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Period doubling in glow discharges: local versus global differential conductivity

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ABSTRACT

Short planar glow discharges coupled to a resistive layer exhibit a wealth of spontaneous spatio-temporal patterns. Several authors have suggested effective reaction-diffusion-models to explore similarities with other pattern forming systems. To test these effective models, we here investigate the temporal oscillations of a glow discharge layer coupled to a linear resistor. We find an unexpected cascade of period doubling events. This shows that the inner structure of the discharge is more complex than can be described by a reaction-diffusion-model with negative differential conductivity.

Period doubling in glow discharges: local versus global differential conductivity

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Short planar glow discharges coupled to a resistive layer exhibit a wealth of spontaneous spatio-temporal patterns. Several authors have suggested effective reaction-diffusion-models to explore similarities with other pattern forming systems. To test these effective models, we here investigate the temporal oscillations of a glow discharge layer coupled to a linear resistor. We find an unexpected cascade of period doubling events. This shows that the inner structure of the discharge is more complex than can be described by a reaction-diffusion-model with negative differential conductivity.

Glow discharges are part of our daily environment in conventional and energy saving lamps, beamers, flat TV screens, car and street lamps as well as in various industrial applications. While applications typically try to avoid any instabilities, experiments actually exhibit a realm of spontaneous pattern formation, see, e.g., [1].

An interesting series of experiments has been performed on short planar dc driven glow discharges with wide lateral extension [2–13] where the formed patterns were explored very systematically. The observed structures resemble those observed in Rayleigh-Benard convection in flat cells, in electroconvection in nematic liquid crystals, or in various chemical or biological pattern forming systems. All these systems show the formation of stripes, spots, spirals etc. In comparison to the other systems, the glow discharge system has the advantage of particular convenient experimental handling and time scales [14]. Besides structures known from other physical systems, it continues to exhibit new structures that might be specific for this system [2–13]. We will focus on the experiment in [9], where a complete phase diagram of different patterns was identified: homogeneous stationary and homogeneous oscillating modes, patterns with spatial and spatio-temporal structures etc.

The theory for these systems has largely focussed on effective reaction-diffusion models in the two transversal directions, and on the negative differential conductivity of the glow discharge as the driving force of pattern formation. Such models actually have been developed independently by a number of authors [15–21,4]. On the other hand, the observation of unconventional patterns like zigzag-destabilized spirals raises doubts whether reaction-diffusion patterns are sufficient to understand the observed patterns.

In the present paper, we examine the concepts of reaction-diffusion models and negative differential conductivity on the particular case of a short DC driven glow discharge in a parameter range that exhibits spontaneous temporal oscillations but no spatial structures transverse to the current [9]. In short, we find (i) that a discharge on the transition from Townsend to glow discharge can combine a positive local differential conductivity with a negative global differential conductivity; (ii) that a glow discharge in a simple electric circuit shows more complex behavior than can be expected from the proposed reaction-diffusion models [15–21,4] for voltage U and current J with (global) negative differential conductivity $dU/dJ < 0$; (iii) in particular, that the system can show period doubling bifurcations. Period doubling actually has been observed experimentally in glow discharges, but in more complex geometries and in longer systems [22,23]. (iv) Finally, we derive a new effective dynamical model in terms of a parameter and a function by adiabatic elimination of the electrons. There is no systematic way to reduce this model to a simpler one [15–21,4] with two scalar parameters like voltage U and current J. We draw this conclusion both from direct analysis and from the occurence of period doubling in the numerical solutions.

To be precise, in the experiments of [2–13], a planar glow discharge layer with short length in the forward direction and wide lateral dimensions is coupled to a semiconductor layer with low conductivity. The whole structure is sandwiched between two planar electrodes to which a DC voltage U_t is applied. Theoretical predictions on how the different spatio-temporal patterns depend on the parameters of the gas discharge, hardly exist. In [16–18,4], an effective reaction-diffusion model in the two dimensions transversal to the current is proposed. Roughly, it consists of two nonlinear partial differential equations for the current J and the voltage U of the form

$$\partial_t U(x,y,t) = \mathcal{F}(U,J) \quad , \quad \partial_t J(x,y,t) = \mathcal{G}(U,J) , \quad (1)$$

where the nonlinear operators $\mathcal{F}$ and $\mathcal{G}$ contain spatial derivatives ∂_x, ∂_y and possibly also integral kernels. The model is of reactor-inhibitor form as studied extensively in the context of chemical and biological systems in the past decades. If applicable to gas discharges, this identification lays a connection to a realm of analytical and numerical results on reaction-diffusion systems.

To test whether a model like (1) is applicable to the gas discharge system, we will focus on its temporal oscillations that can occur in a spatially completely homogeneous mode [9]; hence a one-dimensional approximation

is appropriate. Similar oscillations have been observed in [19,20,24,25], and similar effective models for current J and voltage U of the general form (1) have been proposed in [15,19–21].

Why have different authors come up with the same type of model? The equation for U directly results from the simplest form of an external electric circuit: a semiconductor layer of thickness d_s, linear conductivity σ_s and dielectricity constant ϵ_s will evolve as

$$\partial_t U = \frac{U_t - U - R_s J}{T_s} \qquad (2)$$

where U_t is the voltage on the total system, J is the total current, and $U = \int_0^{d_g} E\, dz$ is the voltage over the gas discharge which is the electric field E integrated in the z direction over the height d_g of the discharge. For the experiments in [9], $R_s = d_s/\sigma_s$ is the resistance of the whole semiconductor layer, and $T_s = \epsilon_s \epsilon_0/\sigma_s = C_s R_s$ is the Maxwell relaxation time of the semiconductor. In other experimental systems, the quantities R_s and T_s can have different realizations. Hence the form of the equation for U in a reaction-diffusion model (1) is clear.

However, the equation for J in a reaction-diffusion model as (1) is based on guesses and plausibility. Different choices have been suggested by different authors, but one thing is clear: to be physically meaningful, the current-voltage characteristics of the glow discharge has to be a stationary solution, so $\mathcal{G}(U, J) = 0$ on the characteristics. Beyond that, there are different suggestions for the functional form of $\mathcal{G}$ and the intrinsic time scale.

If a model like (1) is applicable to oscillations in glow discharge systems, then the following predictions apply: 1) an oscillation can only occur in a region of negative differential conductivity of the glow discharge characteristics, 2) only a single period can be formed, period doubling is not possible, since this requires at least three independent parameters, 3) in a phase space plot in U and J, the trajectory of an oscillation can intersect the load line $U = U_t - R_s J$ only parallel to the J-axis (since $\partial_t U = 0$ and $\partial_t J \neq 0$), and it can intersect the characteristics of the glow discharge $U = U(J)$ only parallel to the U-axis (since $\partial_t U \neq 0$ and $\partial_t J = 0$).

We now introduce the simplest classical model for a glow discharge [26–28], solve it numerically and confront its results with the predictions above.

A discharge between Townsend and glow regime consists of a gas with Ohmic conductivity for the rare charged particles, electrostatic space charge effects and two ionization mechanisms, namely impact ionization by accelerated electrons in the bulk of the discharge (the so-called α-process) and secondary emission from the cathode (the γ-process). In its simplest form, it can be modelled by continuity equations for electron particle density n_e and ion particle density n_+

$$\partial_t\, n_e + \nabla \cdot \mathbf{J}_e = \mathcal{S} \quad , \quad \partial_t\, n_+ + \nabla \cdot \mathbf{J}_+ = \mathcal{S} \, , \qquad (3)$$

and the Poisson equation for the electric field $\mathbf{E}$ in electrostatic approximation,

$$\nabla \cdot \mathbf{E} = \frac{e}{\varepsilon_0}\, (n_+ - n_e) \quad , \quad \mathbf{E} = -\nabla \Phi \, . \qquad (4)$$

The particle currents are approximated as purely Ohmic

$$\mathbf{J}_e = -\mu_e\, n_e\, \mathbf{E} \quad , \quad \mathbf{J}_+ = \mu_+\, n_+\, \mathbf{E} \, . \qquad (5)$$

The source of particles in the continuity equation (3) is written as a sum of generation by impact ionization in Townsend approximation and recombination

$$\mathcal{S} = |n_e \mu_e E|\, \alpha_0\, e^{-E_0/|E|} - \beta n_e n_+ \, . \qquad (6)$$

Finally, the secondary emission from the cathode enters as a boundary condition at the position d_g of the cathode

$$\mu_e n_e(d_g, t) = \gamma \mu_+ n_+(d_g, t) \, . \qquad (7)$$

This is the classical glow discharge model [26–28].

We reduce the problem to one spatial dimension z transverse to the layers which is an excellent approximation for the experiment [9]. Furthermore, we introduce dimensionless quantities as in [28] by rescaling all parameters and fields as $z = r_z/X_0$, $\tau = t/t_0$, $L = d_g/X_0$, $\sigma(z, \tau) = n_e(r_z, t)/n_0$, $\rho = n_+/n_0$, $\mathcal{E} = E_z/E_0$ with the scales $X_0 = \alpha_0^{-1}$, $t_0 = (\alpha_0 \mu_e E_0)^{-1}$ and $n_0 = \epsilon_0 \alpha_0 E_0/e$. A key role is played by the small parameter $\mu = \mu_+/\mu_e$, which is the mobility ratio of ions and electrons.

The gas discharge layer is now modelled by

$$\partial_\tau \sigma = \partial_z(\mathcal{E}\sigma) + \sigma \mathcal{E} \alpha(\mathcal{E}) \quad , \quad \alpha(\mathcal{E}) = e^{-1/|\mathcal{E}|} \, , \qquad (8)$$

$$\partial_\tau \rho = -\mu \partial_z(\mathcal{E}\rho) + \sigma \mathcal{E} \alpha(\mathcal{E}) \, , \qquad (9)$$

$$\sigma(L, \tau) = \gamma \mu \rho(L, \tau) \, , \qquad (10)$$

$$\rho - \sigma = \partial_z \mathcal{E} \, , \qquad (11)$$

where recombination was neglected [$\beta = 0$ in (6), a discussion of this approximation follows below], while the external circuit is described by

$$\partial_\tau \mathcal{U} = \frac{\mathcal{U}_t - \mathcal{U} - \mathcal{R}_s j}{\tau_s} \quad , \quad \mathcal{U}(\tau) = \int_0^L \mathcal{E}(z, \tau)\, dz \qquad (12)$$

with the dimensionless voltage $\mathcal{U} = U/(E_0 X_0)$, time scale $\tau_s = T_s/t_0$ and resistance $\mathcal{R}_s = R_s/R_0$, $R_0 = X_0/(e\mu_e n_0)$ and with a spatially conserved total current

$$j(\tau) = \partial_\tau \mathcal{E} + \mu \rho \mathcal{E} + \sigma \mathcal{E} \quad , \quad \partial_z j(\tau) = 0 \, , \qquad (13)$$

where $\partial_z j = 0$ follows from (8), (9) and (11) as usual.

As a result, the gas discharge is parametrized by the three dimensionless parameters of system length over ionization length L, secondary emission coefficient γ and mobility ratio μ (as discussed in [28]), and the external

circuit is parametrized by relative resistance $\mathcal{R}_s$, ratio of time scales τ_s and dimensionless applied voltage $\mathcal{U}_t$.

For calculational purposes, the ion density ρ can be completely eliminated from the one-dimensional gas discharge equations (8)–(11) with the help of the Poisson equation (11) and the total current j, see [28]. The result are two equations of motion for $\partial_\tau \sigma$ and $\partial_\tau \mathcal{E}$. In our numerical calculations, the system was implemented in this form. Our choice of parameters was guided by the experiments in [9]: we chose the secondary emission coefficient $\gamma = 0.08$, the mobility ratio $\mu = 0.0035$ for nitrogen and the dimensionless system size $L = 50$ which amounts to 1.4 mm at a pressure of 40 mbar. The external circuit has $\mathcal{R}_s = 30597$, $\tau_s = 7435$ and a dimensionless total voltage $\mathcal{U}_t$ in the range between 18 and 20. This corresponds to a GaAs layer with $\epsilon_s = 13.1$, conductivity $\sigma_s = (2.6 \cdot 10^5 \Omega\text{cm})^{-1}$ and thickness $d_s = 1.5$mm, and a voltage range between 513 and 570 V.
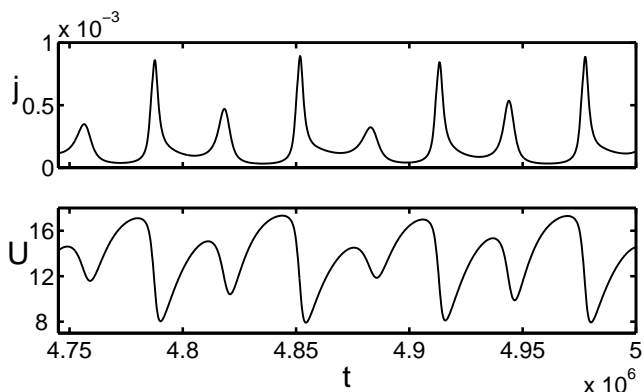

FIG. 1. Spontaneous oscillations of current j and voltage $\mathcal{U}$ as a function of time τ for $\gamma = 0.08$, $\mu = 0.0035$, $L = 50$, $\mathcal{R}_s = 30597$, $\tau_s = 7435$, and applied total voltage $\mathcal{U}_t = 19$.

Fig. 1 shows electric current j and voltage on the gas discharge $\mathcal{U}$ as a function of time for a total stationary voltage $\mathcal{U}_t = 19$ applied to the complete system of gas discharge and semiconductor layer. The system exhibits spontaneous oscillations with sharp current peaks: when the voltage $\mathcal{U}$ on the gas layer becomes high enough, the discharge ignites. The conductivity of the gas increases rapidly and produces a current pulse that deposits a surface charge on the gas-semiconductor interface. Therefore the voltage $\mathcal{U}$ over the gas layer breaks down. Due to the low conductivity of the semiconductor, the voltage $\mathcal{U}$ recovers only slowly. Eventually the gas dicharge ignites again, and the cycle is repeated.

Note that the oscillations in Fig. 1 are not quite periodic. This is not due initial transients since the system is observed after the long relaxation time $\tau = 4.745 \cdot 10^6$. The nature of this temporal structure becomes clear when the trajectory is plotted in the plane spanned by current j and voltage $\mathcal{U}$ in Fig. 2(b). The figure contains the data of the time span from $\tau = 3 \cdot 10^6$ to $6 \cdot 10^6$

which amounts to approximately 90 current pulses. The phase space plot shows that the system is actually periodic, with a period of 8 current pulses. Fig. 1 shows precisely one period.

This discovery raises the question whether our system actually follows the well-known scenario of period doubling. Indeed, it does. Fig. 2(a) for $\mathcal{U}_t = 18$ shows an oscillation where one current pulse is repeated periodically as observed experimentally in [9]. For $\mathcal{U}_t = 18.5$, a period consists of two current pulses (not shown). For $\mathcal{U}_t = 19$, the period is 8 pulses as in Fig. 1 and Fig. 2(b). For $\mathcal{U}_t = 20$, the systems seems to have reached the chaotic state as can be seen in Fig. 2(c).

A detailed comparison of the experiments in [9] with simple oscillations as in Fig. 2(a) will be given elsewhere, and we only state here that there is semi-quantitative agreement of several features. Here we emphasize that period doubling events in glow discharges have been observed experimentally in other systems [22,23]. However, this was always in systems with more complicated geometries like long narrow tubes, and the authors allude to general knowledge on nonlinear dynamics rather than to solutions of explicit models. We state that period doubling can be a generic feature of a simple, strictly one-dimensional glow discharge when coupled to the simple circuit (2). We propose to search experimentally for a period doubling route to chaos in such simple systems which would then allow quantitative comparison with theory.

Let us return to the initial question: is a 2-component reaction diffusion model like (1) with negative differential conductivity appropriate for the present system? Above Eq. (3), we gave a list of predictions for the reaction diffusion model (1) to be applicable. Prediction 2 is falsified by the observation of period doubling. Prediction 3 is also falsified by a simple check of either of the three figures in Fig. 2: the trajectories definitely do not intersect with the characteristics or the load line with the angle prescribed by (1), in particular not in the upper part of the figures that represent the rapid current pulses.

There rests prediction 1: is negative differential conductivity required for the oscillations to occur? We have not found a numerical counterexample where oscillations would occur while the current voltage characteristics of the gas discharge shows a positive differential conductivity, but we have found no reason to exclude its existence. Furthermore we note that the characteristics is a global property of the whole discharge layer with its boundary conditions [28] while the local differential conductivity in our model is always positive: the field dependent stationary ionization is $n_+ = |\mu_e E| \alpha_0 e^{-E_0/|E|} / \beta$ according to (6); hence the local conductivity increases monotonically with the applied field $|E|$. The global negative differential conductivity is due to electrode effects being much stronger than bulk recombination β.

Last but not least, we have derived an analytical approximation of the model (8)–(13) that can be confronted

with the suggested form (1). Electron and ion current in the gas are of the same order of magnitude. Since the electrons are much more mobile, their density is appropriately lower. Rescaling this density like $s = \sigma/\mu$ and time like $\bar{\tau} = \mu\tau$, the electrons can be eliminated adiabatically in the limit of $\mu \to 0$. Space charges in the gas discharge are then due to the ions only $\rho = \partial_z \mathcal{E}$, and ρ can be expressed by $\mathcal{E}$. Splitting the field $\mathcal{E}(z,t) = \mathcal{E}_L(t) + \epsilon(z,t)$ into the field on the cathode $\mathcal{E}_L$ and a correction ϵ with $\epsilon(L,t) = 0$, the complete system for $\mu \to 0$ can be expressed by two dynamic equations

$$\partial_{\bar{\tau}} \mathcal{E}_L(t) = F(\mathcal{E}_L, \epsilon) \quad , \quad \partial_{\bar{\tau}} \epsilon(z,t) = G(\mathcal{E}_L, \epsilon) , \qquad (14)$$

details will be given elsewhere. While the equation for the time dependent parameter $\mathcal{E}_L$ corresponds to the equation for U in (1), the space dependent field correction ϵ within the gas layer cannot be reduced to a single component like the current J in (1). E.g., for the ion density on the cathode $\rho_L = \partial_z \epsilon|_L$, we can derive the equation of motion

$$\partial_{\bar{\tau}} \rho_L = -\rho_L^2 - \mathcal{E}_L(\partial_z \rho)|_L + \gamma \rho_L \mathcal{E}_L \alpha(\mathcal{E}_L) \qquad (15)$$

A two component reaction-diffusion equation for ρ_L and $\mathcal{E}_L$ could result from the completely unsystematic approximation $(\partial_z \rho)|_L = 0$. Rather the transport of ions ρ from the bulk of the gas towards the cathode is a central feature of the system. The field $\epsilon(z,t)$ in (14) indeed accounts for the ion distribution within the gas gap with its intricate dynamics.

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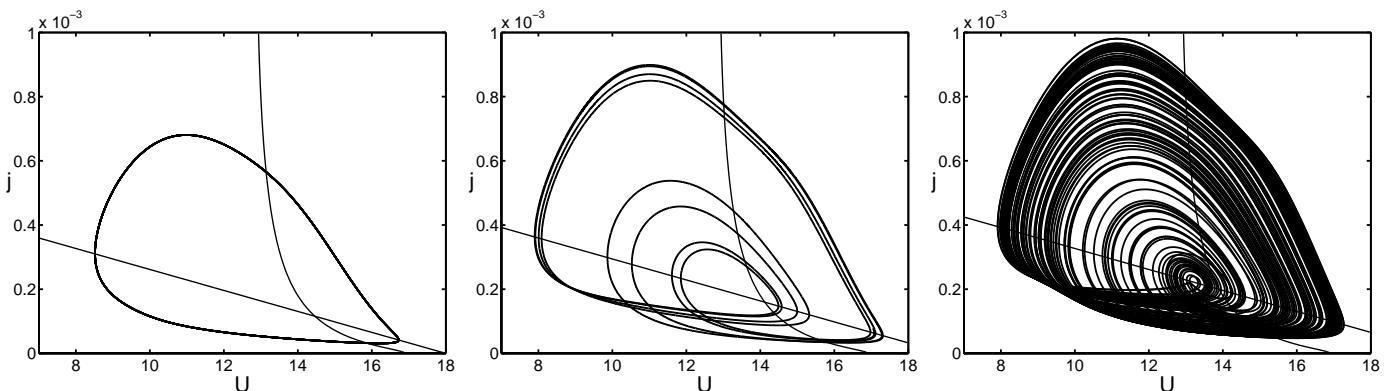[28] D.D. Šijačić, U. Ebert, Phys. Rev. E **66**, 066410 (2002).

FIG. 2. Phase space plots of the trajectories of the oscillations in the plane of current j and voltage $\mathcal{U}$. The time range is $3 \cdot 10^6 \leq \tau \leq 6 \cdot 10^6$ in all figures. Shown are the orbits, the straight load line $\mathcal{U} = \mathcal{U}_t - \mathcal{R}_s j$ and the curved current voltage characteristics $\mathcal{U} = \mathcal{U}(j)$ of the gas discharge [28]. The intersection of load line and characteristics marks the stationary solution of the system. (b) represents the data of Fig. 1 with total voltage $\mathcal{U}_t = 19$, (a) is for $\mathcal{U}_t = 18$, (c) for $\mathcal{U}_t = 20$.