J. Adhes. Sci. Technol.* **24**, 77 (2010).
- ⁸A. Fridman, A. Chirokov, and A. Gutsol, *J. Phys. D: Appl. Phys.* **38**, R1 (2005).
- ⁹J. Park, I. Henins, H. W. Herrmann, and G. S. Selwyn, *J. Appl. Phys.* **89**, 15 (2001).
- ¹⁰D. B. Kim, J. K. Rhee, B. Gweon, S. Y. Moon, and W. Choe, *Appl. Phys. Lett.* **91**, 151502 (2007).
- ¹¹D. F. Opaits, M. N. Shneider, R. B. Miles, A. V. Likhanskii, and S. O. Macheret, *Phys. Plasmas* **15**, 073505 (2008).
- ¹²N. Balcon, A. Aanesland, and R. Boswell, *Plasma Sources Sci. Technol.* **16**, 217 (2007).
- ¹³J. Dedrick, R. W. Boswell, H. Rabat, D. Hong, and C. Charles, *Plasma Sources Sci. Technol.* **21**, 055016 (2012).
- ¹⁴K. Allegraud, O. Guaitella, and A. Rousseau, *J. Phys. D: Appl. Phys.* **40**, 7698 (2007).
- ¹⁵J. B. Boffard, C. C. Lin, and C. A. De Joseph, Jr., *J. Phys. D: Appl. Phys.* **37**, R143 (2004).
- ¹⁶T. Gans, D. O'Connell, V. Schulz-von der Gathen, and J. Waskoenig, *Plasma Sources Sci. Technol.* **19**, 034010 (2010).
- ¹⁷K. Niemi, S. Reuter, L. M. Graham, J. Waskoenig, N. Knake, V. Schulz-von der Gathen, and T. Gans, *J. Phys. D: Appl. Phys.* **43**, 124006 (2010).
- ¹⁸A. R. Hoskinson, L. Oksuz, and N. Hershkowitz, *Appl. Phys. Lett.* **93**, 221501 (2008).
- ¹⁹J. Dedrick, R. W. Boswell, P. Audier, H. Rabat, D. Hong, and C. Charles, *J. Phys. D: Appl. Phys.* **44**, 205202 (2011).
- ²⁰A. Sobota, A. Lebouvier, N. J. Kramer, E. M. van Veldhuizen, W. W. Stoffels, F. Manders, and M. Haverlag, *J. Phys. D: Appl. Phys.* **42**, 015211 (2009).
- ²¹L. Sun, X. Huang, J. Zhang, J. Zhang, and J. J. Shi, *Phys. Plasmas* **17**, 113507 (2010).