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# Oscillatory instability in super-diffusive reaction – diffusion systems: Fractional amplitude and phase diffusion equations

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**Abstract** – Non-linear evolution of a reaction – super-diffusion system near a Hopf bifurcation is studied. Fractional analogues of the complex Ginzburg-Landau equation and Kuramoto-Sivashinsky equation are derived, and some of their analytical and numerical solutions are studied.

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**Introduction.** – It has been recently realised that in many random physical processes the conceptions of Gaussian distribution and Fickian diffusion are invalid. Many such processes can be described by models of sub- or super-diffusion, where the displacement moments of the corresponding random walk grow slower or faster than for normal diffusion, respectively. A typical example of *super-diffusion* is the enhanced transport in fluids, predicted for flows with velocity correlation functions slowly decaying in space or time [1]. A specific type of super-diffusion, the Lévy flight, has been reported in observations of transport in two-dimensional rotating flows [2] and in a freely decaying two-dimensional turbulent flow [3]. Other examples of super-diffusive transport include wave turbulence [4], non-local transport in plasma [5,6], transport in porous media [7], surfactant diffusion along polymer chains [8], cosmic-rays propagation [9], motion of animals [10–13]. A widely used description of super-diffusive transport relies on the continuous time random walk model with a power law asymptotics of the particle jump length distribution. In the case of Lévy flights this leads in the macroscopic limit to a diffusion equation with the Laplacian replaced by its *fractional power* [14–16].

An important problem is the influence of super-diffusion on processes with chemical reactions [17–20]. Normal reaction – diffusion systems exhibit different types of instability [21,22]. Profound understanding of pattern formation and spatio-temporal chaos in these systems was achieved through generic equations valid near the instability threshold, such as complex Ginzburg-Landau [23–25] and Kuramoto-Sivashinsky equations [1]. The evolution of instabilities in reaction – diffusion systems can be

accompanied by advection of components. For instance stirring, which changes the effective diffusion properties of species, is one of the means to control dynamical regimes in systems with chemical reactions [26,27], including those generated by instabilities in reaction – diffusion systems [28–33]. Thus, one can expect that in some cases flows can give rise to an enhanced diffusion of reagents [34]. While studies of instabilities in systems with sub-diffusion have started (see [35] and references therein), super-diffusive reaction – diffusion systems are still unexplored, with the exception of the front propagation phenomenon, which is strongly influenced by fluctuations [17–19]. In this letter, weakly non-linear dynamics of a reaction – diffusion system characterised by Lévy flights [36] near a long wave bifurcation point is investigated.

**Fractional Ginzburg-Landau equation.** – Consider a two-component reaction – diffusion system in the general case of distinct anomaly exponents:

$$\frac{\partial n_j}{\partial t} = d_j \mathfrak{D}_{|x|}^{\gamma_j} n_j + f_j(n_1, n_2), \quad j = \{1, 2\}, \quad (1)$$

where  $n_j$ ,  $d_j$  and  $f_j$  are the species concentrations, diffusion coefficients and general kinetic functions, respectively. The fractional operator of order  $1 < \gamma < 2$  is defined as [37]

$$\mathfrak{D}_{|x|}^{\gamma} n(x) = -\frac{\sec(\pi\gamma/2)}{2\Gamma(2-\gamma)} \frac{\partial^2}{\partial x^2} \int_{-\infty}^{\infty} \frac{n(\zeta)}{|x-\zeta|^{\gamma-1}} d\zeta. \quad (2)$$

The equivalent definition in Fourier space allows for a simple generalisation of the operator to higher spatial dimensions:

$$\mathfrak{D}_{|x|}^{\gamma} e^{i\mathbf{q}\cdot\mathbf{x}} = -|\mathbf{q}|^{\gamma} e^{i\mathbf{q}\cdot\mathbf{x}}, \quad (3)$$

also making evident the virtue of the rotational invariance of (2), as opposed to the basic fractional derivative [38].

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This definition serves as a good continuous (macroscopic) model of a random walk describing Lévy flights, as it correctly captures the asymptotic behaviour of a particle probability to perform long jumps. Equation (1) proposes a suitable description of a reactive system, where the medium enables enhanced diffusion of Lévy flight type, while each species' exponent is determined by its own mobility properties, for example, a reaction – diffusion process in a disordered flow with long tail spatial correlation function [1,16] or on a catalyst surface, where long jumps of the reacting particles are permitted through the bulk phase with a turbulent gas flow.

Suppose that there exists a homogeneous steady state  $\mathbf{n}_0$  satisfying  $\mathbf{f}(\mathbf{n}_0) = \mathbf{0}$ . A vanishing trace of the sensitivity matrix,  $(\nabla \mathbf{f})_{jk} = \partial f_j / \partial n_k$ ,  $j, k \in \{1, 2\}$ , leads to a Hopf bifurcation at the long wave limit  $q = 0$ . Take  $\varepsilon \ll 1$  and  $0 < \mu \sim O(1)$  so that  $\text{tr} \nabla \mathbf{f}|_{\mathbf{n}_0} = \varepsilon^2 \mu$  and invoke a multiple scales analysis with

$$\mathbf{n}(x, t) = \mathbf{N}(\xi, t_0, t_2, \dots; \varepsilon), \quad \xi = \delta(\varepsilon)x, \quad (4a)$$

with  $t_j = \varepsilon^j t$ ,  $j = 0, 2, \dots$  and

$$\mathbf{N} \sim \mathbf{n}_0 + \sum_{j=1}^{\infty} \delta_j(\varepsilon) \mathbf{N}_j(\xi, t_0, t_2, \dots). \quad (4b)$$

For normal diffusion ( $\gamma = 2$ )  $\delta = \varepsilon$ ,  $\delta_j = \varepsilon^j$  and a sequence of problems at successive orders  $\delta_j$  is obtained. For an anomalous system the scaling property of the fractional operator  $\mathfrak{D}_{|x|}^\gamma y(x) = \delta^\gamma \mathfrak{D}_{|\xi|}^\gamma y(\xi/\delta)$  determines the scale of the slow spatial variable  $\delta$ . Namely, in a more common case with  $\gamma_1 = \gamma_2 = \gamma$ , the scale is  $\delta = \varepsilon^{2/\gamma}$  and  $\delta_j = \varepsilon^j$ . Neglecting the phenomena evolving on time scales longer than  $\tau = t_2$ , at order  $O(\delta_1)$  a system of linear homogeneous equations at the bifurcation point is obtained:

$$\frac{\partial \mathbf{N}_1}{\partial t_0} - \nabla \mathbf{f}_0 \mathbf{N}_1 = 0, \quad (5)$$

solved as

$$\mathbf{N}_1 = A(\xi, t_2) e^{i\omega_0 t_0} \mathbf{v}_1 + \text{c.c.}, \quad (6)$$

where  $\mathbf{v}_1$  is an eigenvector of the linearised problem and  $\omega_0$  is the Hopf bifurcation frequency. At subsequent orders the system is not homogeneous. At order  $O(\delta_2)$

$$\frac{\partial \mathbf{N}_2}{\partial t_0} - \nabla \mathbf{f}_0 \mathbf{N}_2 = \sum_k \mathbf{r}_k e^{ik\omega_0 t_0}, \quad (7)$$

with  $k = \{-2, 0, 2\}$ , where  $\mathbf{r}_2 \propto A^2$ ,  $\mathbf{r}_0 \propto |A|^2$  and  $\mathbf{r}_{-2} = \mathbf{r}_2^*$ . The right-hand side contains no secular terms, and thus (7) is solvable. At  $O(\delta_3)$  the equation

$$\frac{\partial \mathbf{N}_3}{\partial t_0} - \nabla \mathbf{f}_0 \mathbf{N}_3 = \sum_{m=-3}^3 \mathbf{r}_m e^{im\omega_0 t_0} \quad (8)$$

is solvable conditionally due to the secular terms corresponding to  $m = \pm 1$ . The vector  $\mathbf{r}_1$  in (8) is a sum of

four terms proportional to  $\partial A / \partial \tau$ ,  $\mu A$ ,  $\mathfrak{D}_{|\xi|}^\gamma A$  and  $A|A|^2$ , and  $\mathbf{r}_{-1} = \mathbf{r}_1^*$ . Hence the amplitude equation, following from the solvability condition, has the form of a *fractional complex Ginzburg-Landau* (FCGL) equation:

$$\frac{\partial A}{\partial \tau} = A + (1 + \alpha i) \mathfrak{D}_{|\xi|}^\gamma A - (1 + \beta i) A |A|^2 \quad (9)$$

(in rescaled form). This equation was formerly derived in [39] in the problem of non-linear oscillators' dynamics with long range interactions. The parameters  $\alpha$  and  $\beta$  coincide with those of a normal reaction – diffusion system, but the Laplacian is replaced by the fractional operator. If  $\gamma_1 \neq \gamma_2$ , the super-diffusion term with the larger index is negligible in the long-wave region and  $\delta_j = \varepsilon^j$  for  $j \leq 3$  only. Higher-order powers are fractional and depend on the ratio of the anomalous exponents. Then the appropriate scaling is  $\delta = \varepsilon^{2/\gamma}$  with  $\gamma = \min\{\gamma_1, \gamma_2\}$ , and the expressions for  $\alpha$  and  $\beta$  are obtained by taking  $d_2 = 0$  if  $\gamma_1 < \gamma_2$  and  $d_1 = 0$  if  $\gamma_1 > \gamma_2$ .

**General properties.** – The integro-differential equation (9) retains the basic symmetries of a normal complex Ginzburg-Landau equation (with respect to time and space translations and the phase change  $A \mapsto A e^{i\vartheta}$ ). It is interesting that its solutions in the form

$$A(\xi, \tau) = B(\xi) e^{i(q\xi - \omega\tau)}, \quad q, \omega \in \mathbb{R} \quad (10)$$

have a symmetry similar to that found by Hagan [40]. If a solution of this type is known for a pair  $(\alpha, \beta)$ , the solution for a new pair  $(\alpha', \beta')$  located on one of the curves

$$(\alpha - \beta)/(1 + \alpha\beta) = \text{const}$$

can be found by the transformation  $B = aB'$ ,  $\xi = b\xi'$ , where

$$a^2 b^\gamma = \frac{1 + \alpha' \beta'}{1 + \alpha \beta} \frac{1 + \alpha^2}{1 + \alpha'^2}, \quad (11a)$$

$$b^\gamma = \frac{1 + \alpha^2}{1 + \alpha \alpha' + (\alpha - \alpha') \omega}, \quad (11b)$$

and the new wave number and frequency are  $q' = bq$ ,

$$\omega' = \alpha' - b^\gamma (1 + \alpha'^2) (\alpha - \omega) / (1 + \alpha^2). \quad (11c)$$

When  $\alpha = \beta$ , by a phase shift  $A \mapsto A e^{-i\beta\tau}$  eq. (9), like a normal complex Ginzburg-Landau equation [25], can be written in a variational form,

$$\frac{\partial A}{\partial \tau} = -(1 + i\beta) \frac{\delta \Upsilon}{\delta A^*}, \quad (12)$$

where  $\Upsilon = \int_{-\infty}^{\infty} U(\xi, \tau) d\xi$ , and

$$U = \frac{1}{2} (1 - |A|^2)^2 - \frac{\sec(\pi\gamma/2)}{2\Gamma(2-\gamma)} \left\{ \frac{\partial A^*}{\partial \xi} \frac{\partial}{\partial \xi} \int_{-\infty}^{\infty} \frac{A(\zeta) d\zeta}{|\xi - \zeta|^{\gamma-1}} + \frac{1}{2} A \int_{-\infty}^{\infty} \frac{\partial^2 A^*}{\partial \zeta^2} \frac{d\zeta}{|\xi - \zeta|^{\gamma-1}} + \text{c.c.} \right\} + \text{const.} \quad (13)$$

The constant is chosen so that  $\Upsilon$  converges. Then

$$\frac{\partial \Upsilon}{\partial \tau} = -2(1 + \beta^2)^{-1} \int_{-\infty}^{\infty} \left| \frac{\partial A}{\partial t} \right|^2 d\xi < 0, \quad (14)$$

and the system relaxes to a certain “stationary” solution (the original variable  $A$  oscillates with the frequency  $\beta$ ).

**Traveling-wave solutions.** – Consider the traveling-wave solutions of (9),

$$A_q = \sqrt{1 - |q|^\gamma} e^{i(q\xi - \omega\tau)}, \quad \omega = \beta - (\beta - \alpha)|q|^\gamma. \quad (15)$$

A small perturbation  $a(\xi, \tau)$  about  $A_q$  comprises longitudinal and transverse waves of the form

$$a = A_{q+k}(\tau) e^{i(q+k\xi)\xi + ik_\eta\eta} + A_{q-k}(\tau) e^{i(q-k\xi)\xi - ik_\eta\eta}, \quad (16)$$

with  $k_\xi, k_\eta$  being the respective wave numbers. The solution (15) is neutrally stable with respect to disturbances  $k_\xi = k_\eta = 0$ . Further analysis of long perturbations reveals that for  $O(k_\xi/q) \sim O(k_\eta/q) \sim o(1)$  to leading order the growth rate of  $A_{q\pm k} \sim e^{\lambda\tau}$  satisfies

$$\Re \lambda \sim \frac{\gamma}{2} |q|^\gamma \left[ -(1 + \alpha\beta) \left( (\gamma - 1) \frac{k_\xi^2}{q^2} + \frac{k_\eta^2}{q^2} \right) + \gamma(1 + \beta^2) \frac{|q|^\gamma}{1 - |q|^\gamma} \frac{k_\xi^2}{q^2} \right]. \quad (17)$$

Therefore, all solutions (15) are unstable if  $1 + \alpha\beta < 0$ , *i.e.* the Benjamin-Feir criterion for a normal CGLE is recovered. However, if  $1 + \alpha\beta > 0$ , a  $\gamma$ -dependent set of unstable wave vectors exists, generalising the Eckhaus instability criterion:

$$|q_m| < |q| < 1, \quad |q_m|^{-\gamma} = 1 + \frac{\gamma}{\gamma - 1} \frac{1 + \beta^2}{1 + \alpha\beta}. \quad (18)$$

No new instability criteria emerge in the opposite limit  $q \ll k_\xi, k_\eta \ll 1$ . In particular, the spatially homogeneous oscillation  $A_0 = \exp(-i\beta\tau)$  is unstable in the same region  $1 + \alpha\beta < 0$  with respect to disturbances whose wave numbers  $k$  satisfy

$$0 < |k|^\gamma < -\frac{2(1 + \alpha\beta)}{(1 + \alpha^2)}, \quad 1 + \alpha\beta < 0. \quad (19)$$

**Fractional Kuramoto-Sivashinsky equation.** – The evolution of perturbations near Benjamin-Feir instability domain boundary is expected to be described by an analogue of the Kuramoto-Sivashinsky equation [1]. Define  $1 + \alpha\beta = -\epsilon$ ,  $0 < \epsilon \ll 1$ , rewrite (9) with  $\chi = \epsilon^{1/\gamma}\xi$  and  $\tau_2 = \epsilon^2\tau$ , take

$$A = \exp(-i\beta\tau_2/\epsilon^2) r(\chi, \tau_2) e^{i\varphi(\chi, \tau_2)}, \quad (20a)$$

where

$$r = 1 + \sum_{j=1}^{\infty} \epsilon^j r_j(\chi, \tau_2), \quad \varphi = \sum_{j=1}^{\infty} \epsilon^j \varphi_j(\chi, \tau_2), \quad (20b)$$

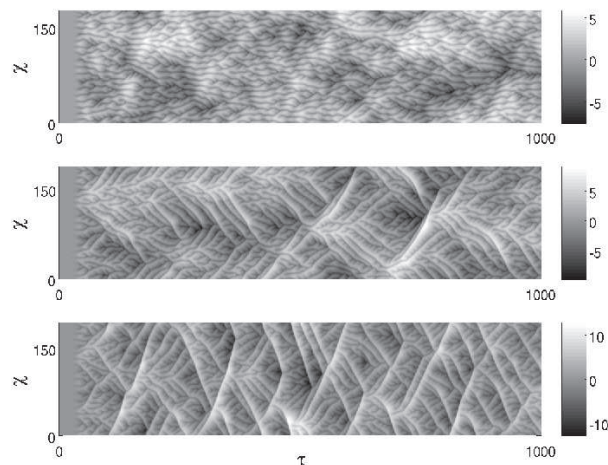


Fig. 1: Spatio-temporal dynamics of solutions of eq. (21) for  $\gamma = 2.0$  (top),  $\gamma = 1.7$  (middle), and  $\gamma = 1.6$  (bottom).

and expand  $e^{\pm i\varphi}$  (as the product rule does not apply here [38]) to obtain the phase diffusion equation at order  $O(\epsilon^3)$  that, after rescaling, has the following form (notations for the rescaled space and time variables are the same):

$$\frac{\partial \phi}{\partial \tau} = -\mathfrak{D}_{|\chi|}^\gamma \phi - (\mathfrak{D}_{|\chi|}^\gamma)^2 \phi + \frac{1}{2} \mathfrak{D}_{|\chi|}^\gamma \phi^2 - \phi \mathfrak{D}_{|\chi|}^\gamma \phi. \quad (21)$$

The operator  $(\mathfrak{D}_{|\chi|}^\gamma)^2$  is defined in Fourier space by  $(\mathfrak{D}_{|\chi|}^\gamma)^2 e^{iqx} = |q|^{2\gamma} e^{iqx}$  and cannot be simply related to the operator  $\mathfrak{D}_{|\chi|}^{2\gamma}$  as the order  $2\gamma$  exceeds the definition range in (2). Equation (21) is the *fractional Kuramoto-Sivashinsky equation*.

**Numerical simulations.** – Figure 1 shows spatio-temporal dynamics of the numerical solutions of (21) obtained by means of a pseudospectral code, using periodic boundary conditions and starting from small amplitude random data. The upper figure shows the dynamics for  $\gamma = 2$  corresponding to a normal KS equation: this is a well known spatio-temporal chaos, exhibiting merging and splitting of “cellular” structures [1]. The middle figure corresponds to  $\gamma = 1.7$ . One can see that along with the chaotic dynamics of “cells” large amplitude traveling “shocks” develop which emit cells still displaying chaotic dynamics. With further decrease of  $\gamma$  the shocks appear more frequently, propagate faster and their amplitude grows (see the lower panel corresponding to  $\gamma = 1.6$ ).

When  $\gamma$  decreases below a certain threshold that depends on the domain length, a single traveling shock is formed in the whole domain. An example of such a shock is shown in fig. 2. Here the shock is traveling with a constant speed (fig. 2a) while its “wings” exhibit spatio-temporally chaotic modulations (fig. 2b). Decreasing  $\gamma$  results in the increase of the shock amplitude

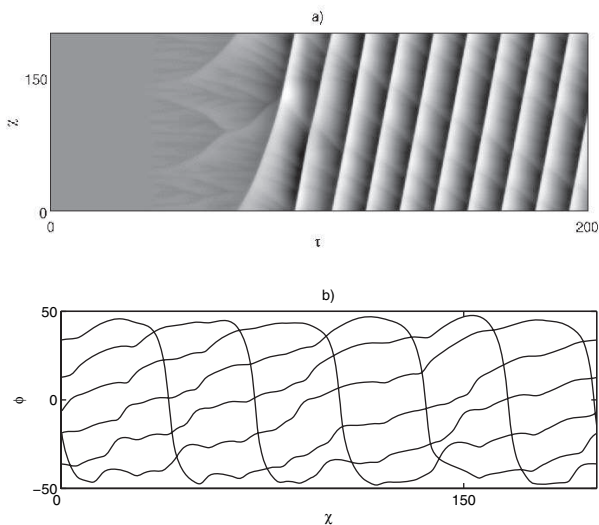


Fig. 2: Spatio-temporal dynamics of solutions of eq. (21) for  $\gamma = 1.5$  (a) and solutions at successive moments of time (b).

and after certain critical  $\gamma$  the shock starts accelerating with its amplitude growing exponentially. The shock amplitude grows with the size of the computational domain. An asymptotic analysis carried out for large amplitude solutions of (21) shows that the solution is of the form  $\phi = a(\tau)f(\xi - \zeta(\tau))$ , where  $f$  is an odd periodic function and  $a(\tau)$  grows exponentially (despite the problem non-linearity) with a certain dependence on the domain size and the velocity  $d\xi/d\tau$  proportional to  $a(\tau)$ . The numerical simulations confirm the asymptotic analysis.

Next, numerical simulations of the FCGL eq. (9) in 1D have been performed for the phase turbulence regime. Figure 3a shows spatio-temporal dynamics typical of the normal CGL equation, starting from the Benjamin-Feir unstable, spatially homogeneous oscillations: it is well described by the normal KS equation (see fig. 1a). Figure 3b shows the similar dynamics of eq. (9) for  $\gamma = 1.6$ . One can see that, after some period of phase turbulence, accelerating shocks form triggering the transition to defect turbulence shown in fig. 3c. The formation of the accelerating shocks seen in figs. 3b, c is consistent with the formation of shocks in the FKS equation discussed above.

Figure 4 shows the spatio-temporal dynamics of numerical solutions of (9) corresponding to the defect turbulence regime emerging from the Benjamin-Feir unstable wave (15) with  $q = 0.5$  for  $\gamma = 2.0$  and  $\gamma = 1.1$  (figs. 4a and b, respectively). One can see that in the anomalous case the defect turbulence has a stronger phase turbulence component and does not consist of propagating holes.

Finally, numerical simulations of FCGL (9) in 2D have been performed for the parameter values corresponding to the formation of spiral waves in the normal CGL equation. Periodic boundary conditions and small amplitude random initial data were used. The results are shown in

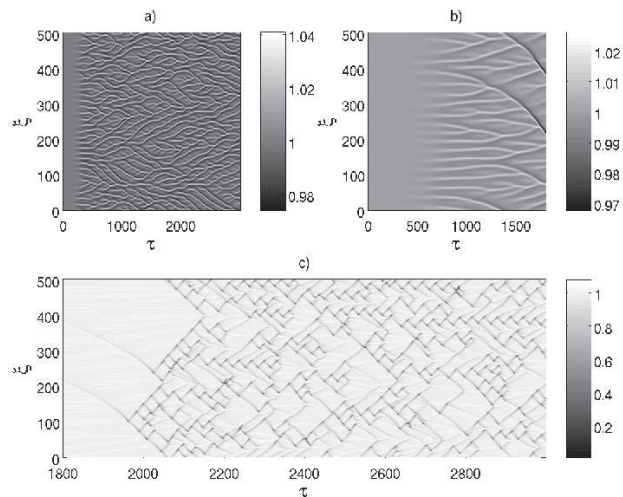


Fig. 3: Spatio-temporal dynamics of  $|A|$  — solution of eq. (9) for (a)  $\alpha = -1, \beta = 1.33, \gamma = 2.0$ ; (b)  $\alpha = -1, \beta = 1.2, \gamma = 1.6$ ; (c) continuation of (b) for the same parameter values.

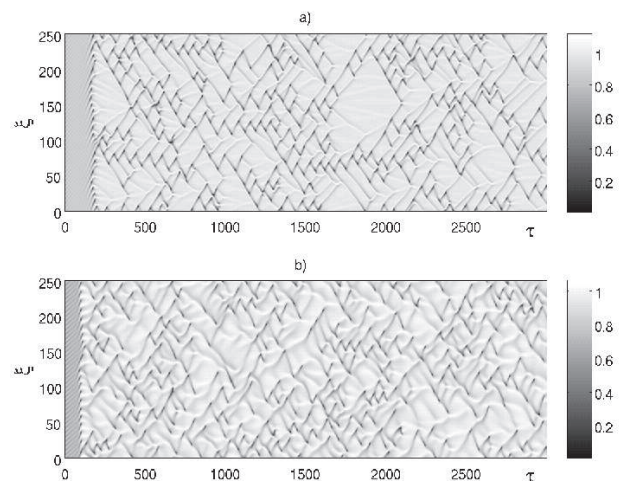


Fig. 4: Spatio-temporal dynamics of  $|A|$  — solution of eq. (9) for  $\alpha = 1, \beta = -1.3$  and (a)  $\gamma = 2.0$ ; (b)  $\gamma = 1.1$ .

fig. 5. One can see that for  $\gamma$  close to 2 (see figs. 5a, d) the formation of a spiral wave is still observed. With the decrease of  $\gamma$  the spiral wave regime is replaced by a defect chaos, however remnants of the spiral waves still can be seen (figs. 5b, e), with each “spiral” occupying a small domain with the domain walls partially melted. Further decrease of  $\gamma$  results in the decrease of the number of defects, the domain walls are almost completely melted (figs. 5c, f), and the local wave number created by each defect decreases. The regimes shown in figs. 5b, e and figs. 5c, f are similar to the defect chaos typical of the normal CGL equation [41]. In the normal CGL equation this regime appears when  $\alpha$  is increased. It is found that the regime in figs. 5b, e returns to the spiral glass state with the decrease of  $\alpha$ . No regimes qualitatively different

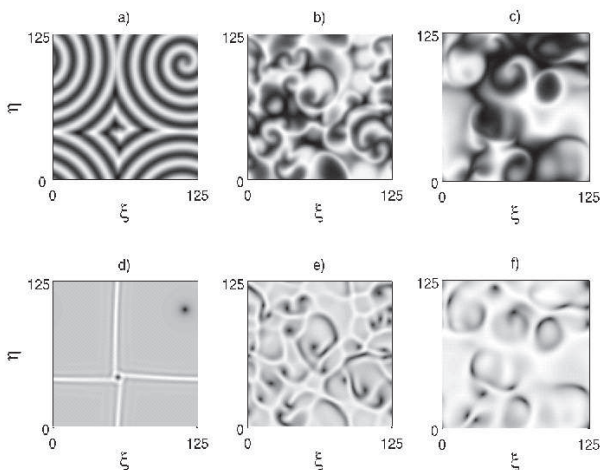


Fig. 5: Snapshots of solutions of eq. (9) for  $\alpha = 1.5, \beta = -0.6$  and  $\gamma = 1.9$  (a, d);  $\gamma = 1.8$  (b, e);  $\gamma = 1.05$  (c, f); upper panels:  $\text{Re}(A)$ ; lower panels:  $|A|$ .

from those observed for the normal CGL equation were found. Thus, based on the performed computations, one may conjecture that the anomaly shifts the boundaries of different regimes on the phase diagram in the parameter space [41] (corresponding to the normal CGL equation).

**Conclusion.** – Fractional Ginzburg-Landau and Kuramoto-Sivashinsky equations have been derived, describing weakly non-linear dynamics of a super-diffusive reaction – diffusion system characterised by Lévy flights, and some of their solutions have been studied analytically and numerically. Solutions of the FCGL equation in the form of traveling waves have been found and their stability studied. It has been shown that the Benjamin-Feir stability criterion holds also for the FCGL equation, whereas the Eckhaus stability domain is different from the normal CGL equation. Changes in the phase diagram boundaries of different regimes such as defect chaos, phase turbulence and spiral waves have been observed due to super-diffusion. It has been found that with the anomaly exponent diminution the spatio-temporal chaotic dynamics of cells is replaced by formation of moving shocks and ultimate blow-up. It is noted that investigating the effects of fluctuations on non-linear dynamics of instabilities in a super-diffusive reaction – diffusion system would be of interest since, as shown in [19], the fluctuations can have a profound influence on the non-linear behaviour in such systems. However, this topic is beyond the scope of the present paper.

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