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# SELF-ORGANIZATION SCENARIO RELEVANT FOR NANOSCALE SCIENCE AND TECHNOLOGY

M. Sanduloviciu, D. G. Dimitriu, L. M. Ivan, M. Aflori, C. Furtuna,
S. Popescu, E. Lozneanu*

Complex System Laboratory, Al. I. Cuza University of Iasi, Romania

Based on a scenario of self-organization suggested by plasma experiments we explain the emergence of metastable complex space charge configurations in the form of luminous spots of micrometer sizes and also its ordered spatial distribution in dc micro-discharges. Endowed with memory, these metastable complex space charge configurations give to the system the ability to work as an intelligent multifunctional network. Starting with the assertion that the intrinsic mechanism of self-organization is the same in all states of aggregation of the matter and that only the dimensions of the metastable configurations depend on the matter concentration it becomes possible to offer a new insight concerning the genuine origin of self-organized pattern with sizes in the range of micro and nanometers.

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## 1. Introduction

New ideas concerning the physical basis of self-organization phenomena observed in the so called intelligent materials miniaturized at micrometer and nanometer sizes gained substantial interest because of the wide range of their technological purposes. Thus, miniaturization makes possible the implementation of better computer architecture but the presence of physical limits imposed by quantum and thermal fluctuation phenomena makes such systems less and less reliable. The recognition of these ultimate limits has lead computer scientists to seek inspiration from biology. This is because living organisms operate with functional elements that are of mesoscopic scale dimensions and actually exploit collective quantum effects and thermal energy for ensuring its “living” state. The hope to break the barrier of miniaturization seems to lie in the knowledge of the self-organization mechanism able to explain the self-assemblage and the working regime of the simplest organisms created by nature.

In solid-state physics and electronics, a large variety of different nonequilibrium phenomena accompany the spontaneous self-assemblage of spatial and spatio-temporal patterns [1]. Thus, attention has been paid to thyristor-like semiconductor structures with large active area, as these nonlinear systems with bistable properties show several spatial and spatio-temporal current density patterns. Such semiconductors could potentially be used as multi-stable elements for integrated circuits, self-organizing devices for image recognition and image processing. It is remarkable that the instability mechanism observed in such samples exibits some features very similar to those studied in biological or chemical media.

Self-organization phenomena can be revealed also in non-crystalline materials in which metastable configuration of atoms at nanometer scale gives to the material the possibility to choose one among various pathways to change free energy [2]. This occurs when, by providing some energy, the system is pushed to modify the quasi-equilibrium state in one of the multiple states of higher/lower energy. As assumed, the self-organization phenomenon ensures the reversibility of the atomic state during energy exchange with the surrounding medium.

* Corresponding author: : erloz@uaic.ro

Referring to nonlinear phenomena related to pattern formation observed in gaseous conductors (plasma), the so-called micro-discharges are interesting for nanoscience and technology. These are produced in devices whose main parts are a semiconductor plate of high resistivity and a gas discharge gap in direct contact with each other [3]. Working under relative high gas pressures, the observed spatial patterns in such devices, usually known as anode glows or spots, have sizes in the order of magnitude of micrometers. Such devices are used, for example, as high-speed conversion of infrared images into visible one.

Our attempt in this paper is to present experimental arguments demonstrating that the self-organizing process observed in all aggregation states of the matter is essentially based on the same intrinsic mechanism. This mechanism explains the emergence of self-organized systems in laboratory [4-8] but also in the nature [5,9,10]. For nanoscience and technology the consideration of this self-organization mechanism offers a new insight into a phenomenology not yet considered.

## 2. Experimental results and discussion

The answer to the problem if self-organized systems working as multifunctional materials have intelligence depends on what we mean by intelligence. Associating the intelligence with what human brain can do, it results that memory associated with learning is one of the most important characteristic by which a system can be classified to be intelligent. In a gaseous conductor the quality of a self-organized complex space charge configuration (CSCC) to work as a multifunctional intelligent system is based on a special kind of memory that endowed it to perform operations "learned" during its spontaneous self-assembling process.

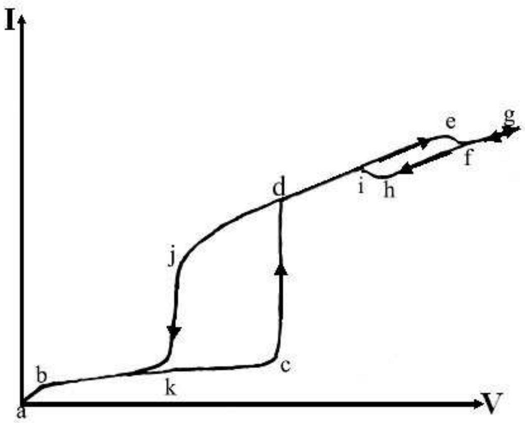


Fig. 1. Static current-voltage characteristic.

As well established, self-organized CSCC appears in gaseous conductors when an external constraint, in the form of a local electric field created by an electrode immersed in plasma in thermodynamic equilibrium, accelerates the electrons delivered by the plasma at kinetic energies for which in adjacent regions the excitation and ionization rates suddenly increase [4-8]. The net negative space charge appears in the region where, after atom excitation, a part of the thermal electrons delivered by the plasma gets kinetic energy for which the excitation rate suddenly increases. The net positive space charge appears in the adjacent region where the accelerated thermal plasma electrons have obtained kinetic energies for which the ionization cross-section suddenly increases. Because of their small mass with respect to the positive ions the electrons that produced and that result after ionizations are quickly collected by the positive electrode so that a net positive space charge appears. This is placed in a region adjacent to the net negative space charge formed by electrons that have lost their kinetic energy after excitations. Consequently, electrostatic forces acting as long-range correlations, determine the self-assemblage of a double layer space charge arrangement. In spite of the loss of charged particles by recombination, diffusion and so on, this double layer continues to exist because new carriers that replaced the lost one accumulate in the adjacent regions where the excitation and ionization cross sections suddenly increase. This means that the existence of the double layer is ensured in a dynamical state in which it is continuously assembling. By local acceleration of electrons collected from the surrounding plasma the double layer contributes itself to this self-assembling process. The double layer becomes self-consistent when, after increasing the voltage applied on the sample, the long-range correlation forces acting between the adjacent net space charges become dominant with respect to the external forces that have initiated the double layer formation. More in detail these phenomena were already described [4-9].

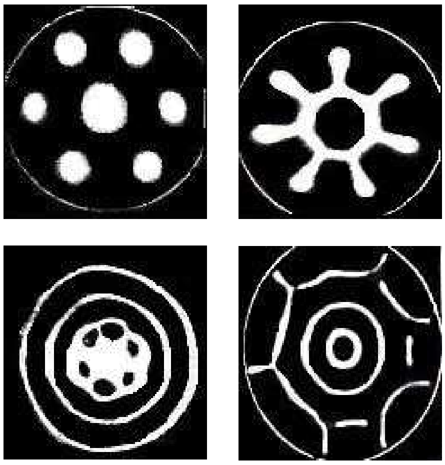

Fig. 2. Anode spots and a network of structures formed at large anodes in dc gas discharge in different experimental conditions [11].

In the following we will discuss the physical basis of the special kind of memory emphasized by systems emerged after self-organization. In gaseous conductors the presence of such systems is emphasized by plotting the static current-voltage *I(V)* characteristic. Such a typical *I(V)* characteristic is shown in Fig 1. It was obtained from a diode where the conductor is a plasma created in a plasma source that acts as cathode. Because of the presence of a load resistor the voltage drop *V* on the plasma conductor is different from the voltage $V_{PS}$ delivered by the external dc power supply (PS). This static *I(V)* characteristic emphasizes abrupt increase and decrease of *I* when $V_{PS}$ is gradually increased and decreased. Self-organized CSCC in a metastable state emerges at the anode of the diode concomitant with an abrupt increase of the electrical conductivity. In Fig 1 the abrupt increase of *I* appears when *V* reaches the critical value marked by **c**. When $V_{PS}$ is further gradually increased an abrupt decrease of *I* related to periodic limitation of the current *I* are observed when *V* reaches the value marked by **e**. When the voltage delivered by the PS is gradually decreased, two hysteresis loops are revealed in Fig 1. The first reveals that the self-organized CSCC attached to the anode is able to perform all operations needed for its existence also when *I* runs through the branch **d-j,** *i.e.*, when *V* has values smaller than that required to initiate its self-assembling process. This means that the CSCC was endowed during its self-assembling process with a special kind of memory that manifests in its ability to continuing all operations "learned" during its self-assembling process also when the external causes that have initiated its emergence are removed. For working as a bistable system, the voltage *V* supported by the gaseous conductor must be varied in such a way that the current *I* runs from the value marked by **c,** for which the CSCC self-assemblies, to the value marked by **j,** for which the CSCC disrupts. This means that the bistability is controlled by $V_{PS}$ that also controls the current *I*. When the circuit that includes the CSCC contains a system able to oscillate naturally these oscillations can trigger the bistability so that oscillations are maintained in the system. Under such conditions the gaseous conductor works as an S shaped NDR. For generating oscillations the current *I* runs through a "duty cycle" that corresponds to the hysteresis loop **c-d-j-k-c**.

The spontaneous transition of the gaseous conductor from a state in thermodynamic equilibrium into a state when, after self-organization, the CSCC emerges is accompanied by a change of free energy produced in the moment when the matter and energy injected by the PS reaches a critical value. In this moment the gaseous conductor is pushed to modify its quasi-equilibrium state in a new state in which the CSCC acts as a new internal source of free carriers so that its internal resistance decreases abruptly. This means that, under the conditions that at the anode a “network” of mettastable CSCCs emerges, the sample can transit between multiple states of higher/lower energy.

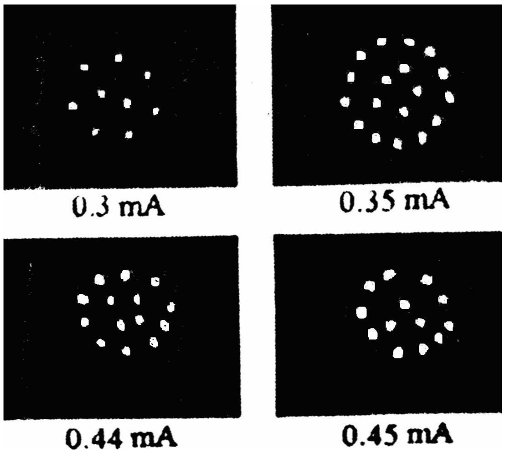


Fig. 3. Anode spots emerged in helium discharge at high pressure

The presence of a network of anode glows and of “chain-line” structures emerged after self-organization in usual dc gas discharge is showed in the photographs presented in Fig 2 [11]. More interesting for nanoscience are similar photographs of “networks” of anode spots formed after self-organization in micro-discharges [3]. These self-assembly more frequently at the anode but, as recently reported [12], a similar self-organization process was observed in the cathode layer of a dc gas discharge. As mentioned in [12], at the date of the publication of the paper the authors were not able to explain the observed self-organization phenomena.

In our opinion, the phenomena observed in micro-discharges can be explained considering the same self-organization process as described in [4-8]. As known the anode spot, *i.e.*, the CSCC, transits into a mettastable state when the double layer from its border becomes self-consistent. This is realized when the electrons accelerated in its potential drop ensure an ionization rate at its high potential side sufficiently to balance the net negative space charge at its low potential side [8]. Since the ionization rate strongly depends (beside of the flux and the kinetic energy of the electrons that transverse the double layer) on the concentration of the atoms in the ground state, the size of the CSCC decreases simultaneously with the increase of the atom concentration.

Referring only to the CSCCs self-assembled in the next vicinity of the cathode [12] we take into account that the dc xenon discharge was produced at pressures in the order of magnitude of 200 Torr. The CSCCs emerge at a flat cathode separated at a distance of 250 micrometers from a ring shaped anode. Under such conditions, the local acceleration of electrons originated from the surface of the cathode at energies sufficient to produce excitations and ionizations in the cathode layer. The presence of excitation phenomena in this region is directly proved experimentally by the light emission observed in the cathode layer. The problem if ionization processes also take place in this region can be answered considering the static $I(V)$ characteristic presented in the same paper. This characteristic proves abrupt increase of $I$ for a critical value of $V$ fact related to the spontaneous self-assembling of an electric double layer similar to the double layer self-assembled in the next vicinity of the anode in usual dc gas discharges. For illustrating the presence of the same self-organization mechanism we present in Fig 3 the formation of spatial ordered patterns (anode spots) obtained from a dc Helium discharge at 20 Torr obtained between a heated electron emitting cathode and a plate anode placed at 10 cm one from the other. These photographs are similar to that published in [12]. Increasing the voltage applied on the anode, a little luminous anode sheet attached to the anode is firstly observed. When the voltage applied on the anode reaches a critical value, a well located nearly spherical very luminous space charge configuration (anode spot) appears in a certain point of the luminous sheet. It is accompanied by an abrupt increase of the current. The anode spot appears at a point of the anode where the current is concentrate owing to local causes. Such causes are, for example, the presence of protuberances on the anode surface or local gas emission. The local current increase involves an increase of the flux of the accelerated electrons so that locally the excitation and ionization rates increase at the critical values for which a CSCC self-assembles by self-organization [4-8]. Its appearance demonstrates that the dc gas discharge is able to create by self-organization CSCC that acts as new internal source of free carriers helping the external dc PS to maintain the high conductive state of the gaseous conductor [13]. When the voltage applied on the anode is further increased the dc gas discharge creates new CSCCs in the form of luminous anode spots attached to the anode. Since every of these anode spots are laterally bordered by a double layer the external side of which is negatively charged, repulsive electrostatic forces act between the spots so that they are symmetrically distributed on the surface of the anode. Their spontaneous emergence is accompanied by an abrupt increase of the current *I* collected by the anode. Returning to the self-organized luminous spots observed in the cathode layer of a microdischarge their appearance can be explained considering that, under the described experimental conditions, by bombardment with positive ions, the cold cathode emits electrons. These electrons with very small initial kinetic energy are accelerated in the cathode fall at energies able to produce atom excitations. By returning in the ground state the atoms emit photons explaining the light emission in this region. A part of them collide the surface of the cathode emitting photoelectrons. Their appearance increases the concentration of the current. Owing to the fluctuations of the light emission there exists the possibility of the local increase of the concentration of the electronic current so that in those points CSCC self-assembles by a self-organizing mechanism similar to that determining the self-assembling of the anode spots in usual dc gas discharges. Because every spot spontaneously self-assembled in the cathode sheet is surrounded by a double layer able to accelerate electrons at energies sufficient to produce ionizations it results that, by self-organization, the dc gas discharge is able to establish its discharge regime in which its existence is ensured by a minimal “effort” of the external dc PS [13].

Since every self-assemblage of a metastable CSCC is accompanied by the appearance of a new internal source of free carriers, the gaseous conductor works as a multifunctional system ably to transit in multiple conducting states after interaction with the surroundings, in our case, with the PS.

A more advanced self-organization state with respect to the formation of the CSCC in a metastable state is revealed by the gaseous conductor if, by increasing $V_{PS}$, $V$ reaches the value marked by **e** for which the current collected by the anode becomes periodically limited. As already shown [6,8], these current limitations are related to a detachment of the double layer from the border of the CSCC followed by its propagation through the plasma and de-aggregation at a certain distance from the region where it self-assembled. Since during the departure from the region where it self-assembled a new double layer self-assembles at the border of the CSCC, an internal acting positive feedback mechanism acts so that the detachment process of double layer from the border of the CSCC becomes a periodical one.

Decreasing $V_{PS}$, the periodic limitation of $I$ is maintained also for values of $V$ smaller than that required to produces the transition of the CSCC into a dynamical state in which double layers that propagate as moving patterns in the gaseous conductor periodically self-assembles at its border. This proves that the CSCC in a dynamical steady state was endowed with another kind of memory that appears after its transition into a dynamical state. This memory is emphasized by the hysteresis loop **e-f-h-i-j** on which a periodic exchange of matter and energy with the surroundings takes place. In this case the memory is related to the concomitant presence of two double layers, one in the moving phase and another one in the self-assembling phase. Since the existence of the moving double layer involves operations learned in its self-assembling process it is evidently that the memory related to the aforementioned hysteresis loop has the same origin as that of a metastable CSCC whose bistability is related to the hysteresis loop **c-d-j-k-c**.

As already shown, by increasing $V_{PS}$ after the abrupt increase of $I$, when $V$ reaches the value marked by **e** the gaseous conductor works as an N-shaped negative differential resistance [6,8]. Starting from the strong similarities emphasized between the non-linear behaviours of gaseous conductors and semiconductors it was proposed [14,15] to consider a scenario of self-organization similar to that which explains self-organization phenomena in gaseous conductors to be at the origin of the S-shaped and N-shaped, respectively, negative differential resistances of semiconductors. As known, the genuine origin of the two kinds of negative differential resistances proper to different kinds of semiconductors remained at the present date an enigma that requires elucidation.

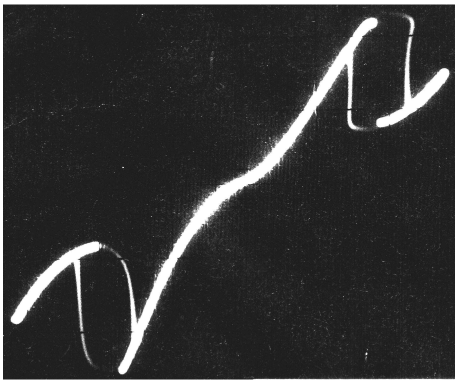

Fig. 4. Current-voltage characteristic of a special kind of semiconductors generating ultra-high frequencies [16].

It is interesting to remark that the *I(V)* characteristics obtained from so called quantum electronic devices [16] as that shown in Fig. 4 reveal hysteresis loops similar to that obtained from a gaseous conductor shown in Fig 1. This seems to prove that in every region where two different semi-conducting materials are joined, complex self-organized space charge configurations appear, very probable with planar shape. These are able to generate periodically moving double layers that, after a very short time span reach the distance at which they de-aggregate. Under such conditions, an internal feedback mechanism similar to that taking place in gaseous conductors can explain the working regime of semiconductors, implicitly that used for generating ultrahigh frequencies.

## 3. Conclusions

The ability of the so-called intelligent materials to work as multi-functional systems is related to the spontaneous self-assemblage of a CSCC endowed with memory. In gaseous conductors the intrinsic self-organization mechanism at the origin of the spontaneous emergence of a single CSCC is well know [4-9]. Thus, it was experimentally emphasized that depending on the distance from thermodynamic equilibrium imposed by the external constraint (the voltage applied on the conductor) the CSCC appears in a metastable state or in a state in which a periodic exchange of matter and energy with the surroundings ensures its existence by a proper dynamics of the double layer at its border.

The emergence of ordered or nearly-ordered distributed CSCCs at the surface of electrodes in usual dc gas discharge but, especially, in micro-discharges is, in our opinion, a strong argument to consider that a similar self-organization mechanism is able to determine the emergence of a network of CSCCs. Since apparently similar networks of metastable configurations in the form of chain-like objects at nanometer scale were observed also in non-crystalline solids [17], it is very probable that the self-organization process at their origin implicate the presence of electrostatic forces that, acting as long-range correlation, ensure their self-consistence and implicitly the high degree of its ordered or quasi-ordered distribution.

In quantum electronic devices as that used in miniaturized technology to generate oscillation with ultrahigh frequencies [16,18] the presence of a memory effect is proved by the hysteresis loops shown in Fig 4. Considering at its origin moving double layers similar to those observed in gaseous conductors, it results that the time-span in which these are self-assembling and de-aggregating are very short with respect to the same time-spans observed in gaseous conductors. The causes of these differences reside in the fact that the self-enhancing process of the production of positive ions, related to the self-assembling of the double layers develops in a time-span, that because of the high atom concentration in solids, is very small.

Taking into account the above said, we consider that the scenario of self-organization explaining the emergence of CSCC in gaseous conductors is potentially able to explain the behaviour of multifunctional systems as those used in micro and nano-technologies. The news of this scenario of self-organization consists in the fact that it describes the succession of physical processes able to explain the self-assembling of a single CSCC and implicitly of a network of CSCCs. Since every CSCC is endowed with memory it is, in principle, possible to explain also why a sample containing a network of CSCCs works as an intelligent material.

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