Formation of Clusters of Localized States in a Gas Discharge System via a Self-Completion Scenario

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Formation of extended structures composed of localized states via the self-completion scenario is observed. The localized states are actually current filaments in a dc driven planar semiconductor–gas discharge system. The structure grows from a solitary localized state in the subcritical domain, where the homogeneous state is stable. It is proved that compact hexagonal clusters form due to the attractive interaction between distant filaments. A semiphenomenological model of the activator-inhibitor type is proposed which suggests that the Turing mechanism triggers the phenomenon. [S0031-9007(97)04156-2]

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Extended systems driven out of equilibrium are known to form structures where the homogeneous background coexists with excited spatially localized domains [1]. Such components of patterns have been referred to as localized states (LS) of active media [2]. In different treatments LS's became known also as autosolitons [3] or dissipative solitons [4]. The intriguing problem is the formation of complex structures composed of the LS's with their quasiparticle character (see Refs. [5–7]). In their pioneering numerical simulations of morphogenesis, Meinhardt and Gierer have demonstrated development of structures from LS's via the appearance of new LS's around those emerged earlier [8,9]. Later such a scenario was named *self-completion* [3]. We are not aware of any unambiguous experimental proof of the self-completion mechanism, whereas the other scenario of multiplication of LS's, via their division, has been observed [10,11].

In the present Letter we report the formation of clusters of LS's via the self-completion scenario in a quasi two-dimensional (2D) gas discharge system. In our case the LS's are filaments of electric current. The experimental results are supported by simulations based on the original semiphenomenological model, which demonstrates the Turing type of instability. Both experiment and numerical results reveal the attractive interaction between distant LS's. Thus, the latter can form compact hexagon clusters. However, LS's retain their individual particlelike nature. In the experiment, under the action of unavoidable noise, they can escape from a cluster, wander over the active area of the system, and stick again to a cluster edge.

The experimental setup used is a planar gas discharge cell equipped with a resistive electrode made of silicon doped with deep impurity of Zn or Au. The thickness of the silicon wafers d_s is in the range 0.45–1.0 mm and the diameter of the active area of the cell is 20 mm. The discharge gap, whose width d_g is in the range 0.8–1.4 mm, is filled with gaseous nitrogen at the pressure $P = (1–2) \times 10^4$ Pa. To provide high resistance of the electrode the cell is cooled down to $T \approx 90$ K. The value of the discharge current is controlled by the feeding dc voltage U_b and by the resistance of the electrode which in turn is controlled by the homogeneous stationary illumination from a tungsten lamp. More comprehensive descriptions of the device have been given in previous works, where spontaneous formation of small-amplitude stripe and hexagon patterns [12] and zigzag instability of solitary stripes [13] has been reported. As compared with experimental conditions of these works lower resistance of the electrode has been applied in the present research. The destabilization of the homogeneous state is followed in this case by the filamentation of current. In contrast to other works dealing with gas discharges where spontaneous formation of patterns was observed (see Refs. [10,14–16]), the system under study exhibits pronounced planar geometry on the one hand, and reveals pattern formation at low dc electric current on the other.

At low voltage U_b the discharge is not ignited and the system is characterized by the "dielectric" branch (A) in Fig. 1. At some threshold voltage U_0 a self-sustained homogeneous stationary discharge develops in the gap. Its current grows nearly linearly with voltage as shown by the "conductive" branch (B) in Fig. 1. With increasing voltage we observe the spontaneous formation of a hexagon pattern composed of current filaments. This occurs at the voltage U_{up}; see branch (C) in Fig. 1. Generally, the number of filaments in the pattern grows with further increase in voltage. When diminishing the control voltage along branch (D) in Fig. 1, the number of filaments in the pattern decreases. At the point U_{down} the last filament disappears, and no current can be detected in the device. In order to reignite the discharge, we again go along the dielectric branch (A) on the hysteretic loop. The observed subcritical behavior is in accordance with general regularities of spontaneous formation of hexagon patterns

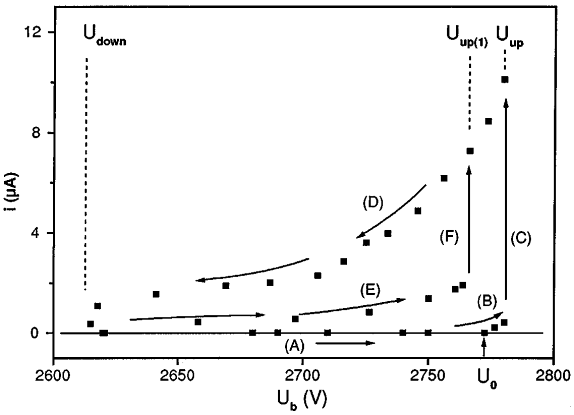

FIG. 1. The characteristic current-voltage loop connected with the filamentation of current in the device. (*A*) dielectric branch; (*B*) conductive branch: Ignition of the homogeneous discharge at the critical voltage $U_b = U_0$; (*C*) spontaneous formation of a hexagon pattern from the homogeneous state at the critical voltage U_{up}; (*D*) successive decay of the number of filaments in the pattern; (*E*) branch for one filament (solitary LS); (*F*) formation of a hexagon cluster, induced by a LS, at the critical voltage $U_{\mathrm{up}(1)} < U_{\mathrm{up}}$. The diameter of the discharge area is 20 mm. Parameters: $d_g = 1.0$ mm, $d_s = 1.0$ mm, N_2 pressure $P = 1.68 \times 10^4$ Pa.

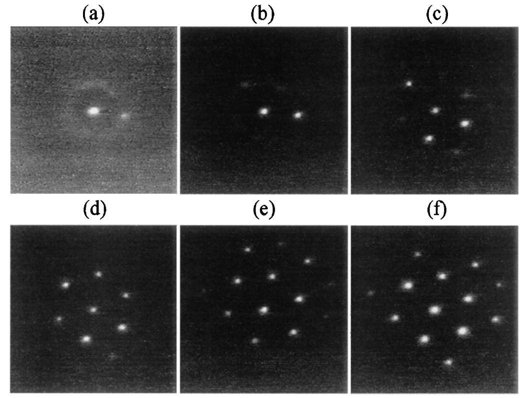

FIG. 2. Growth of the hexagon cluster from a solitary LS (the self-completion scenario). The sequence of snapshots (a)–(f) is obtained under a slight increase of the feeding voltage in the domain of transition (*F*) in Fig. 1. An image acquisition technique working with the framing rate 25 Hz was used. The linear dimension of the area shown is $L = 14$ mm. Parameters: $d_g = 1.4$ mm, $d_s = 0.45$ mm, $P = 1.5 \times 10^4$ Pa, $U_b \approx 3200$ V.

in nonequilibrium systems [1]. However, in our case the value U_{down} is much lower than the voltage U_{up} needed to form a hexagon pattern. This indicates the strong subcriticality of the system.

Decreasing U_b along branch (*D*) of the loop in Fig. 1, we can achieve a state with a single stable filament, which is actually the solitary LS. Branch (*E*) in Fig. 1 also illustrates the response of the system containing one LS to the voltage increase. The arrow (*F*) marks the voltage level where the hexagonal structure develops from the solitary LS. The intrinsic noise of the device makes it difficult to determine exact values of the critical voltages U_{up} and $U_{\mathrm{up}(1)}$: The lower U_b is compared to these values, the longer is the time that is needed for a corresponding transition to occur. We also note that pattern formation occurs at small current, i.e., at low power dissipated in the device. This is also valid for the data presented in Figs. 2–4, where we do not specify the current amplitudes.

Figure 2(a) shows an example of a single filament state. The filament consists of a bright core which is surrounded by a ring. Such a structure of LS's has been discussed in a number of theoretical (analytical and numerical) works [3,4,6,8]. An increase in the voltage leads to the appearance of other filaments, which grow almost exactly on the ring [see Figs. 2(b) and 2(c)]. Figures 2(d)–2(f) show the further growth of the pattern, which develops through the building up of new filaments on hexagon lattice sites. The final pattern in Fig. 2(f) for the given voltage is a localized hexagon cluster on the homogeneous background. Since the cluster formation from the initial LS occurs in the very narrow range of the feeding voltage, it corresponds to vertical branch (*F*) in Fig. 1. Formation of a structure via successive building up of additional LS's on maxima of oscillating tails of existing LS's has been described in several theoretical works [3,6,8]. Following Ref. [3] we refer to this phenomenon as the "self-completion" process.

The nonlinear properties of the system depend on experimental parameters (such as the values of P and d_g). The self-completion scenario is pronounced when the thresholds for spontaneous and induced generation of LS's (voltages U_{up} and $U_{\mathrm{up}(1)}$) are well separated. This implies the strong subcriticality of the system. The amplitude of LS's against the homogeneous background also depends on the experimental parameters, and the role of noise can be fairly important for the stability of a cluster. Noise can

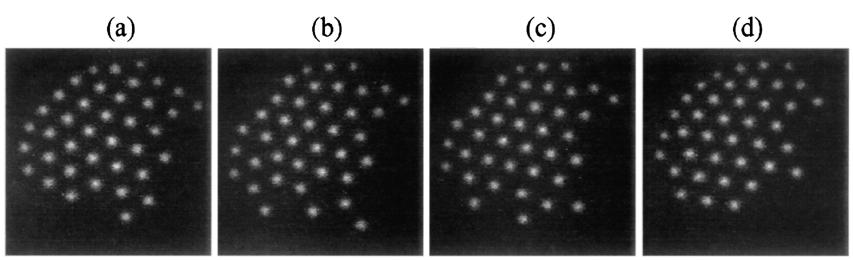

FIG. 3. Example of the interaction of a large cluster of LS's with "evaporated" LS's. Snapshots have been obtained at a fixed $U_b > U_{\mathrm{up}}$ (see Fig. 1) at successive times (a) 0 sec, (b) 8 sec, (c) 10 sec, and (d) 23 sec. $L = 20$ mm. Parameters: $d_g = 0.8$ mm, $d_s = 1.0$ mm, $P = 1.3 \times 10^4$ Pa, $U_b = 1900$ V.

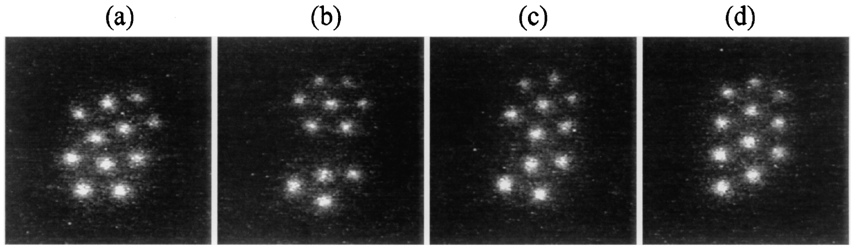

FIG. 4. Illustration of the attractive interaction between LS's in the cluster. The system is at $U_{\mathrm{down}} < U_b < U_{\mathrm{up}(1)}$ on branch (D) of Fig. 1. Shown are (a) the nonperturbed state; (b) the reaction of the system to the local perturbation; successive stages of pattern relaxation at $t = 6$ sec (c) and $t = 9$ sec (d) after the perturbation is removed. $L = 15$ mm. Parameters: $d_g = 0.8$ mm, $d_s = 1.0$ mm, $P = 1.37 \times 10^4$ Pa, $U_b = 1900$ V.

destroy the hexagonal order of a cluster. This is demonstrated in Fig. 3, which presents the evolution of a large cluster at fixed parameters. The noise induces the "evaporation" of LS's from hexagon facets into the surrounding active space of the system, their wandering and subsequent condensation onto the same or other boundary cites of the cluster "lattice." Such a behavior exemplifies quite clearly the quasiparticle nature of LS's. It strongly resembles the process of interaction of a condensed phase with its vapor, which is responsible for such phenomena in condensed matter physics as the Ostwald ripening [17].

The next experiment shows quite unambiguously the existence of attractive interaction between LS's, which gives rise to their condensation into a compact hexagonal cluster. While going along the route $(A) \rightarrow (B) \rightarrow (C) \rightarrow (D)$ of Fig. 1, a stable hexagon had been prepared; see Fig. 4(a). Then the active area of the device containing the cluster was partly screened from the light exciting the semiconductor electrode. In this way the cluster has been separated into two parts; see Fig. 4(b). After removing the perturbation the system relaxes, so that the LS's again form a cluster, which now can be of a different shape; see Figs. 4(c) and 4(d).

It has been shown during the last decade that formation of patterns in some gas discharge [15] and semiconductor gas discharge [16] systems can be properly interpreted on the basis of reaction-diffusion equations. Trying to comprehend the observed phenomena, we apply below a simple semiphenomenological reaction-diffusion model for the transport of electrical current in our two layer system. As variables we use the potential drop U across the discharge gap and the density of charge carriers N. Proceeding from the previous study concerning the local kinetics of these variables in the system [18], the equations can be written as

$$\frac{\partial U}{\partial t} = \frac{U_b - \gamma \overline{N} - U}{\tau_U} - cNU + D_U \Delta U\,, \tag{1}$$

$$\frac{\partial N}{\partial t} = -\frac{N}{\tau_N} + NU\left[a + b\left(\frac{N}{N + N^*}\right)^2\right] + D_N \Delta N\,, \tag{2}$$

where the first equation describes the charging up of the capacity of the discharge gap from an external voltage source U_b with the characteristic time τ_U [19] and its discharging due to the presence of free carriers in the gap (c is the constant of the discharging). The term $\gamma\overline{N}$ takes into account the global negative feedback existing in an electrical system: Voltage supplied by a battery drops partly on passive resistive components of the device and on external loads. We consider this feedback to be proportional to the total number of carriers in the gap. To include it in the local Eq. (1), its value is normalized to the average density

$$\overline{N} = S^{-1} \int_S N\, dS\,,$$

where S is the area of the system.

The dynamics of the carrier density is governed by their decay with lifetime τ_N and avalanche multiplication, the first and second terms on the right-hand side of Eq. (2), respectively. The efficiency of the autocatalytic avalanche process is determined by the constants a, b, and N^*. For low current (i.e., for small N) the rate of multiplication of the carriers during their drifting through the gap does not essentially depend on N. An increase in current is accompanied by a decrease in the voltage drop across the gap. This "falling" part of the current-voltage characteristic reflects the negative differential conductance (NDC), which indicates transition to the glow regime of the discharge. Thus, Eq. (2) takes into account phenomenologically the main features of low-current discharges including the Townsend and glow domains (see Ref. [20]). The last terms in Eqs. (1) and (2) describe the diffusive spreading of variables in the plane of the system. We stress that it is the NDC of the discharge domain which gives rise to pattern formation in the systems considered; see Refs. [10,16].

The proposed model demonstrates two successive bifurcations when the control parameter U_b is varied. First, with increasing U_b the transcritical bifurcation from a dielectric state $N = 0$ to the homogeneous conductive state $N > 0$ takes place. This bifurcation occurs at $U_b = 1/a\tau_N$ and corresponds to the point U_0 of the experimental characteristic in Fig. 1. For $D_U \gg D_N$ and $\tau_U \gg \tau_N$, further increase of U_b is followed by the destabilization of the homogeneous state via the Turing bifurcation; i.e., a mode with some spatial period Λ_c grows [21].

Numerical simulations using the standard Euler scheme confirm stability analysis results: At parameter U_b exceeding some value (U_{up} in Fig. 1), a pattern forms which

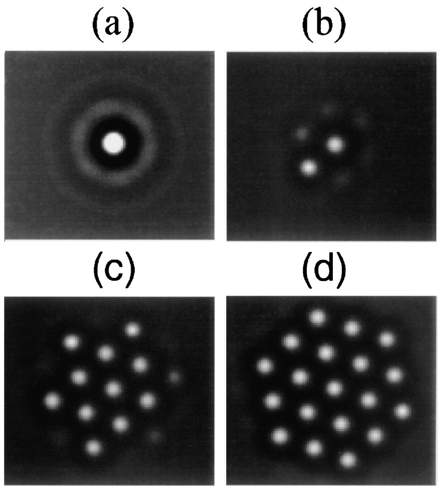

FIG. 5. Growth of the cluster from a solitary LS as obtained with Eqs. (1) and (2), where $\tau_U = 10^{-2}$ sec, $\tau_N = 10^{-3}$ sec, $D_U = 0.625$ cm^2/sec, $D_N = 0.045$ cm^2/sec, $\gamma = 10^{-1}$, $a = 1$, $b = 0.4$, $c = 1.64 \times 10^{-4}$ cm^3/sec, $N^* = 1.5 \times 10^5$ cm^{-3}. The calculated threshold voltages U_{up}, $U_{\mathrm{up}(1)}$, and U_{down} (see Fig. 1) for a domain with dimension $L = 2.72$ cm are 7225, $\approx$6388, and $\approx$2160 V, respectively. Initiation of the growth is due to the stepwise increase of U_b from the value $U_{\mathrm{down}} < U_b < U_{\mathrm{up}(1)}$ to $U_b = 6390$ V.

consists of large amplitude spots arranged in a hexagonal lattice with spatial period on the order of Λ_c. Subsequent decreasing of U_b leads to the successive dying out of some spots, and the structure becomes more crumbly. If we stop decreasing U_b and continue the integration, the surviving spots drift close to each other and organize themselves in a compact hexagon cluster. This indicates the attractive interaction between LS's in our case. Repeating this procedure we can achieve the state with just a single LS. Thus, our simulations reproduce branch (*D*) on the experimental loop of Fig. 1. The radial distribution of N in a solitary spot has the form of the core surrounded by an oscillating tail which decays in the radial direction.

Starting simulations from the state with the stationary solitary LS, we also reproduce the scenario corresponding to branches (*E*) and (*F*) in Fig. 1. Snapshots obtained from these simulations are presented in Fig. 5. The increase in U_b is accompanied by the swelling of the nearest ring surrounding the LS and breaking the rotational symmetry of the ring with a tendency to create six additional spots; see Fig. 5(a). Like in the experiments [see branch (*F*) in Fig. 1], at some threshold value $U_b = U_{\mathrm{up}(1)} < U_{\mathrm{up}}$ additional spots of the same amplitude are generated. Other conditions remaining unchanged, the number of spots in a stable cluster is controlled by U_b and γ. For the transient process presented in Fig. 5, the value of U_b is high enough to support the existence of a cluster of 19 spots. Similar to the experiment, we refer to this transient as a process of self-completion. The amplitude of the spots in the cluster exceeds about 7 times that for the homogeneous background and strong subcriticality of the process is observed.

In conclusion, in the activator-inhibitor system we have observed formation of complex structures via the self-completion process. The localized states which build an extended pattern to a large extent retain their individual (quasiparticle) nature. Phenomena like clustering and evaporation of LS's from clusters are registered. This shows the striking similarity of some properties of dissipative structures and crystals.

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