Forced random walks with memory in a glow mode
dielectric barrier discharge

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Abstract. In this paper, we present experimental results on dielectric barrier
gas-discharge (DBD) systems in helium where the surface is prepared with some
humidity. We observe well-defined solitary current filaments with self-propelled
motion. The trajectories of the filaments in the discharge plane resemble a random
walk motion with memory. The mechanism leading to motion is attributed to the
mutual interaction of gas-discharge and local humidity at the dielectric surface.
The phenomenon is a new drift mechanism for filaments in DBD and may be
important in applications being related to plasma surface treatment.

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1. Introduction

Random walk, e.g. Brownian motion, is a universal concept being used to describe many phenomena in natural sciences. There are several mechanisms leading to an interaction between the moving particle and the background resulting in memory effects. Depending on the interaction the memory leads to a self trapping walk with the particle being attracted by its own trajectory or a self-avoiding walk where the trajectory repells the particle [1]. Especially the latter one is an important topic of recent research, e.g. in the field of polymer chain formation [2]. Random walk with memory like self-avoiding walks is also interesting in the framework of self-propelled droplets on surfaces. Through chemical interaction between the liquid and the surface, a surface tension gradient emerges, that propels the droplet [3]–[5]. The droplet never returns back to a position on the surface that it had occupied before. A similar phenomenon is observed if microscopic islands of tin are deposited on a copper surface. The islands migrate across the surface, leaving a trace of bronze behind and avoiding their own path [6].

A typical self-avoiding walk on a square lattice [7] is shown in figure 4(a). The walker started on the lower left and came to a rest at the upper right side. In each time step the walker moves to an arbitrary neighbouring grid point avoiding those he visited before. Sooner or later every self-avoiding walk comes to an end as the walker gets trapped by his own path. In figure 4(a), this situation occurred in the upper right and is represented by a •. In the experiment, depending on the system, the walker may have a chance either to vanish and re-appear somewhere else or to escape from the trap, e.g. by exceptionally crossing its own path. The migrating tin islands mentioned above show the latter kind of behaviour [6].

In the present study, we will describe the discovery of random walk motion with memory in a dielectric barrier gas-discharge (DBD) device. The systems are well-known pattern forming systems [8, 9] exhibiting stationary or, often due to impurities in the gas, dynamic patterns [10, 11]. Beside periodic patterns also well-defined solitary current filaments are observed. These filaments have often been observed either as stationary or as travelling objects [12]–[14] but the origin of their motion remained largely obscure so far. In the present study, we demonstrate that humidity on the dielectric surfaces can cause self-avoiding motion.

In section 2, we will describe the experimental set-up of the gas discharge system. In section 3, we present experimental results on the dynamical behaviour of the discharge filaments. In section 4, we then develop a simple model that allows for an understanding of the mechanism that drives the motion of the current filament. This model will also be tested in various respect. Finally in section 5, we relate the observations of the present study to other systems where random walk motion with memory has been observed and we discuss the possible importance of the discovery for application.

2. Experimental set-up

The set-up of our system is sketched in figure 1(a). Basically it consists of two plane parallel glass plates with thickness \( d \). On the outer sides, the plates are coated with an ITO layer. These electrically conductive surfaces serve as electrodes and at the same time they are transparent with respect to the visible. The inner sides of the glass plates are roughened and become prepared as described below. Between the glasses there is a narrow gas gap with a circular cross-section, which is bounded by a dielectric spacer. The discharge space has a thickness of \( d = 0.5 \text{ mm} \) and
Figure 1. (a) Experimental set-up: two glass plates of thickness $a$ are separated by a spacer defining a circular gas-discharge space being filled with helium at pressure $p$ and having the discharge length $d$ and the diameter $D$. On the outside the glass plates are coated with ITO which represents the metallic electrodes. Through one of them the gas discharge can be observed via the emitted luminescence radiation density using sufficiently fast cameras. The system is driven by a sinusoidal high voltage supply with frequency $f$ and amplitude $\hat{U}$. Common parameters for all experiments are $d = 0.5\,\text{mm}$, $D = 40\,\text{mm}$ and $f = 200\,\text{kHz}$. (b) Light emitted from the dielectric barrier gas-discharge (DBD) prepared with dry surfaces. Exposure time is 50 ms. Parameters: $a = 0.7\,\text{mm}$, $\hat{U} = 634\,\text{V}$, $p = 201\,\text{hPa}\text{ He}$, other parameters see panel (a).

If the discharge system with dry surfaces is ignited, a filamentary discharge will occur typically as seen in figure 1(b). The filaments form a dense irregular arrangement and stay at their positions.

With the dielectric surfaces being prepared with humidity, a very different scenario is observed. After ignition, the whole discharge area is covered with current filaments that are arranged in a nearly hexagonal lattice, and in the beginning the filaments exhibit strong dynamics. The left picture in figure 2 represents this discharge mode.
Figure 2. ‘Freezing’ hexagons. Emitted light from the discharge gap at 0 s, 13.2 s, and 22.8 s after ignition. The driving voltage is just above the ignition voltage. Parameters: $a = 0.7$ mm, $\hat{U} = 465$ V, $p = 196$ hPa He, other parameters see figure 1(a). Camera repetition rate: 250 fps, exposure time: 100 $\mu$s.

While the discharge burns, beginning at the border more and more filaments stop moving and stay at a fixed position. In figure 2, at 13.2 s after ignition two regions in the discharge area can be identified. In the left and the right-hand side panel, standing filaments form a hexagonal arrangement. In the middle panel, we find a region with moving filaments. We note that directly at the boundary the hexagonal symmetry is broken and the filaments are arranged along the circular boundary with equal distance of adjacent filaments.

In the image being taken after 22.8 s in figure 2 all filaments stay at fixed positions. From now on there is hardly any variation in the pattern. The system has reached a stationary state. Neither a change of the parameters like pressure and driving voltage nor a re-ignition of the discharge can re-establish motion. Also an exchange of the working gas by fresh helium will not help. The only way to return to moving filaments is to repeat the preparation with humidity.

Two conclusions can be drawn at this point. Firstly, the humidity on the surfaces is essential for the motion of the filaments. And secondly, the surface humidity becomes consumed, while the gas discharge burns.

In a second series of experiments, again with humid dielectric surfaces, a single filament state is prepared. The system is initially operated closely above ignition voltage ($\approx 700$ V). This leads to a dense arrangement of filaments being arbitrarily scattered across the discharge area and being constantly in motion (figure 3, 0 s). We now switch the driving voltage to an amplitude of $\hat{U} = 640$ V which is below the ignition voltage. Waiting for some time, we observe that the number of filaments decreases due to collision processes until a single filament is left (figure 3; 1.864, 2.224, 2.246 s). The remaining filament moves across the discharge area with a typical trajectory being depicted in figure 4(b). This trajectory is recorded with a temporal resolution of 125 fps. If much smaller timescales are resolved, the trajectory becomes overlayed by a small amplitude chaotic motion. Such a scenario is observable for a few seconds up to more than a minute. Finally the filament will come to rest somewhere and stay there for ever (figure 3, 22.392 s).

Note that apparently surface humidity is necessary for filament motion and becomes consumed. As natural evaporation plays a minor role, the filamentary discharge is responsible for the drying of the surfaces. Consequently, there should be some relation between the sites that the filaments have visited and the position where the filaments come to a final rest. To clarify this point all frames in this record, beginning with ignition and ending with a stationary filament, were added (figure 3, integrated brightness). The brighter an area is, the more often it is visited by a filament. The arrow points to the position where the last filament stopped.
Figure 3. Preparation of a single filament. The grey border depicts the boundary of the discharge area. Parameters: $a = 1.0 \text{ mm}$, $\hat{U} = 640 \text{ V}$, $p = 200 \text{ hPa He}$, other parameters see figure 1(a). Camera repetition rate: 125 fps, exposure time: 100 $\mu\text{s}$.

Figure 4. (a) Exemplary trajectory of a self-avoiding walk on a square lattice ending when the walker becomes trapped ($\bullet$). (b) Typical experimental trajectory of the luminescence radiation density of a single filament projected on to the discharge plane covering a time-interval of $\approx 1 \text{ s}$. The end is marked by an image of the filament. Parameters: $a = 1.0 \text{ mm}$, $\hat{U} = 750 \text{ V}$, $p = 200 \text{ hPa He}$, other parameters see figure 1(a). (c) Detail view of the round shaded area in (b). The arrowheads indicate the direction of motion.

Apparently the filament came to a rest at a position that is rather bright and that, in contrast to its neighbourhood, has been visited very often by a filament before. Hence this position is supposed to be dry, whereas its surrounding is still covered with some attached water. We conclude that the discharge is responsible for the change of the surface properties forcing the filament to stop.

To be sure with the foregoing argument, we carried out a control experiment. We again prepared humid dielectric surfaces and mounted them in the discharge chamber. After the evacuation of the chamber and the subsequent filling with the working gas, we delayed the ignition for 5 min. In the previous experiments being performed without any delay, the final single filament comes to a rest after at most 3 min. So if the surface humidity simply evaporates
instead of being consumed by the discharge, there should be no motion in the pattern when the discharge is ignited after this delay. However, a scenario similar to that shown in figure 3 is observed. So obviously, it is the discharge that changes the surface humidity.

The next experiment demonstrates that indeed humid preparation is essential. Here, only the right half of the discharge area is prepared with humidity. The other one is left dry. In figure 5, some representative frames are shown. The first frame is captured immediately after ignition and depicts motion of filaments everywhere in the discharge plane. 1.88 s later, motion is suppressed on the left-hand (dry) side, whereas the filaments on the right-hand (humid) side are still in active motion. In the course of time ‘freezing in’ is observed which is very similar to what has been observed in the first experiment. At the boundary some filaments stop moving and stay there. But most of the filaments collide at the boundary with standing filaments and vanish. After 4.16 s, the pattern is stationary all over the domain. This proves that indeed humidity is the main cause for the filaments motion. Additionally, this experiment gives an upper boundary for the diffusion constant of the humidity on the dielectric surfaces. The assembly of the set-up, i.e. the duration from the humid preparation to the ignition of the discharge, lasts about 1 or 2 min. Within this time span the humidity smears out at least not more than a filament diameter. Hence, the diffusion of humidity on the surfaces play no or only a minor role for these experiments.

4. Qualitative explanation

In general, the generation and stability of current filaments in DBD is understood theoretically and can be reproduced numerically [8, 15]. This is possible as all relevant phenomena take place on a timescale of several breakdowns, which now a days is within the reach of numerical calculations. However, in the present experiments the movement of the filaments occurs on a comparatively slow timescale. Typically it takes some 10000 breakdowns for the filament to move the distance of its proper diameter. Therefore, it is understandable that numerically solving model equation is almost impossible.

Consequently, to get some insight into the mechanism of propagation of filaments, we use a heuristic model. The latter is based on three well-known physical properties of DBD systems. Firstly, we note that surface charges on the dielectrics resulting from transfer charges of the foregoing half cycle favour the reappearance of the filament at the same site. This is because during the onset of a breakdown the residual charges of the foregoing breakdown contribute to the
applied driving voltage in a constructive way. Secondly, impurities of electronegative gases, e.g. water, in the working gas, capture electrons and hence weaken the electron avalanche processes necessary for the ignition. So, the ignition voltage of the gas increases. Thirdly, the diffusion of water in helium is virtually zero on the timescale of a few breakdowns.

Of course, there are several physical processes that might influence the observed phenomena, too. For some of them, like possible changes of the secondary emission coefficient or the diffusion of humidity on the dielectric surfaces, we know from experiment that they are too weak to have an significant influence. Others, like the increase of surface conductivity due to humidity, would stabilize the resting filament and hence cannot play a dominant role. Therefore, we will now review our experimental data on the background of the physical mechanisms of which we think that they are the predominating ones.

We start with a situation in which the dielectrics is covered with humidity (figure 6(a)). At the position where a filament is re-ignited every half cycle some humidity is released into the working gas due to energy transfer from the plasma. As water is electronegative, within this cloud the ignition voltage is locally higher than in pure helium. This situation is unstable against lateral fluctuations of the filaments position. As soon as the filament is shifted to the right (figure 6(b)), the right edge of the filament is no longer exposed to the previously generated vapour cloud. Hence, at the right-hand side the ignition voltage is lower than in the filament centre. At the next rising slope of the driver voltage the ignition voltage is reached at the right edge first, and the filament is re-ignited there. The filament size is determined by fast timescales (particle travel time) and cannot change. The result is a moving filament leaving behind a cloud of impurities in the gas and a trace with less humidity on the surfaces. On a short timescale the cloud of impurities acts as a path memory with repulsion making the walk of the filament a self-avoiding walk. On a long timescale, the cloud of impurities and therefore the memory vanishes due to diffusion of water molecules in the gas. If one takes into account that the filament can move on a two-dimensional plane, the direction of movement is not fully determined by this mechanism. The probability to move in a certain direction is larger for forward directions and smaller for backward directions. The exact direction of movement is determined by any inhomogeneities of the surfaces, also the degree of humidity, and any other fluctuations that may be present in the gas discharge.

When many filaments have moved across the discharge area, a very inhomogeneous distribution of surface humidity is left. The integrated brightness in figure 3 may give an idea

Figure 6. Qualitative model for the interaction of filaments in gas discharge with humidity. (a) Some humidity is detached from the surface by the filamentary gas discharge and stays in the gas as vapour. (b) As the vapour increases the ignition voltage, the filament avoids the vapour cloud and moves aside. (c) Standing filament on a dry area, surrounded by humidity. Fluctuations to the side release water to the gas and force the filament to turn back.
of how the humidity distribution on the surface might look like. Bright places being occupied by a filament often before are nearly free of humidity. Figure 6(c) represents schematically a filament in such a dry place. As long as the filament is at rest, it does not produce any vapour any more. But if it shifts due to fluctuations, vapour is created forcing the filament to move back.

To support the introduced mechanism of motion, we estimate the diffusion properties of vapour in helium. The diffusion constant of water in helium is \( D_0 = 0.90 \text{ cm}^2 \text{ s}^{-1} \) at 1013 hPa (760 mmHg) \cite{16} or \( D = 0.18 \text{ cm}^2 \text{ s}^{-1} \) at 200 hPa. The filament velocity known from experiment is in the range of \( v = 100 \text{ mm s}^{-1} \). The filament diameter, \( d \) is about 1 mm. So the filament needs \( t = 10 \text{ ms} \) to move a distance as long as the own diameter. The diffusion length of vapour in helium during this time is \( l = \sqrt{Dt} = 0.42 \text{ mm} \). As \( l < d \), on the timescale \( d/v \) of the filaments motion the local concentration of vapour is not changing much. However, as \( l \) is not much smaller than \( d \), after the filament has moved the distance of several diameters enough time has passed so that the vapour concentration could be reduced significantly by diffusion in the vicinity of the former site. Now the latter can be revisited.

Let us now return to the experiment. From the previous considerations, we conclude that the filament performs—at least for short distances—a self-avoiding random walk. To check this prediction we have a closer look at the trajectory of a single moving filament shown in figure 4(b). At first glance there seem to be many intersections in the trajectory. But if studied in detail, we recognize two types of intersections. Firstly, there are intersections of parts of the path being visited long time ago. These intersections may occur with an arbitrary angle (figure 4(b); arrowhead). Secondly, if the filament is trapped by its proper path (figure 4(c)); it can escape from the trap by perpendicular crossing the trajectory exceptionally.

The situation described above is typical for the further course of the filament trajectory. Whenever possible, the filament avoids the recently visited path. Only if it is captured by a trap it performs a crossing.

A further test of the proposed model might be to examine the dependency of the motion from the gas temperature. As the memory depends on the diffusion coefficient, one might expect a longer lasting memory for lower temperatures. Preliminary measurements with a set-up cooled with liquid nitrogen (120 K) exhibit fewer crossings per time period than at room temperature. Unfortunately, more detailed measurements are not possible with the present equipment.

Another interesting point is a direct observation of the vapour. However, a direct observation via the spectral line at 309 nm of the OH-radical turned out to be impossible. Apparently, the corresponding excited state is not occupied in the gas discharge.

5. Conclusion

To conclude, our experimental investigations have demonstrated an important influence of humidity on the surface conditions on the behaviour of self-organized structures in AC gas discharge. On one the hand the surface humidity is important to get filaments moving. On the other hand the humidity is evaporated by a filament. Informal, we can consider this effect as a ‘smart’ cleaning process. During the treatment of the surface the filamentary pattern is in motion but when the treatment is finished, the pattern becomes stationary.

Within the framework of moving solitary objects the filaments can be regarded as self-propelled quasi-particles due to mutual interaction of the particle and the surface. On a short
timescale the type of motion is a self-avoiding walk. But in contrast to the self-avoiding walk in the strict mathematical sense, the current filaments can escape from traps and revisit sites that have been visited long ago.

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