Traveling Pairs of Spots in a Periodically Driven Gas Discharge System: Collective Motion Caused by Interaction

I. Brauer, M. Bode, E. Ammelt, and H.-G. Purwins
Institute of Applied Physics, University of Münster, Corrensstrasse 2/4, D-48149 Münster, Germany
(Received 12 November 1999)

We report on a new type of interaction between solitary current density spots observed in an ac-driven gas discharge system. These spots behave as independent particles as long as they are far apart from each other. Upon collision, in most cases one of the spots is extinguished. However, we also observe the formation of stable bound pairs of spots that move rather fast. We argue that both the rapid motion and the binding itself are due to a pronounced symmetry breaking with respect to size and breakdown phase of the individual spots. Basic features of pairs of spots traveling due to a true two-spot mechanism are discussed.

PACS numbers: 47.54.+r, 52.80.Pi, 82.20.Mj

The formation of patterns has been observed in various physical, chemical, and biological systems [1–5]. A special interest in the field of pattern formation lies in the investigation of localized structures. In particular, so-called spots, essentially unstructured, localized patterns of exact or approximate radial symmetry, can be generated by self-organizing processes in continuous media. Although such objects are built up of many microscopic elements, the macroscopic patterns may be treated as coherent objects, suggesting a description in terms of individual particles. Spots have been studied extensively, both experimentally and theoretically, in continuously supplied systems [6–10] as well as in setups with alternating driver where they have been called "oscillons" [11,12]. A rich variety of complex patterns may be interpreted as two-, three-, or even multiparticle structures which, in the case of bound states, leads to the notion of "molecules," "clusters," or, in special one-dimensional cases, "pulse trains" [13,14].

One important aspect concerning localized states is related to their motion that may set in either due to external influences such as parameter gradients or to internal degrees of freedom, the latter being referred to as self-motion; see, e.g., [15]. From the point of view of the particle picture described above, self-motion of a two- or more-particle structure may, in general, be induced either by a mere (nonlinear and in the simplest case symmetric) superposition of single-particle self-motion modes (class I) or by some more involved collective mechanisms (class II). In the first case, a single spot may undergo a transition to a moving state [16], and a traveling pair, e.g., is nothing but a bound state of two spots traveling in the same direction [13]. Different combinations of these single-spot motion modes give rise to more complicated cluster dynamics as, e.g., in the case of a rotating pair [17]. On the other hand, the single-spot modes that have to be combined to cause the collective propagation may be completely unrelated to single-spot motion, i.e., they may be mere shape modes, changing, for instance, the size of a spot that does not couple to its own translational degrees of freedom.

Albeit, such a classification is certainly possible and very general, up to now there was no example belonging to the second class of "molecule" motion truly based on interaction. The experiment we report on in the present article provides clear evidence for this most interesting kind of propagation.

We investigate traveling bound states consisting of two spots in a periodically driven, laterally two-dimensional gas discharge system. We will show that the formation of a traveling pair of spots is the result of a transition from simultaneous to subsequent ignitions of the respective spots. This leads to an attraction-repulsion cycle: first, the leading spot pulls the rear one and, shortly afterwards, it is pushed by the latter. Propagation of the spot pair is a consequence of this asymmetric interaction.

The gas discharge system under investigation is a dielectric barrier discharge consisting of a sandwich structure of two dielectric layers being separated by a gas layer (Fig. 1). Usual glass is used as dielectric. The outer surfaces of the dielectrics are in contact with an ac-voltage source. One of the electric contacts is an aluminum layer, while the other is a transparent and conductive ITO (indium tin oxide) layer, allowing the observation of the luminous density distribution emitted from the discharge gap. The entire arrangement is 1.6 mm thick, and is therefore thin

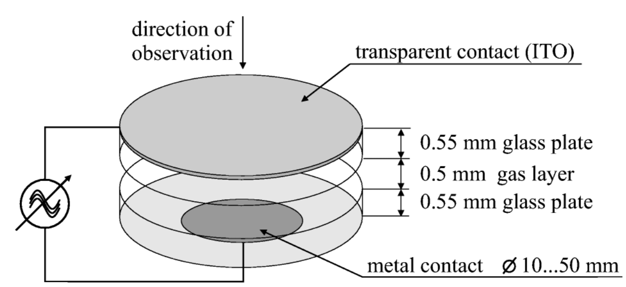

FIG. 1. Experimental setup: Two dielectric layers separated by a gas gap are electrically contacted on their outer surfaces. The device is supplied by a sinusoidal voltage of frequencies of typically $f = 200\,\mathrm{kHz}$ and amplitudes $\hat{V}$ up to 1 kV.

 0031-9007/00/84(18)/4104(4)$15.00

compared to its lateral extension of 10 mm. Experimental setup and system parameters are close to those used in flat plasma display panels [18,19], ozone generators, and excimer lamps [20].

The experimental system shows an extreme richness of current density patterns (visualized by the emitted light), many of which are composed of current density spots [21–23]. Such a spot is a channel in the gas layer of several 100 μm in diameter with a current density several orders of magnitude higher than the density in the surroundings. Single spots can be stationary or move randomly at velocities in the order of mm/s.

What seems to be a stationary spot of high luminous intensity for slow recording devices turns out to be a rapid switching on and off when using cameras with temporal resolutions higher than the period of the driver. When the external voltage reaches ignition potential on the rising edge of the sinusoidal driver, a breakdown takes place accompanied by charge carrier multiplication. Because of the applied electric field, charge carriers drift towards the dielectric walls where they accumulate, generating an electric field opposite to the externally applied field. Below a certain threshold value the total electric field becomes too weak to sustain breakdown, and the discharge current practically drops to zero. In the following half cycle, the externally applied electric field and the field of the wall charges that have been accumulated in the previous half cycle add up and a new breakdown takes place as soon as the ignition potential is reached once more. The current is now sustained until the old wall charges have reversed their sign and the threshold value of the total electric field below which no more breakdown is possible will be reached once again (Fig. 2).

The stability of an isolated spot as a localized pattern can be explained on a semimicroscopic level by the lateral interaction of drifting and diffusing electrons and ions in the gas and wall charges deposited on the dielectric surfaces during the breakdown process [24,25].

A pattern of particular interest is represented in Fig. 3. On a long time scale we observe a closed loop of high luminous intensity [Fig. 3a]. On the time scale of the driver period, however, the loop turns out to consist of a bound state of two spots [Fig. 3b], suggesting that the pair moves along the circular orbit of Fig. 3a. This was proved by taking high speed framing camera images presented in Fig. 4. Since it is impossible for a symmetric pattern to get into motion on its own, one should expect reflection symmetry to be broken in these structures, and, indeed, the two spots involved are of different size and total brightness. This difference is associated with different amounts of transferred charge: The small spot deposits fewer charge carriers on the dielectric walls than the big one. Now follow the rising edge of the sinusoidal driver voltage. Since external and wall charge field add up, the minimum electric field required for the ignition of the discharge is reached earlier at the location of the big spot than at the small spot. In the global signal one should consequently expect two distinct current peaks, and indeed this turns out to be the case (Fig. 5a). This phenomenon is specific for traveling pairs of filaments and was never observed in the case of patterns consisting of two or more well separated spots. By taking pictures with an exposure time of 100 ns we could show that the current peaks are in fact correlated with the successive breakdowns of the two spots [Fig. 5b].

The velocity of a traveling pair is 5–10 m/s and is thus significantly higher than the velocity of single, randomly moving spots. The displacement of a pair during one half cycle is about 2% of a spot's diameter, compared to about 10^{-4}% for isolated spots. The velocity of a traveling pair increases with increasing amplitude of the applied voltage, as can be seen in Fig. 6.

Pairs of spots do not form spontaneously out of the off state of the discharge, but are the result of a collision of single spots. Generally, in a collision of two slowly drifting spots one of the spots vanishes as a result of an attractive, short-range interaction, while the other one remains unchanged in form and size. Under the conditions investigated here, a bound state can be formed in the collision process instead. Single spots and pairs of spots can

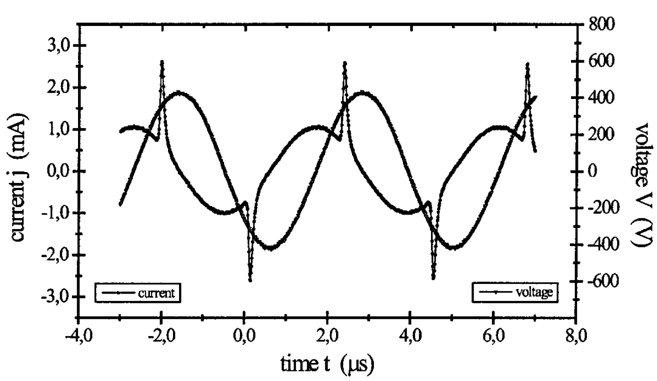

FIG. 2. Typical example for the driving voltage and the global current in the system shown in Fig. 1 using helium as gas at a pressure of 5000 Pa. Short current pulses arise when the voltage at the gas reaches the ignition level.

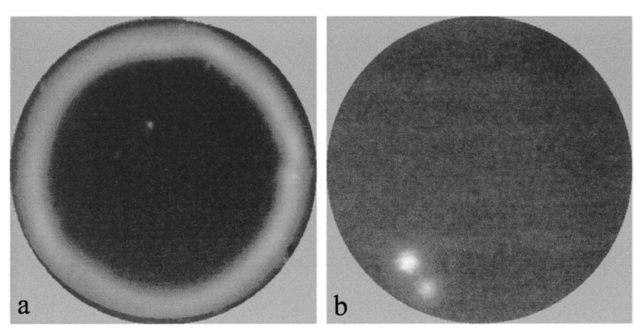

FIG. 3. When using a helium-air mixture as gas ($p =$ 5000 Pa + 2000 Pa air), a loop structure at the border of the active domain is observed when recording the luminous density distribution with a video camera [(a) exposure time 40 ms]. This loop turns out to be an asymmetric pair of spots on the time scale of the driver period [(b) exposure time 22.4 μs]. (Parameters: $\hat{V} = 620$ V, $f = 179$ kHz.)

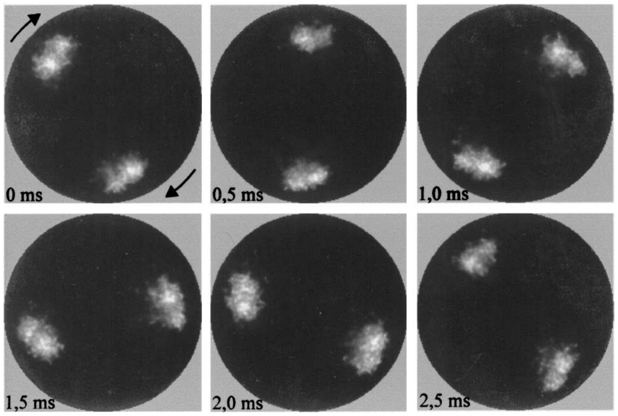

FIG. 4. Subsequent luminous density distributions recorded with a framing camera system. Two pairs of spots travel along the systems boundary. The small spot is the leading one in both pairs. Each picture was taken with an exposure time of 100 μs. (Parameters: $p = 5000$ Pa + 2200 Pa air, $\hat{V} = 650$ V, $f = 196$ kHz.)

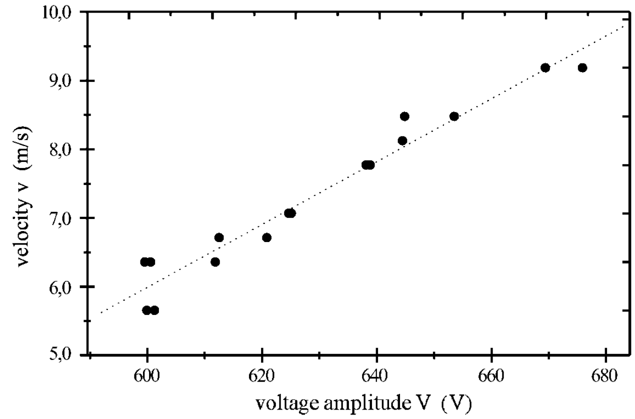

FIG. 6. Velocity of a pair of spots as a function of applied voltage amplitude. (Parameters: $p = 5000$ Pa + 2000 Pa air, $f = 196$ kHz.)

therefore coexist. As an example, Fig. 7 shows a snapshot of four equidistant pairs traveling along the systems boundary, circling around a stationary spot in the center.

The key to understanding the phenomenon of traveling pairs of spots is to develop a fairly detailed comprehension of the coupling of neighboring ignition processes. It is useful for a start to assume that the two spots are well separated, i.e., the distance between them is large, and interaction is weak. As a consequence of an individual spot's stability, both spots will almost behave as individual structures having identical shape and amplitude, and the driver phases at their respective ignitions will be the same. However, different from individual spots, they will move, due to their neighbors influence breaking translation symmetry.

Because of the wall charges, spots are associated with an electric field that resembles a dipole. From the point of view of a given spot at position x, its remote neighbor at position y enhances its own field and contributes a gradient prior to ignition. The same is true, of course, vice versa. So the maxima of the total field of both spots will increase slightly as compared to an individual spot and they will shift towards each other. As a consequence, both spots will ignite a bit earlier, and they will come a little closer to each other. The crucial point is to realize that, in addition, the interaction may destabilize the symmetrical behavior as soon as it is strong enough.

Assume that spot 1 (the big spot) ignites a little earlier on the rising edge of the sinusoidal driver than its neighbor.

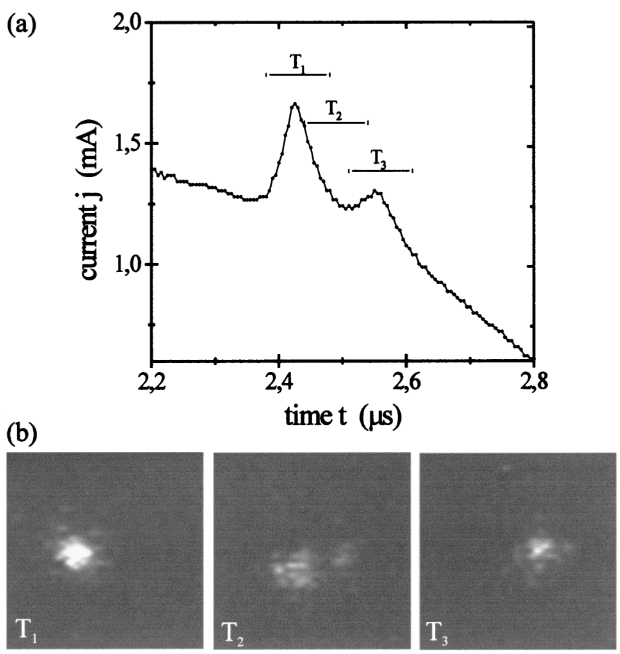

FIG. 5. In traveling pairs of spots the active current splits up into two pulses, the first one transferring more charge than the second one. The pictures of Fig. 5b taken with an exposure time of 100 ns reveal that the first and the second current pulse are correlated with the big and the small spot, respectively. The exposure time intervals T_1–T_3 are indicated in Fig. 5a. (Parameters: $p = 5000$ Pa + 2000 Pa air, $\hat{V} = 605$ V, $f = 220$ kHz.)

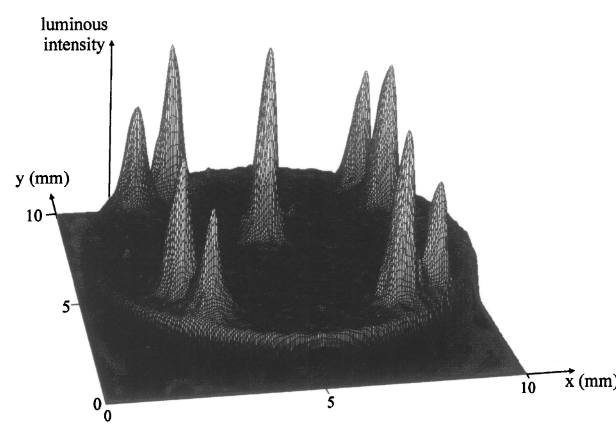

FIG. 7. Luminous intensity distribution of four pairs of spots traveling around a stationary spot in the center of the system. (Parameters: $p = 5000$ Pa He + 2000 Pa air, $\hat{V} = 670$ V, $f = 196$ kHz, exposure time 25 μs.)

As long as spot 1 burns, the corresponding wall charges will be compensated, weakening the support for ignition of spot 2. Before spot 2 starts to break down, there is a time interval in which its field does not change and will strongly support ignition 1. Hence, the amount of charges deposited on top of the walls at position 1 is increased as compared to the symmetrical situation. On the other hand, spot 2 will end up smaller than before, increasing the delay of the ignition times between the two spots. We may call this a phase instability referring to the driver phases at the respective times of ignition. Let us discuss the consequences in the extreme situation, when both burning intervals are completely separated. As before, spot 1, being ignited early, is attracted by spot 2. After spot 1 has stopped burning, it is associated with an inverted dipole field. This field influences spot 2: It will (a) delay ignition 2 and (b) repulse spot 2. This combination of opposite forces—the large and early spot is attracted, whereas the small and late one is pushed—is what makes a pair of spots move.

We simulated the experimental system by using a very simple phenomenological model, taking account of the most crucial aspects of the physical system as the periodic driver and the memory provided by the wall charges, only. We supposed that a localized spot of a given form is ignited as soon as the sum of external driver field and memory field reaches a given ignition threshold. After ignition, the spot, being activated by the memory field at the beginning of breakdown, inverts the memory field in the course of breakdown so that, finally, the extinction condition is reached and the spot is turned off. Simulating two spots with this model, we observe that the ignition of the spots is symmetric as long as they are far apart. When the distance between them falls below a critical value, symmetry breaking with respect to the moment of ignition and the size of the spots sets in. As in the experiment, we observe that the symmetry breaking leads to an attraction-repulsion cycle between the spots and hence a motion of the pair in the direction of the smaller spot. Details of the model and the numerical results will be published elsewhere.

In conclusion, we have presented basic features of traveling pairs of spots. We have shown that the motion of the pairs is due to a new mechanism based on a two-spot mode. Whereas single spots are more or less stationary the coupling of two spots leads to a symmetry breaking inducing a very effective mechanism for the motion of the pair. As this mechanism, interpreted as an attraction-repulsion cycle between the spots, is very general and does not include detailed physical aspects, we believe that this approach may also be applicable to other systems as, e.g., to dissipative solitary states in driven fluids [26] or current density filaments in ZnS:Mn based thin film electroluminescence structures [27].

[1] G. Ahlers, Physica (Amsterdam) **51D**, 421 (1991).
[2] M. C. Cross and P. C. Hohenberg, Rev. Mod. Phys. **65**, 85 (1993).
[3] H. Meinhardt, Rep. Prog. Phys. **55**, 797 (1992).
[4] R. Imbihl and G. Ertl, Chem. Rev. **95**, 697 (1995).
[5] S. C. Müller and V. S. Zykov, *Spiral Waves in Active Media* (World Scientific, Singapore, 1999).
[6] T. Ohta, M. Mimura, and R. Kobayashi, Physica (Amsterdam) **34D**, 115 (1989).
[7] K. J. Lee, W. D. McCormick, J. E. Pearson, and H. L. Swinney, Nature (London) **369**, 215 (1994).
[8] M. Bode and H.-G. Purwins, Physica (Amsterdam) **86D**, 53 (1995).
[9] A. L. Hodgkin and A. F. Huxley, J. Physiol. **117**, 500 (1952).
[10] L. M. Pismen, J. Chem. Phys. **101**, 3135 (1994).
[11] P. B. Umbanhowar, F. Melo, and H. L. Swinney, Nature (London) **382**, 793 (1996).
[12] C. Crawford and H. Riecke, Physica (Amsterdam) **129D**, 83 (1999).
[13] C. P. Schenk, P. Schütz, M. Bode, and H.-G. Purwins, Phys. Rev. E **57**, 6480 (1998).
[14] C. Elphick, E. Meron, and E. A. Spiegel, SIAM J. Appl. Math. **50**, 490 (1990).
[15] A. Mikhailov and D. Meinköhn, in *Stochastic Dynamics*, edited by L. S. Schimansky-Geier and T. Pöschel (Springer, New York, 1997).
[16] M. Or-Guil, M. Bode, C. P. Schenk, and H.-G. Purwins, Phys. Rev. E **57**, 6432 (1998).
[17] C. P. Schenk, A. W. Liehr, M. Bode, and H.-G. Purwins (to be published).
[18] A. Sobel, IEEE Trans. Plasma Sci. **19**, 1032 (1991).
[19] C. Punset, J.-P. Boeuf, and L. C. Pitchford, J. Appl. Phys. **83**, 1884 (1998).
[20] U. Kogelschatz, B. Eliasson, and W. J. Egli, J. Phys. IV (France) **7**, C4-47 (1997).
[21] E. Ammelt, D. Schweng, and H.-G. Purwins, Phys. Lett. A **179**, 348 (1993).
[22] I. Müller, E. Ammelt, and H.-G. Purwins, Phys. Rev. Lett. **82**, 3428 (1999).
[23] W. Breazeal, K. M. Flynn, and E. G. Gwinn, Phys. Rev. E **52**, 1503 (1995).
[24] I. Müller, C. Punset, E. Ammelt, H.-G. Purwins, and J. P. Boeuf, IEEE Trans. Plasma Sci. **27**, 20 (1999).
[25] I. Brauer, C. Punset, H.-G. Purwins, and J. P. Boeuf, J. Appl. Phys. **85**, 7569 (1999).
[26] O. Lioubashevski, Y. Hamiel, A. Agnon, Z. Reches, and J. Fineberg, Phys. Rev. Lett. **83**, 3190 (1999).
[27] S. Zuccaro, F.-J. Niedernostheide, B. Kukuk, M. Strich, and H.-G. Purwins, Phys. Rev. E (to be published).