Defect-Mediated Turbulence in Systems with Local Deterministic Chaos

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Defect-mediated turbulence is shown to exist in media where the underlying local dynamics is deterministically chaotic. While many of the characteristics of defect-mediated turbulence, such as the exponential decay of correlations and a squared Poissonian distribution for the number of defects, are identical to those seen in oscillatory media, the fluctuations in the number of defects differ significantly. The power spectra suggest the existence of underlying correlations that lead to a different and nonuniversal scaling structure in chaotic media.

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Weakly driven, dissipative, pattern-forming systems often exhibit spatiotemporal chaos in the form of defect-mediated turbulence, where the dynamics of a pattern is dominated by the rapid motion, nucleation, and annihilation of point defects (vortices or dislocations) [1]. Examples can be found in electroconvection in liquid crystals [2], nonlinear optics [3], fluid convection [4,5], autocatalytic chemical reactions [6], cardiac tissue [7], and Langmuir circulation in the oceans [8], to name only a few. These results suggest that the dynamics of these very different systems can be characterized by a universal description which is based only on the defect dynamics.

A statistical description of defect-mediated turbulence based on a simple model for the defect dynamics was given by Gil *et al.* [9]. Treating the defect pairs as statistically independent entities, the nucleation rate for pairs of defects was taken to be independent of the number of pairs n and, based on the topological nature of the defects, the annihilation rate was taken to be proportional to n^2. This directly led to a squared Poissonian distribution for the probability distribution function (PDF) of n. Gil *et al.* found that simulations of a spatiotemporal chaotic state of the complex Ginzburg-Landau equation (CGLE), which is the prototype of oscillatory media, agreed with their prediction. Rehberg *et al.* [2] measured the PDF of n for defect-mediated turbulence in electroconvection of liquid crystals and found it to be consistent with the predicted squared Poissonian. Later, Ramazza *et al.* [3] investigated a defect turbulent state in optical patterns and found that their data were not conclusive. Very recently, Daniels and Bodenschatz [5] studied the defect-mediated turbulent state of undulation chaos in fluid convection and found that the observed pair nucleation and annihilation rates as well as the PDF agree with the theoretical predictions in [9] when boundary effects are taken into account.

The experimental and theoretical studies of defect-mediated turbulence have focused exclusively on first-order statistics like the PDF of n and creation and annihilation rates. Moreover, theoretical investigations have been carried out only for media with underlying oscillatory dynamics, generically described by the CGLE. Yet, in many cases the local dynamics may differ from simple oscillatory behavior; instead, complex-periodic or even chaotic attractors may exist (see, e.g., Ref. [10]). Chemically reacting systems, notably the Belousov-Zhabotinsky (BZ) reaction, are known to exhibit deterministic chaos [11]. This leads to a variety of new spatiotemporal states [12]. In this Letter, we show that defect-mediated turbulence can exist in media where the underlying local dynamics is chaotic and that second-order statistics can be used to distinguish between different media. This implies that a universal description of defect-mediated turbulence cannot encompass second-order or higher correlations.

Our focus is on systems where the dynamics of the spatially homogeneous system, described by ordinary differential equations, has a deterministic chaotic attractor; hence, at least three phase space variables are required in contrast to the two-variable descriptions of simple oscillatory media. A specific example of such a chaotic system is the Willamowski-Rössler (WR) reaction-diffusion model [13]

$$\partial_t \mathbf{c}(\mathbf{r}, t) = \mathbf{R}[\mathbf{c}(\mathbf{r}, t)] + D\nabla^2 \mathbf{c}(\mathbf{r}, t), \tag{1}$$

where $R_1 = \kappa_1 c_1 - \kappa_{-1} c_1^2 - \kappa_2 c_1 c_2 + \kappa_{-2} c_2^2 - \kappa_4 c_1 c_3 + \kappa_{-4}$, $R_2 = \kappa_2 c_1 c_2 - \kappa_{-2} c_2^2 - \kappa_3 c_2 + \kappa_{-3}$, and $R_3 = -\kappa_4 c_1 c_3 + \kappa_{-4} + \kappa_5 c_3 - \kappa_{-5} c_3^2$. These rate equations were derived from the mass action kinetics of a reaction scheme with quadratic kinetics where certain pool species are taken to be fixed. Here $c_i(\mathbf{r}, t)$ is the local concentration of species i at site $\mathbf{r}$ in a two-dimensional (2D) space of size L^2 with periodic boundary conditions. The parameters $\kappa_{\pm j}$ are rate coefficients that contain the concentrations of the pool species that are fixed to maintain the system out of equilibrium. The diffusion coefficients D of all three species are taken to be equal: $D = 1$. In this case, the value of D determines the spatial scales but does not affect the dynamics [14]. Even though the WR model is very simple, it exhibits a phenomenology [15] with

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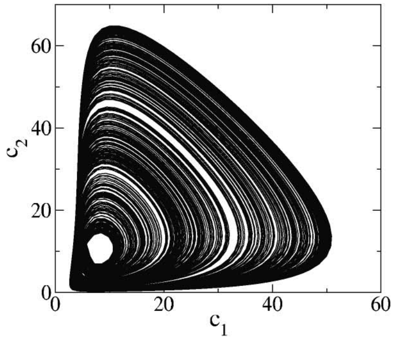

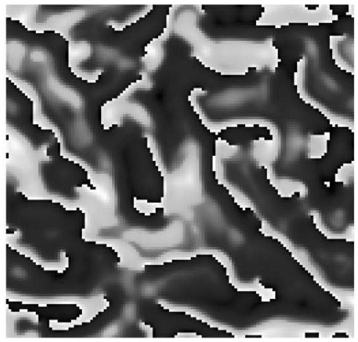

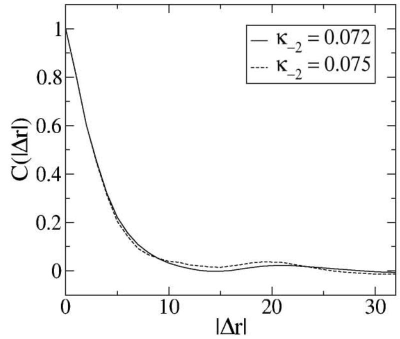

FIG. 1. Left: projection of the chaotic attractor in the (c_1, c_2) plane of the homogeneous WR model for $\kappa_1 = 31.2$, $\kappa_{-1} = 0.2$, $\kappa_2 = 1.45$, $\kappa_{-2} = 0.072$, $\kappa_3 = 10.8$, $\kappa_{-3} = 0.12$, $\kappa_4 = 1.02$, $\kappa_{-4} = 0.01$, $\kappa_5 = 16.5$, $\kappa_{-5} = 0.5$. Snapshots of the inhomogeneous **c** field in the defect turbulent state closely resemble this attractor. Center: phase field for $L = 128$ [16]. Right: normalized correlation function in the defect-mediated turbulent state for $L = 128$ and different values of κ_{-2}.

many features in common with those observed in chemical experiments on the BZ reaction [12]. Consequently, this model may be expected to capture the qualitative features of chemical systems whose chaotic attractors arise from a period-doubling cascade. The chaotic attractor is shown in the left panel of Fig. 1.

A defect is characterized by its integer topological charge (or winding number) m_{top} which is defined by $\frac{1}{2\pi}\oint \nabla\phi(\mathbf{r}, t) \cdot d\mathbf{l} = m_{\text{top}}$ [17], where $\phi(\mathbf{r}, t)$ is the local phase and the integral is taken along a closed curve surrounding the defect. A topological defect corresponds to a point in the medium where the local amplitude is zero and the phase is not defined. Typically only topological defects with $m_{\text{top}} = \pm 1$ are observed. One-armed spiral waves with such defects at their centers are the only stable spiral waves for the CGLE [18]. The phase has to be defined in order to apply the notion of defects to chaotic media. This is not a trivial issue and for many chaotic systems it is impossible to introduce a phase field. The rather simple shape of the WR chaotic attractor (see left panel of Fig. 1) which arises from a period-doubling cascade admits a simple definition of the phase and the WR model belongs to the class of chaotic-oscillatory media which is well-known from the study of phase synchronization (see Ref. [19] for a review). We chose $\phi(\mathbf{r}, t) = \arctan\{[c_2(\mathbf{r}, t) - c_2^0]/[c_1(\mathbf{r}, t) - c_1^0]\}$ with $(c_1^0, c_2^0) = (8.0, 9.0)$ as the center of rotation. For such simple chaotic attractors, the particular choice of a phase variable for chaotic systems does not influence the results [20]. In the center panel of Fig. 1, the phase field of the WR medium is shown for a certain set of parameters [16]. The topological defects can be identified as the termini of the white equiphase contour lines. As for the CGLE in the defect-mediated turbulent state [21,22], the defects in the WR system for these parameter values rarely emit waves. They behave as passive objects and are merely advected by the surrounding chaotic fluctuations. The right panel in Fig. 1 shows the correlation function $C(|\Delta\mathbf{r}|) = \langle \text{Re}[A(\mathbf{r}, t)\bar{A}(\mathbf{r} + \Delta\mathbf{r}, t)]\rangle_{\mathbf{r},t}$, where $A(\mathbf{r}, t)$ is the complex amplitude in the (c_1, c_2) plane with respect to the center of rotation and $\langle\cdots\rangle_x$ signifies an average over the (optional) argument x. It decays exponentially with a very short characteristic length scale, verifying the existence of a turbulent state. This is the most typical characteristic for defect-mediated turbulence and has been found in the CGLE as well [1].

The fluctuations in the number of pairs of topological defects shown in the left panel of Fig. 2 provide further evidence that a defect-mediated turbulent state can exist in chaotic-oscillatory media. The total number of defects in the medium is exactly twice the number of pairs because the net topological charge is conserved and equal to zero due to the periodic boundary conditions. Hence, topological defects can be created and annihilated only in pairs of opposite topological charge. One can easily derive a PDF $p(n)$ for the number of defect pairs provided that the defects are statistically independent entities [5,9]. In the stationary state and for periodic boundary conditions, the master equation reduces to $p(n) = p(n - 1)c(n - 1)/a(n)$, where $c(n)$ and $a(n)$ are the creation and annihilation rates, respectively. Provided that $c(n) = c = \text{const}$ and $a(n) = an^2$, $p(n) \propto (c/a)^n/(n!)^2$. The right panel of Fig. 2 shows that the PDF for the WR model agrees with the predicted form of a squared Poissonian distribution reasonably well. Moreover, the assumptions leading to this distribution seem to be justified: the annihilation rate scales approximately with n^2 and the creation rate is approximately independent of n as can be deduced from Fig. 3.

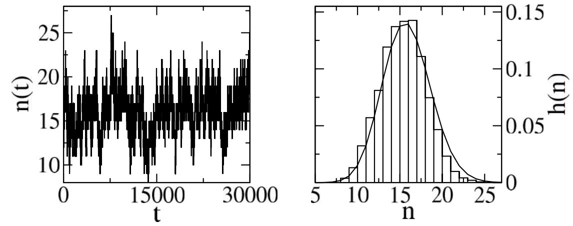

FIG. 2. Left: $n(t)$ for the parameters given in Fig. 1 and $L = 128$. Right: normalized histogram $h(n)$. The solid curve is the corresponding squared Poissonian distribution.

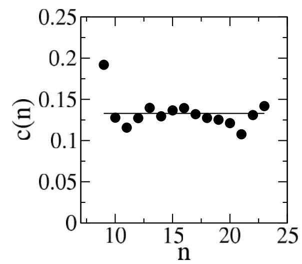

FIG. 3. Left: $c(n)$ in the WR system for the parameters given in Fig. 1 and $L = 128$. The solid line corresponds to a constant creation rate. Right: $a(n)$ in the WR system for the same parameters. The solid line corresponds to an increase in the annihilation rate proportional to n^2.

The fluctuations in the number of defect pairs do not have the properties one would expect if they arose from independent random events. If this were the case the power spectrum $S_L(f) = \lim_{T\to\infty} \frac{1}{2T} \mid \int_{-T}^{T} dt\, n(t) \times \exp^{-i2\pi f t}|^2$ for system size L would have a Lorentzian shape. The power spectrum does not have this form for the defect-mediated turbulence in the WR model. Figure 4 shows that $S_L(f) \propto 1/f^\gamma$ for intermediate frequencies with an exponent γ that is far from the value $\gamma = 2$ expected for a Lorentzian shape. For $k_{-2} = 0.072$, we find $\gamma = 1.43$ and, for $k_{-2} = 0.075$, $\gamma = 1.60$ [23]. Although in both cases the system exhibits defect-mediated turbulence, the exponents are significantly different from each other. This implies that different chaotic-oscillatory media can have different second-order statistics. To confirm that these results are not specific to the WR model, we have also studied the autocatalator reaction-diffusion system [24,25] which also has a chaotic attractor arising from a period-doubling cascade but with very different Lyapunov spectra. We find a power-law decay with values of γ that are similar to those reported here for the WR model [26]. These results suggest that the power spectrum of $n(t)$ may exhibit power-law decay with nontrivial exponents for defect-mediated turbulent states when the local dynamics exhibits deterministic chaos.

For large enough system sizes, $S_L(f) \propto L^2$ for all f; thus, the power spectrum for large systems can be considered to be the superposition of the power spectra of subsystems which is expected in view of the short correlation length. It implies that $(\langle n^2\rangle - \langle n\rangle^2)^{1/2}$ scales as $\sqrt{\langle n\rangle}$, which is proportional to L. Such a scaling is a consequence of the law of large numbers and has been observed for the CGLE as well. It follows that γ and the low-frequency cutoff—which is due to the fact that $n(t)$ is bounded—are independent of L. The cutoff's location depends on the density of defects in the medium. For lower densities it moves to lower frequencies.

To compare our results for $S_L(f)$ in chaotic media with those in oscillatory media, we simulate the CGLE [1,21]

$$\partial_t A = A + (1 + i\alpha)\nabla^2 A - (1 + i\beta)|A|^2 A, \qquad (2)$$

in a domain of size L^2 with periodic boundary conditions [16]. Here, A is the complex amplitude field and α and β are control parameters. The power spectrum of the CGLE in the defect-mediated turbulent state has a power-law decay for intermediate frequencies, and the scaling of $S_L(f)$ with L^2, as well as the dependence of the low-frequency cutoff on the density of defects, agrees with those in the WR medium. Yet, as Fig. 4 shows, $\gamma = 1.9$ [23] which is very different from the values observed for the chaotic media we analyzed. Thus, different media can have different second-order statistics for $n(t)$ depending on the underlying local dynamics. This argues against a universal description of defect-mediated turbulence. We note that the exponent for oscillatory media is extremely close to 1.87—the value we found for the simple model of Gil *et al.* Thus, even second-order statistics of oscillatory media can be quite well described by a purely random process where the interaction between defects of opposite charge is restricted to annihilating collisions.

To understand how the nontrivial correlations in $n(t)$ at intermediate time scales for the WR model arise, we have analyzed the series of waiting times between consecutive creation events and consecutive annihilation events separately. In both cases, the waiting times are exponentially distributed and statistically uncorrelated as for a random walk. This implies that the correlations in $n(t)$ are due to the interaction of creation and annihilation events. This is

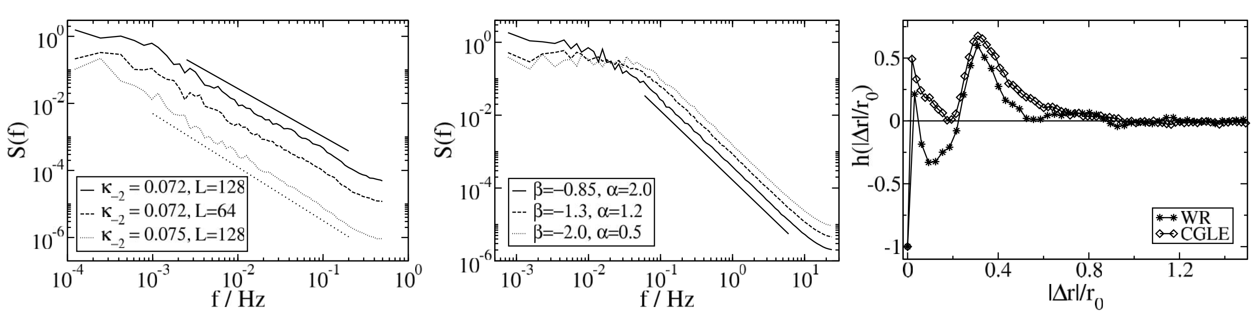

FIG. 4. Left: power spectrum of $n(t)$ for different parameters in WR. For clarity, the curve for $\kappa_{-2} = 0.075$ has been shifted down by one decade. The thick lines are to guide the eye. The thick solid line decays with $\gamma = 1.43$ and the thick dotted line with $\gamma = 1.60$. Center: power spectrum of $n(t)$ for different parameters in the CGLE and $L = 128$. The thick line is to guide the eye and decays with $\gamma = 1.9$. Right: normalized pair correlation function $h(|\mathbf{\Delta r}|/r_0)$ with $r_0 = \sqrt{L^2/\langle n\rangle}$.

further confirmed by the normalized pair correlation function for the defects, defined as [27]

$$h(|\Delta\mathbf{r}|) = \frac{\langle n_+(\mathbf{r}, t)n_-(\mathbf{r} + \Delta\mathbf{r}, t)\rangle_{\mathbf{r},t}}{\langle n\rangle^2} - 1, \qquad (3)$$

where $n_{+(-)}(\mathbf{r}, t)$ is the number of defects with $m_{\rm top} = +1(-1)$ at site $\mathbf{r}$ at time t. This function is shown in Fig. 4. For the CGLE, the first peak and its decay are very similar to what one would expect for a process where the motion of single defects is basically unaffected by defects of opposite charge as long as they do not collide: The peak is mainly due to the creation of pairs and decays to zero approximately proportional to $1/r$ as expected in 2D. In contrast, $h(|\Delta\mathbf{r}|)$ for the WR model has a pronounced negative value in the vicinity of $|\Delta\mathbf{r}| \approx 0.1r_0$. These strong anticorrelations imply that, with high probability, defects with opposite topological charge annihilate each other directly after their creation, or they separate quickly. This manifests itself in the behavior of $n(t)$ and is likely responsible for the nontrivial exponents in the power spectrum.

We have shown that defect-mediated turbulence can arise both in oscillatory and chaotic-oscillatory media and, thus, applies to a broad range of systems. While most common diagnostic measures do not allow one to distinguish between oscillatory and various chaotic media, the fluctuations in the number of defects can be different for different media: for the CGLE they resemble those of a simple creation-annhilation random process; for the WR and autocatalator models pronounced nontrivial correlations exist. Our results may be tested experimentally on systems like the Belousov-Zhabotinsky reaction whose local temporal dynamics can be complex periodic or even chaotic [12], and where defect-mediated turbulence has been observed [6].

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[1] P. Coullet, L. Gil, and J. Lega, Phys. Rev. Lett. **62**, 1619 (1989).
[2] I. Rehberg, S. Rasenat, and V. Steinberg, Phys. Rev. Lett. **62**, 756 (1989).
[3] P. Ramazza, S. Residori, G. Giacomelli, and F. Arecchi, Europhys. Lett. **19**, 475 (1992).
[4] S.W. Morris, E. Bodenschatz, D.S. Cannell, and G. Ahlers, Phys. Rev. Lett. **71**, 2026 (1993); A.L. Porta and C.M. Surko, Physica (Amsterdam) **139D**, 177 (2000).
[5] K.E. Daniels and E. Bodenschatz, Phys. Rev. Lett. **88**, 034501 (2002).
[6] Q. Ouyang and J.M. Flesselles, Nature (London) **379**, 143 (1996); Q. Ouyang, H.L. Swinney, and G. Li, Phys. Rev. Lett. **84**, 1047 (2000); L.Q. Zhou and Q. Ouyang, J. Phys. Chem. A **105**, 112 (2001).
[7] J.M. Davidenko, A.V. Pertsov, R. Salomonsz, W. Baxter, and J. Jalife, Nature (London) **355**, 349 (1993).
[8] T.M. Haeusser and S. Leibovich, Phys. Rev. Lett. **79**, 329 (1997).
[9] L. Gil, J. Lega, and J.L. Meunier, Phys. Rev. A **41**, 1138 (1990).
[10] A. Goryachev, R. Kapral, and H. Chaté, Int. J. Bifurcation Chaos Appl. Sci. Eng. **10**, 1537 (2000).
[11] S. Scott, *Chemical Chaos* (Oxford University Press, New York, 1991); J.-C. Roux and H.L. Swinney, *Nonlinear Phenomena in Chemical Dynamics*, edited by C. Vidal and A. Pacault (Springer-Verlag, New York, 1981), p. 38; J.L. Hudson and J.C. Mankin, J. Chem. Phys. **74**, 6171 (1981).
[12] J.-S. Park and K.J. Lee, Phys. Rev. Lett. **83**, 5393 (1999); **88**, 224501 (2002).
[13] K.D. Willamowski and O.E. Rössler, Z. Naturforsch. **35**, 317 (1980).
[14] Note that this is not true for nonidentical diffusion coefficients. Qualitatively similar results were obtained if diffusion driven instabilities were absent.
[15] A. Goryachev and R. Kapral, Phys. Rev. Lett. **76**, 1619 (1996); Phys. Rev. E **54**, 5469 (1996).
[16] The integrations were performed by a simple Euler scheme with time step $\Delta t = 0.0002(0.02)$, resolution $\Delta x = 1(0.5)$, and a 5(9)-point Laplacian for WR (CGLE).
[17] N. Mernin, Rev. Mod. Phys. **51**, 591 (1979).
[18] P.S. Hagan, SIAM J Appl. Math. **42**, 762 (1982).
[19] A.S. Pikovsky, M. Rosenblum, and J. Kurths, *Synchronization: A Universal Concept in Nonlinear Science* (Cambridge University Press, Cambridge, UK, 2001).
[20] K. Josić and D.J. Mar, Phys. Rev. E **64**, 056234 (2001); A.S. Pikovsky, M.G. Rosenblum, G.V. Osipov, and J. Kurths, Physica (Amsterdam) **104D**, 219 (1997).
[21] I.S. Aranson and L. Kramer, Rev. Mod. Phys. **74**, 99 (2002).
[22] H. Chaté and P. Manneville, Physica (Amsterdam) **224A**, 348 (1996).
[23] The error given by the regression analysis for γ is 0.02 (0.01) for WR (CGLE). The actual error is difficult to estimate and significantly larger due to different effects including the choice of the interval used for the fit.
[24] B. Peng, S.K. Scott, and K. Showalter, J. Phys. Chem. **94**, 5243 (1990).
[25] R. Kapral and X.-G. Wu, Physica (Amsterdam) **103D**, 314 (1997).
[26] For the parameters given in Ref. [25] and $\kappa_2 = 11.52$, we find $\gamma = 1.40$. The positive Lyapunov exponent in the homogeneous system is approximately a factor of 2 smaller than in the WR model for the parameters we have considered here. This and the γ variations in the WR systems with similar Lyapunov exponents suggest that γ does not depend solely on the largest Lyapunov exponent. This is further confirmed by the fact that γ varies for nonidentical diffusion coefficients.
[27] M. Hildebrand, M. Bär, and M. Eiswirth, Phys. Rev. Lett. **75**, 1503 (1995).