Large spiral and target waves: Turbulent diffusion boosts scales of pattern formation

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Pattern formation in reaction-diffusion-advection (RDA) systems is an important process in many natural and man-made systems, e.g., plankton growth and iron fertilization in the ocean [1], dispersion of pollutants in the atmosphere, and optimal mixing in chemical reactors [2]. Spiral and target waves have been observed on small scales in various active media, e.g., in chicken retina [3], cardiac tissue [4] or chemical reactions [5, 6]. From a geophysical viewpoint it is of crucial interest if these reaction-diffusion patterns can also be found in large scale systems involving turbulent advection, as for example plankton dynamics in the ocean affecting CO₂ absorption [1, 2, 7].

However, so far, experimental evidence of these patterns in turbulent flows is lacking. Despite the importance of pattern formation in RDA systems only very few laboratory experiments on turbulent fluid flow involve reaction kinetics [9], and to our knowledge, none has considered excitable kinetics so far. Considerable numerical and experimental effort has focused on cellular and chaotic flows due to the simpler FKPP equation [∗]. Theoretically, the appearance of spiral and target waves should be possible in RDA systems whenever the advection term can be parameterised as a global diffusion coefficient [8]. However, so far, experimental evidence of these patterns in turbulent flows is lacking. Despite the importance of pattern formation in RDA systems only very few laboratory experiments on turbulent fluid flow involve reaction kinetics [9], and to our knowledge, none has considered excitable kinetics so far. Considerable numerical and experimental effort has focused on cellular and chaotic flows due to the simpler FKPP equation [∗].

We create a quasi two-dimensional turbulent flow using the Faraday experiment [14, 15], i.e. we vertically vibrate a circular container of 30 cm diameter filled with 2 mm of an excitable cyclohexandione and ferroin based Belousov-Zhabotinsky reaction (BZ) [16] (see methods summary and supplementary Fig. S1 [13]). The dynamics of this chemical reaction can be well observed in the visible range due to the oxidation of the reddish catalyst ferroin [Fe(phen)²⁺] to the blue ferriin [Fe(phen)³⁺] [17]. We vary the intensity of the turbulence and thus the turbulent diffusion constant [12] \( D_* \), by altering the amplitude \( a_0 \) of the acceleration and the frequency \( f \) of the vertical forcing.

For a quantitative analysis of the periods of the boosted spirals we varied the turbulent diffusion of the flow. This was achieved by changing only the forcing amplitude \( a_0 \) leaving the forcing frequency, and...
turbulent flow, i.e., voped boosted target waves in the quasi two-dimensional M7[13], as well as up to spirals with two free curling ends (supplementary movie to the spiral and target patterns we also observe double supplementary data, movies M4, M5 and M6[13]). In addition these pinned spirals last for up to \( \sim 50 \) mm diameter, placed in the middle of the container. ral to drift, we pinned its tip to a round obstacle of somewhat lower. Further, in order to prevent the spi-rals seem to be restricted by their own tail[20]. This of the width of the boosted autowaves such that the spi-rals last for up to \( \sim 1 \) h (see supplementary data, movies M4, M5 and M6[13]). In addition to the spiral and target patterns we also observe double spirals with two free curling ends (supplementary movie M7[13]), as well as up to 3 simultaneously existing spirals. All reactive waves had the typical characteristics of autowaves, in particular, they annihilate when they meet.

Figure 2(a) and inset (b) show that the FKPP relation for the front velocity \( v_f \) remains valid for well developed boosted target waves in the quasi two-dimensional turbulent flow, i.e., \( v_f = 2\sqrt{D_\ast/\tau_{\text{reac}}} \), \( \tau_{\text{reac}} \) being the reaction timescale \( \tau_{\text{reac}} \) was estimated from the velocity measurement of the molecular-diffusion-induced target wave to be \( \tau_{\text{reac}} = (0.8 \pm 0.3) \) s and the molecular diffusion coefficient was estimated from the literature to be \( D_{\text{mol}} \approx (1.3 \cdot 2) \times 10^{-3} \) mm\(^2\)/s[19, 24]. Theoretically, when the reaction timescale is small in comparison to the timescale of the fluid flow, the front velocity \( v_f \) is bounded by the unidirectional root-mean-square velocity of the flow instead of obeying the FKPP relation[25]. Inset (c) shows that in our experiments this limit is only approached for low forcing. We noted that the variation of the front velocity is related to the interval in between successive waves which suggests that they might obey a dispersion relation analogue to usual target waves[25].

In Fig. 2(e) the measured turbulent diffusion coefficient \( D_\ast \) is plotted as a function of the estimated Reynolds number for different forcing amplitudes. The turbulent diffusion increases approximately linearly with the Reynolds number as expected, and mixing is enhanced. At these Reynolds numbers the flow is turbulent as can be seen in an exemplary energy spectrum (\( Re \approx 120 \)) revealing a double cascade and a Kolmogorov type scaling \( (x \propto k^{-5/3}) \) in inset Fig. 2(f)[9, 15, 27]. The turbulent diffusion coefficients \( D_\ast \) were estimated from measurements of the absolute dispersion \( A(t) \) shown in Fig. 2 inset (e), by a fit to the regime of linear growth.

Despite the validity of the FKPP prediction for the front speed, Fig. 3 demonstrates that the boosted target waves do not entirely behave like their molecular diffusion counterparts. An important difference is the complex fil-
the root-mean-square flow velocity, estimated to be the ratio of the Faraday wavelength and of the flow and the reaction. The flow timescales were Da, the filaments can be explained by the Damköhler number, Fr, and thus lower forcing, the front appears sharper and its velocity.

For smaller turbulent diffusion (Fig.3(a)) the filamentous structure of the reaction front which is related to the small scale stretching and folding processes in the turbulent dynamics (Fig.3(a), (b) and Fig.1[9, 25, 28]). For larger Da, the fluid flow is fast compared to the reaction timescale, the peak of the diffusion-coefficient. Inset (f) shows exemplary the absolute energy spectra of the flow for Re ≈ 120. A double cascade and a regime with a Kolmogorov type scaling (Ek ∝ k−5/3) can be distinguished. kF is the typical Faraday wavenumber.

FIG. 2. (Color online) Front velocity of reactive waves in dependence of turbulent diffusion. (a) The velocity of the target wave fronts \(v_f\) scales with \(\sqrt{D_r}\) and follows the FKPP prediction \(v_f = 2\sqrt{D/\tau_{reac}}\) (solid line). The time constant of the reaction \(\tau_{reac} = (0.8 \pm 0.3)\) s was derived from the molecular case (circle) but adjusts also well for the turbulent data (crosses). Dashed lines indicate the error bounds estimated from the standard deviation of the velocity measurements from the molecular-diffusion-induced target wave. Inset (b) shows a close up of the turbulent data pairs. (c) Target front velocity \(v_f\) vs. turbulent root-mean-square velocity in one direction \(v' = v_{rms}/\sqrt{2}\), both normalized to the front velocity \(v_{mol}\) of the molecular-diffusion-induced target wave. (d) The measured diffusion coefficients are shown as a function of the Reynolds number \(Re = v_{rms}\lambda_F/\nu\) indicating the turbulence strength, where \(\nu\) is the kinematic viscosity of the fluid. Inset (e) shows exemplary the absolute diffusion for the flows with \(Re \approx 43\). \(Re \approx 120\) and \(Re \approx 194\) and the linear fit for estimation of the turbulent diffusion coefficient. Inset (f) shows exemplary energy spectra of the flow for \(Re \approx 120\). A double cascade and a regime with a Kolmogorov type scaling (\(E_k \sim k^{-5/3}\)) can be distinguished. \(k_F\) is the typical Faraday wavenumber. A double cascade and a regime with a Kolmogorov type scaling (\(E_k \sim k^{-5/3}\)) can be distinguished. \(k_F\) is the typical Faraday wavenumber.
turbulent diffusion coefficients and the reaction front velocities at various Reynolds numbers we find that they obey the FKPP relation for reaction-diffusion systems. The overall patterns resemble those of their molecular counterparts, however, an important difference is the filamentary appearance of the front which leads to an unexpected scaling of the front width. We suggest that this phenomena can be understood by the existence or absence of coherent structures in the flow that are known to exist in many turbulent flows. We expect our results to increase the attention on pattern formation in systems where excitable dynamics evolve in turbulent flows, such as plankton growth in the ocean where a ring-like structure, similar to a target, has been reported [30, 31].

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FIG. 3. (Color online) Front characteristics of boosted target waves. (a), (b) Space-time plots of boosted targets for $D_x \approx 5.4$ mm$^2$/s ($a \approx 1.3$ $g_0$) and $D_x \approx 30.0$ mm$^2$/s ($a \approx 2.2$ $g_0$), arrows indicate the direction of front propagation (supplementary movies M2 and M8 [13]). The target waves are narrower, slower and more filamentous for the smaller diffusion coefficient. (c), (d) Ferritin concentration, $[Fe(phen)]^{2+}$, along a line at three different instances of time, $\Delta t \approx 6.4$ s, for $D_x \approx 5.4$ mm$^2$/s and $D_x \approx 30.0$ mm$^2$/s respectively. (e), (f) Ferritin concentrations for the same values of $D_x$ at three different points in space ($\Delta x \approx 53$ mm, $\Delta t \approx 80$ mm). (g) The mean profile of the target waves for both diffusion coefficients estimated by averaging over all targets measured. (h) Different widths $w_1$ and $w_2$ of the profile in dependence of the diffusion coefficient $D_x$. The full width $w_2$ of the target wave grows with $\sqrt{D_x}$ as expected while the width of the rising edge $w_1$ stays constant.

at the tail of the front is much slower and sees a well-developed diffusive process. Timescales of the forward and the backward reaction can be estimated as the times of rise and fall of the ferritin concentration in Fig 3(e), (f).

In summary, we conclude that complex spatiotemporal patterns, such as target and spiral waves, occur in turbulent fluid flows as was shown experimentally. Measuring...
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