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CHEMICAL TEMPORAL AND SPATIAL STRUCTURES IN STRONG MAGNETIC FIELD

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Temporal and spatial structures in the Belousov-Zhabotinskii systems in constant electric and strong magnetic fields are studied in this paper. Unlike a random generation of chemical wave center in a system without the influence of an electromagnetic field we have observed that only one wave forms in the presence of electric and magnetic fields of suitable intensity and induction. Its speed of propagation depends on the connected fields. A theory of this phenomenon has been developed and the obtained results are in a good agreement with experimentally measured dependences of the temporal period of chemical waves on electric field intensity and magnetic field induction.

Chemical temporal and spatial structures serve well as models for explaining different properties of living systems in different stages of organization¹⁻⁵. As a suitable reaction system for experimental observation the Belousov-Zhabotinskii system can be used^{6,7} enabling visual observation. We know that different local inhomogeneities can serve as a stimulator for spatial wave generation in temporal oscillating systems. The inhomogeneities arise in the system randomly, therefore the generation of these waves is also a random process. This property of structures is used, e.g. in papers^{8,9}, to explain the generation of heart arythmicity and fibrillation. We have studied the possibility of preventing this randomness by an external influence on the system and regulating the characteristic parameters of the observed structures by means of external factors. Objects responsible for observed generation of structures are the ions and so there may be a possibility to use the electric and magnetic fields as an external regulator. The influence of electric field on chemical dissipative structures was studied by many authors. Kondepudi in^{10,11} and together with Prigogine in¹², among others, showed that the reaction of nonlinear chemical oscillating systems to external electric field may appear by the arising of "propagating bands". Schmidt and Ortoleva found, by means of theoretical analysis of interaction of chemical waves with electric field in¹³⁻¹⁵ that as a result of it many nonlinear effects may arise, especially in changes of the wave transport speed and the amplitude. The review of these effects can be found in¹⁶.

Marek and Ševčíková have found in^{17,18} that the wave propagation speed increases with the intensity of the connected field at one polarity and in turn it decreases at the opposite polarity. In the second case at a certain critical intensity the speed of wave shift becomes equal to zero.

The influence of relatively very weak magnetic field on chemical oscillation amplitudes was studied by Agulova and Opalinskaya in the paper¹⁹. They found that this influence is not very significant (the amplitude changes are of a few per cent). Our stimulus for application of electric and magnetic fields to chemical oscillations was the assumption that in this case the possibilities of random local generation of centers of spatial structuralization could be suppressed. It was observed that in an electric and sufficiently strong magnetic fields only one plane perturbation wave was developed which started from the site of the strongest field, i.e. from the electrodes, and propagated in the system with a definite speed. Equally it was found that characteristic parameters, i.e. temporal period, wavelength, wave speed were influenced by electric and magnetic fields. These interesting results were observed by applying relatively strong magnetic field of the induction between 1 and 3 Tesla at relatively low intensities of the electric field (<70 V/m).

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EXPERIMENTAL

We have used about 8.5 ml of the Belousov-Zhabotinskii reaction mixture²⁰. In all cases the experiments were arranged in such a way that this mixture formed a thin horizontal layer (depth ≈ 2 mm) between two vertical platinum parallel plate electrodes (48 mm in the distance) placed in Petri dish of inner diametér 70 mm. The whole experimental apparatus was placed in a relatively strong magnetic field formed by the cavity of Helmholtz coils 20 cm in inner diameter. The field in the cavity could be estimated as homogeneous while the direction of the vector of magnetic induction was perpendicular to the vector of electric intensity. The imposed electric field was of the intensity in the range of $0 \div 62.5$ V/m obtained from undirectional stabilized voltage source. The imposed stationary magnetic field was of the induction in the range of $0 \div 3$ T induced by the current flowing through the superconductive material of Helmholtz coils.

The reaction mixture used consisted of $0.375 \text{M} \text{H}_2\text{SO}_4$, $0.347 \text{M} \text{NaBrO}_3$, 0.057 M NaBr, 0.113 M malonic acid and $2.94 \cdot 10^{-3} \text{M}$ ferroin. The stock solution composed of H_2SO_4 and NaBrO_3 was prepared in advance. Just before the experiment started it was mixed with NaBr solution (Br₂ was formed) and malonic acid was added (the brom-malonic acid was formed). When all formed Br₂ was consumed (2 min after addition of malonic acid) ferroin was added²¹. Immediately after adding ferroin the reaction mixture was introduced into Petri dish with electrodes and centered in the Helmholtz coils. After the first wave began to propagate from the electrode in the solution (see ref.²¹) we started to record the frequency of the oscillations potentiometrically by a double-track recorder (the part of the equipment used for measuring was made from nonmagnetic materials not to influence the imposed magnetic field). The temperature in the center of the coils and the temperature of the reaction mixture were measured repeatedly during the experiment and they did not change considerably (they were approximately of the value 285 K).

RESULTS

One-Dimensional Theoretical Model – Plane Electrodes

The experimental results we have observed, can be interpreted qualitatively as follows: we have found by measuring that the highest potential gradient of the connected electric field is in the surrounding of electrodes. Hence, in accordance with the results of Kondepudi and Prigogine¹² we can suppose that here, in these regions, are favourable conditions for formation of "propagating bands". The connected magnetic field evokes mixing in a certain way and so it inhibits the arise of further random local perturbations. Hence, the perturbation generated at one electrode propagates, practically unchanged, toward the opposite electrode (Fig. 1*a*, *b*). Here we have observed the remarkable fact, that this perturbation is spatially structuralized and that its characteristics, mainly the frequency, depend on the intensity of electric field and the induction of magnetic field.



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Fig. 1

Chemical temporal and spatial structures a without electric and magnetic fields, b in electric ($E = 52 \cdot 1 \text{ V/m}$) and magnetic (B = 2 T) fields

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In the theoretical section we will try to elaborate a phenomenological theory to explain the observed effects. Similarly as Kondepudi we aim to study the influence of external fields on already existing structures and, therefore, we will not deal with examining the conditions of bifurcation arise. This problem is studied in detail, e.g. in the paper of Livshits²². The above mentioned papers deal in the theoretical part only with the influence of an electric field on chemical dissipative structures, mainly on the propagation speed of chemical waves. It was necessary to include into the formalism of the calculation the influence of magnetic field and to find the possibility of deriving the dependence of frequency and wavelength of the observed perturbation on the intensity of electric and on the induction of magnetic fields.

We will use several approximations – their convenience is supported mainly by the fact that our obtained results – when considering only the influence of electric field – as it will be presented in the conclusion of this paper – are in accordance with the results obtained by other authors and by other methods (see, e.g. ref.¹⁶) only when electric field was used. Our method – similarly as most of the mentioned authors – is based on the assumption that we have to do with a two-component autocatalytical reaction system.

The dynamics of chemical system is determined in general by its own mechanism of "chemistry", molecule diffusion of its individual components and their movement initiated by external fields. The evolution equations of the two-component system can be formulated on the base of the general continuity equation in the form

$$\oint (\mathbf{j}_d + \mathbf{j}_v) \cdot d\mathbf{S} = - \int (\partial n/\partial t) \, dV, \qquad (1)$$

where $\mathbf{j}_d = -D \nabla n$ is a diffusion flow, D being the diffusion coefficient, $\mathbf{j}_v = \mathbf{n} \cdot \mathbf{v}$ is a flow induced by external fields, n is concentration, dS is a surface element and dV is a volume element. They are of the form

$$\partial n_1 / \partial t = F_1(n_1, n_2) + D_1 \Delta n_1 - \nabla \cdot (n_1 \mathbf{v}_1)$$
⁽²⁾

$$\partial n_2 | \partial t = F_2(n_1, n_2) + D_2 \Delta n_2 - \nabla \cdot (n_2 \mathbf{v}_2), \qquad (3)$$

where the functions F_1 , F_2 determine the chemical mechanism and Δ is the Laplace operator. Microscopic drift of electrical charge appears in the investigated system at the presence of electric and magnetic fields. Electric current and, in very strong rmagnetic fields, also macroscopically observable mechanical movement is observed in the system. The speed \mathbf{v} can be therefore generally expressed as the sum of drift (\mathbf{v}_d) and mechanical (\mathbf{v}_m) components, i.e. $\mathbf{v} = \mathbf{v}_d + \mathbf{v}_m$. The drift component of the speed is expressed as the product of mobility and electric field intensity \mathbf{E} , i.e. $\mathbf{v}_d = \mu \mathbf{E}$.

Our measurement consisted in recording the structure of perturbation propagating in the direction perpendicular to the plane electrodes. Let it be the direction of the x-axis, so the components of ion speed in this direction are $v_{dx} = \mu E_x$. It is simple to prove that in the magnetic field with the induction oriented perpendicularly to the x-axis $(B \equiv B_z)$ an effective diminishing of this transport speed will follow in the relation $g = (1 - kB^2)$, where k is the constant. The free electrically charged particle would move in magnetic field in the circle with the radius r, derived from the equation $qvB = mv^2/r$, where q is the charge.

In Fig. 2 we can see that instead of the real course s_0 the particle in the presence of magnetic field will follow in the direction of the observed transport (x-axis) only the course

$$s' = r \sin \alpha = r \sin (s_0/r)$$

If we suppose $s_0 \ll r$, what is in our case always fulfilled, we can use the approximation for small angles, so that

$$s' \approx r(s_0/r - s_0^3/6r^3) = s_0(1 - s_0^2/6r^2).$$

Since $1/r \sim B$ we really obtain the relation $s' \approx s_0(1 - k B^2)$. Also in the case of very strong fields $kB^2 \ll 1$ (so it is always in our case because of the low mobility of the ions) and this term can be written in "renormalized" form $s' \approx s_0 \exp(-kB^2)$. The advantage of this form is that for the theoretical limit $B \to \infty$ it gives "reasonable" physical result, i.e. $s' \to 0$. In this case the ion speed, being in the direction of the x-axis, in the presence of electric field and magnetic field which are perpendicular to it, can be written in the form $(E_x = E; B_z = B)$

$$v_x = \mu_0 E \exp(-kB^2) + v_{mx},$$
 (4)

where μ_0 corresponds to the mobility in zero magnetic field. Substituting this in Eqs (2) and (3), considering the ions with index 1 are negative (this choice is in



Fig. 2

Diminishing of ion transport speed in magnetic field perpendicular to the direction of propagation accordance with obtained experimental results) and by substituting the Maxwell equation in the form

$$\mathrm{d}E/\mathrm{d}x = \varrho/\varepsilon = q(n_2 - n_1)/\varepsilon$$

we obtain them in the form

$$\partial n_1 / \partial t = F_1(n_1, n_2) + D_1 \partial^2 n_1 / \partial x^2 + \left[\mu_{01} E \exp\left(-k_1 B^2\right) + v_{1m} \right] \partial n_1 / \partial x + + \left(\mu_{01} q / \varepsilon \right) \exp\left(-k_1 B^2\right) \left(n_2 - n_1 \right) n_1 , \qquad (5)$$

$$\partial n_2 / \partial t = F_2(n_1, n_2) + D_2 \partial^2 n_2 / \partial x^2 - \left[\mu_{02} E \exp\left(-k_2 B^2 \right) + v_{2m} \right] \partial n_2 / \partial x - \left(\mu_{02} q / \epsilon \right) \exp\left(-k_2 B^2 \right) (n_2 - n_1) n_2 , \qquad (6)$$

where v_{1m} and v_{2m} are the components of mechanical speed in the direction of the x-axis.

In these equations the intensity of electric field is represented by the sum of the internal and external intensity, but using solutions with high electric conductivity, what concerns also our observed systems, this internal component in this expression can be neglected as the internal fields are already relatively weak (see, e.g. ref.¹⁶).

When solving the Eqs (5) and (6) we use the known "slaving principle" and the analysis by means of small deviation method (deviation from stationary concentration values $y = n - n^*$). Using the Galilei transformation z = t + x/u, where u is the perturbation propagation speed, we prove that in the solution temporally and spatially structuralized perturbation expressed by the function (see Appendix) can propagate

$$y(z) = C \exp(-\alpha z) \sin(\omega z + \varphi), \qquad (7)$$

where C and φ are integrating constants,

$$\alpha = (u/2D) \left[\mu_0 E \exp(-kB^2) - u \right],$$

$$\omega = u \{ a/D + (2\mu_0 q n^* | \varepsilon D) \exp(-kB^2) - [\mu_0 E \exp(-kB^2) - u]^2 / 4D^2 \}^{1/2}$$
(8)

and a is the constant characterizing here the chemism of the solution (see Appendix) and all characteristic quantities are related to ions which are dominant. That's why we omit the competent indices of individual symbols.

Regarding to the relation $n_1 = n_1^* + y$ both constants C and φ can be determined by two boundary conditions; i.e. $n_1(0, t_0) = n_{10}$ and $n_1(d, t_0) = n_{1d}$, where d is the distance between plane electrodes. We are not interested in this calculation because our aim is to find the dependence of fundamental wave characteristics (the frequency $v = \omega/2\pi$ and the wavelength $\lambda = u/v$) on the electric intensity and magnetic induction. Equation (8) and the known equation $u = v\lambda$ are two equations for three unknown characteristic structural parameters: u, λ and v. It would be possible to obtain the third equation from the expression (7). When enlarging the argument ωz by 2π , i.e. enlarging z by $2\pi/\omega$, then sin $[(\omega z + 2\pi) + \varphi] = \sin(\omega z + \varphi)$ and the quotient of the two deviations y_1 , y_2 corresponding to these states fulfilles the relation

$$y_1/y_2 = \exp\left(2\pi\alpha/\omega\right) \tag{9}$$

and we become the equation

$$\lambda \left[\mu_0 E \exp\left(-kB^2 \right) - u \right] = 2\delta D ; \quad \delta = \ln\left(y_1 / y_2 \right). \tag{10}$$

This equation enables to express explicitly the wavelength of the spatial structure by the relation

$$\lambda = \{ [(\delta^2 + 4\pi^2) D] / [a - (2\mu_0 q n^* / \varepsilon) \exp(-kB^2)] \}^{1/2} .$$
 (11)

It follows from this relation that in a steady regime, i.e. when $\delta^2 \ll 4\pi^2$, the dependence of the wavelength on the magnetic field induction can be expressed in the form

$$\lambda = \lambda_{\infty} \left[1 - \left(2\mu_0 q n^* / a \varepsilon \right) \exp\left(-kB^2 \right) \right]^{-1/2}, \qquad (12)$$

where λ_{∞} corresponds to λ for $B \to \infty$. We can state that the electric field does not influence the wavelength of the spatial structure.

Inserting the relation (12) into the Eq. (8) we get the formula for the frequency of the chemical perturbation wave in electric and strong magnetic field

$$v = (2/\lambda_{\infty}) \left[1 - (2\mu_0 q n^*/a\varepsilon) \exp(-kB^2) \right]^{1/2} \left\{ \left[(aD - 4\pi^2 D^2/\lambda_{\infty}^2) + (2\mu_0 q n^*D/\varepsilon - 8\pi^2 D^2\mu_0 q n^*/a\varepsilon\lambda_{\infty}^2) \exp(-kB^2) \right]^{1/2} + \left[(\mu_0 E/2) \exp(-kB^2) \right] \right\}.$$
(13)

This formula can be written in the more convenient form for an experimental verification

$$v = v_{\infty} [1 - L \exp(-kB^2)]^{1/2} + cE \exp(-kB^2) \}, \qquad (14)$$

where v_{∞} corresponds to v for $B \to \infty$ and L, b, c are constants.

Radial-Symmetric Electrodes

It is easy to realize the experiment with radial-symmetric electrodes. It follows from the Eq. (A8) (see Appendix) that if the perturbation is observed in the distance $r \gg G/a^*$ the term G/r in the Eq. (A8) can be neglected and now this equation becomes

identical with the equation (A3), which was solved in the previous section. It follows from this analysis that all results found for plane electrodes are valid also in the case of radial-symmetric electrodes, only the perturbation function here is multiplied by the factor 1/r. However, the dependence (13) is valid only in the case when the fraction y_1/y_2 is measured in the same distance r.

DISCUSSION

In our experiments a development of only one structuralized perturbation wave was really observed at the presence of electric and sufficiently strong magnetic fields in accordance with our expectation and the results of the presented theory. This result is demonstrated in Fig. 1. Fig. 1*a* corresponds to the situation without electric and magnetic fields, Fig. 1*b* illustrates the structure in an electric field (E = 52.1 V/m) with plane electrodes and magnetic field (B = 2 T). The structuralized perturbation starts to develop from the electrodes in the form of a plane or a little deformed plane boundary. In the case with radial-symmetric electrodes one radial-symmetric wave perturbation was observed. The process repeated periodically in time.

For the verification of the dependence (13), resp. (14) we have made two series of measurements – one for plane electrodes and second one for radial-symmetric electrodes. In the first case magnetic fields of the induction B = 1.5 T, 2 T, 2.5 T and 3 T were applied, in the second case the measurement started with magnetic induction B = 1 T. The measurement was realized at the presence of electric field with applied voltages U = 2 V, resp. U = -2 V in the first case and with voltages U = -0.2 V, -2 V, -3 V in the second case. The aim of the application of these two groups of electric intensity was to prove the dependence (14) in a monotonously increased electric field and in the presence of voltages of opposite signs.

Results of the measurements are presented in Fig. 3 (for plane electrodes) and in Fig. 4 (for radial-symmetric electrodes). The measured frequences were calculated on the basis of potentiometric data. For the interpretation of measured curves the values of constants L, b, c in the relation (14) must be known. It is possible to find approximately only the value of constant L. It is identical with $2\mu_0 qn^*/a\epsilon$ in the relation (12). The fraction $\lambda_{\infty}/\lambda_0 = \lambda_{B\to\infty}/\lambda_{B\to0}$ according to this relation is

$$\lambda_{\infty}/\lambda_0 = (1-L)^{1/2}$$

It follows from the observation that the value of this fraction is approximately 4, so we chose $L \approx -20$. The constant c is determined by the mobility of ions and the constant b is given by many other physical and chemical characteristics. We have calculated series of theoretical curves according to the relation (14) for constants L = -20, b = -1 and -0.075 < cE < 0.075 (Fig. 5) and for L = -20, b = -0.6 and -0.3 < cE < 0.075 (Fig. 6). We can see that the correlation between them and

the experimental curves (Figs 3 and 4) is very good. It is also possible to construct dependences v = f(E) for various values of the magnetic induction from curves in Fig. 4. These dependences are approximately straight lines what is also in agreement with the theoretical formula (13).

If we rewrite the formula (13) only for electric field, we get the relation

$$v = A(1 - J\mu_0 E/\lambda_0 + K),$$

where A and K are constants. This result is in principle the same as it was found by $Ortoleva^{16}$ for "high conductivity media".



FIG. 3

Dependence of the quantity 100/T on magnetic induction B for plane electrodes. (1/T = -v)



Fig. 5

Theoretical curves according to the function (14) for L = -20, b = -1 and -0.075 < cE < 0.075. $x \sim B$, $y \sim 1/T$





Dependence of the quantity 100/T on magnetic induction *B* for radial-symmetric electrodes





Theoretical curves according to the function (14) for L = -20, b = -0.6 and -0.3 < cE < 0.075. $x \sim B$, $y \sim 1/T$

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APPENDIX

The well known "slaving principle" is very often used in solving the equations of the types (5) and (6) (see, e.g. ref.²³). Let us suppose the negative ions to be dominant in the development of structures and we put in Eq. (6) $\partial n_2/\partial t \rightarrow 0$. This expresses the fact that positive ions adapt "adiabatically" to the situation formed by negative ions. When trying to find the relation between the concentrations n_1 and n_2 we remain only by balancing the homogeneous state $\partial n_2/\partial x \rightarrow 0$ without the external fields and for this purpose it is sufficient to solve the equation $F_2(n_1, n_2) = 0$. For simplicity we use the brusselator model described by the equations

$$F_1(n_1, n_2) = a_0 - a_1 n_1 + a_2 n_1^2 n_2 ,$$

$$F_2(n_1, n_2) = b_1 n_1 - b_2 n_1^2 n_2 .$$

From equation $F_2(n_1, n_2) = 0$ it follows the relation $n_2 = b_1/b_2n_1 = b'/n_1$, where b' is the characteristic constant.

It is to be noted that the choice of a suitable model of "chemism" is not important from the point of view of our aim. In the analysis by means of small deviation method all competent expressions are reduced only to an expression comprising the constants and the first power of the deviation. It is important only to know these constants and in our assumptions we consider them as given. The attempts to use a more adequate model of "oregonator" (see refs^{13,14}) were not successful and the authors had to modify even this model.

After these simplifications the Eq. (5) acquires the form

$$\partial n_1 / \partial t = F_1'(n_1) + D_1 \partial^2 n_1 / \partial x^2 + \left[\mu_{01} E \exp\left(-k_1 B^2 \right) + v_{1m} \right] \partial n_1 / \partial x , \quad (A1)$$

where

$$F'_{1}(n_{1}) = a_{0} + an_{1} + (\mu_{01}q/\varepsilon) \exp(-k_{1}B^{2})(b' - n_{1}^{2}); \quad a = a_{2}b' - a_{1}.$$

Now we will use the formalism of small deviation method using the relation $n_1 = n_1^* + y$, where n_1^* is a stationary value of the concentration n_1 . We obtain the equation

$$\partial y/\partial t = D_1 \partial^2 y/\partial x^2 + G \partial y/\partial x + a'y,$$
 (A2)

where

$$G = \mu_{01} E \exp(-k_1 B^2) + v_{1m},$$

$$a' = a - (2\mu_{01}qn_1^*/\epsilon) \exp(-k_1 B^2).$$

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It is easy to prove that the Eq. (A2) describes the one-dimensional perturbation, i.e. the variable y is the function of argument z = t + x/u, where u is the propagation speed of the perturbation. Using this Galilei transformation we get the equation

$$d^{2}y/dz^{2} + 2\alpha \, dy/dz + \beta y = 0, \qquad (A3)$$

where

$$\alpha = u(G - u)/2D_1,$$

$$\beta = a'u^2/D_1.$$

Solution of Eq. (A3) is the function

$$y(z) = C_1 \exp(p_1 z) + C_2 \exp(p_2 z), \qquad (A4)$$

where the parameters p_1 , p_2 are determined by the relations

$$p_{1,2} = -\alpha \pm (\alpha^2 - \beta)^{1/2}$$
.

The condition for development of structuralization is

$$\alpha^2 - \beta < 0$$

i.e. the condition

$$\left[\mu_{01}E\exp\left(-k_{1}B^{2}\right)+v_{1m}-u\right]^{2}-4a'D_{1}<0.$$
 (A5)

From this it follows that at a sufficient value of mechanical speed v_{1m} this condition cannot be fulfilled and the oscillations do not start. In the following consideration we will suppose we are in an subcritical region and we omit the term v_{1m} . Then the angular frequency of oscillations is determined by the relation (8) and the function (A4) acquires, in the oscillating case, the form (7).

In the case of radial-symmetric electrodes which were also used in our experiments the Eq. (A2) is of the form

$$\partial y/\partial t = (D_1/r^2)(\partial/\partial r)(r^2\partial y/\partial r) + G\partial y/\partial r + a'y, \qquad (A6)$$

where r is the radius. Supposing now the perturbation in the form

$$y = r^{-1}f(t + r/u) = r^{-1}f(z),$$
 (A7)

we get the equation

$$\partial f/\partial z = (D_1/u^2) \,\partial^2 f/\partial z^2 + (G/u) \,\partial f/\partial z + (a' - G/r) f \,. \tag{A8}$$

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