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## Preface

Stefan C. Müller



This Special Issue of Physica D is dedicated to Stefan C. Müller on the occasion of his 60th birthday. For many years, Stefan Müller has been at the forefront of research on spatio-temporal organization in nonequilibrium systems. During these years, he has influenced the careers of scores of scientists as their mentor and colleague. Much of Stefan's work focuses on emergent phenomena in chemical and biophysical systems. One of his key achievements is the careful characterization of spiral waves in excitable systems such as the autocatalytic Belousov-Zhabotinsky (BZ) reaction and the cellular slime mold Dictyostelium discoideum [1,2]. Other projects that he has pursued vigorously during his career concern the dynamics of excitation waves under the influence of electric fields, photochemical forcing, and optical feedback as well as the analysis of chemo-hydrodynamical instabilities [3,4]. His research philosophy has always attracted him to biophysical questions as perhaps best exemplified by his work on oscillating microtubules, spreading depression in neuronal tissue, and glycolytic waves.

Many of these research topics are represented in this Festschrift. However, they not only show Stefan Müller's research interests but also show a cross-section of current research activities in the field of emergent phenomena. The 21 articles span from fundamental physics through chemistry into biology, with an emphasis on emergent phenomena in chemical systems. Works in this latter group present approaches that vary widely, from pure chemical research (e.g., analysis of reaction mechanisms) to science that uses chemistry as a model. In this respect, Stefan's research has always been closer to the latter end of this spectrum as it takes its motivation primarily from biophysical problems and fundamental questions in nonlinear dynamics.

An important root of this multidisciplinary research field is the classical work by Onsager and Prigogine on nonequilibrium systems. The first paper [5] in our Focus Issue revisits some related questions in an investigation of entropy production in a pattern-forming reaction–diffusion model. The nonlinear dynamics of spatially extended systems is perhaps most strikingly exemplified by spatio-temporal chaos. Certain aspects of this topic are discussed by Hidaka et al. [6] for the example of soft-mode turbulence in electrohydrodynamics. Their experiments measure the non-thermal Brownian motion of tracer particles and reveal interesting insights into fluctuations and statistical properties of

transport in such a chaotic system. The subsequent paper by Vlad et al. [7] focuses on the kinetic rate terms of complex reaction–diffusion processes and specifically on the analysis of random channel kinetics. This work is motivated by experimental phenomena such as the decay and the preservation of marine organic carbon but should be also applicable to the study of heterogeneous catalytic reactions with surface reconstruction. Heterogeneous catalysis in general has always been an important reaction class in nonlinear reaction dynamics and this importance is perhaps best demonstrated by the work of Nobel laureate Gerhard Ertl [8]. In this Focus Issue, Wehner et al. [9] study surface processes in which intrinsic noise triggers transitions between bistable states.

The papers [10–13] describe studies of front propagation and other spatio-temporal patterns in autocatalytic reaction-diffusion systems. Such investigations have become an integral part of modern physical chemistry and provide a broad range of experimental models for studies in nonlinear dynamics. Rácz et al. consider an interesting autocatalytic reaction involving porphyrin complexes such as the hemin-bromate reaction [10]. Propagating zones of pH change are investigated by Kovacs et al. for the example of a reaction involving bromate, sulfite, ferrocyanide, and aluminum ions [11]. Here protons are the autocatalytic species. The dynamics of autocatalytic wave packets is analyzed in the article by Bordyugov [12], which focuses on onedimensional waves in the 1,4-cyclohexanedione-BZ reaction and their anomalous dispersion. Horvath et al. investigate the thioureaiodate-sulfite system in a one-side-fed continuous reactor [13]. Their investigation establishes this system as the only third aqueous reaction showing stationary Turing patterns.

Perhaps even more complex dynamics are discussed in [14–16]. Tinsley et al. [14] report results on dynamical quorum sensing and synchronization in collections of catalytic particles. The collective behavior of these particles shows a sharp, Kuramototype transition between the unsynchronized and synchronized dynamics. Microscopic catalytic particles are also the focus of the work by Echeverria and Kapral [15] who describe a particlebased mesoscopic simulation method, which – among other results - reveals interesting and strong deviations from the mean-field mass-action kinetics. Zykov et al. discuss novel results on a classic problem in mean-field reaction-diffusion systems, namely the rotation of spiral waves of excitation around unexcitable "holes" [16]. Such pinned vortices were studied experimentally first by Stefan Müller and co-workers [17] and are potentially relevant in the context of cardiac arrhythmia. Furthermore, vortex pinning also has recently been accomplished in spatially threedimensional media [18].

The next group of papers [19–21] is concerned with the "trinity" of chemical reactions, diffusion, and fluid flow. The overall topic not only has applications in engineering but also generates a

multitude of interesting nonlinear phenomena, such as large-scale convection and fingering patterns. The paper by Miike et al. [19], which is a short review of chemo-hydrodynamical instabilities in the Belousov-Zhabotinsky reaction, sets the stage for the two subsequent articles on fingering dynamics. D'Hernoncourt and De Wit study a case in which fingers arise from the buoyancy-driven convection around autocatalytic, exothermic reaction fronts [20]. Rica et al. report unusual cellular fingering patterns in experiments with the acid-catalyzed chlorite-tetrathionate reaction with added polyelectrolyte [21].

Some emergent phenomena in chemical systems have potentially useful and rather direct technological applications. A prominent example is the use of traveling reaction fronts for the production of polymeric materials. This topic is represented in our Focus Issue by the study of Viner et al. [22] who report experimental and theoretical results on the frontal polymerization of triacrylate. Zhai et al. [23] consider applications in the field of electrochemistry and study specifically the desynchronization and clustering of globally coupled oscillators, which also connects this work to the paper by Tinsley et al. [14]. Another exciting application of nonlinear dynamics in chemistry relates to the coupling of autocatalytic reactions and mechanically responsive gels which is analyzed in the theoretical study by Métens et al. [24]. The swelling and shrinking of the gel during chemoelasto-dynamical cycles might allow the construction of artificial muscles, intelligent microfluidic devices, and other self-regulated components. In addition, it generates novel instabilities and creates a large, widely unexplored, playground for scientists in nonlinear dynamics. For instance, Métens et al. stress that complex patterns in the polyacrylamide-methylene blue-oxygen system, which were first observed in Stefan's laboratory in 1999 [25], are still unexplained.

As mentioned above, many of the emergent phenomena in chemical reaction media are believed to exist also in biological systems. A striking example is given by glycolytic oscillations which are revisited by Zimanyi et al. [26] in a study that employs chemometric methods to untangle the dynamics of the multitude of involved chemical species from spectroscopic data. Glycolytic reactions in yeast extract can also cause spatio-temporal concentration patterns such as traveling waves and rotating spirals, which were first observed by Mair and Müller in 1996 [27]. The final papers [28–30] report results on such structures in four different biological systems. Takagi and Ueda [28] describe rotating wave patterns in the "true slime mold" Physarum polycephalum, while Ševčíková et al. [29] study wave propagation in the cellular slime mold D. discoideum. These microorganisms are important models in developmental biology and treasure troves of nonlinear dynamics. An experimental system of biomedical relevance is discussed in the paper by Dahlem et al. [30]. This final article [30] reports work on waves of depolarization (also known as "spreading depression") in the chicken retina and the gyrencephalic feline cortex. Close ties to strokes and migraine attacks are discussed.

We believe that this Focus Issue will provide the reader with a broad view of modern trends in the field of emergent phenomena. Although certainly not comprehensive, it nevertheless demonstrates the remarkable breadth of the field and its fascinating multidisciplinarity. These features are also evident in Stefan's great and many contributions. The editors express their personal thanks to Stefan Müller and join with the authors in wishing him all the best for the future.

## References

- [1] J. Ross, S.C. Müller, C. Vidal, Chemical waves, Science 240 (1988) 460-465.
- [2] P. Foerster, S.C. Müller, B. Hess, Curvature and spiral geometry in aggregation patterns of Dictyostelium discoideum, Development 109 (1990) 11–16.

- [3] O. Steinbock, V. Zykov, S.C. Müller, Control of spiral wave dynamics in active media by periodic modulation of excitability, Nature 366 (1993) 322-324.
- [4] M. Böckmann, S.C. Müller, Growth rates of the buoyancy-driven instability of an autocatalytic reaction front in a narrow cell, Phys. Rev. Lett. 85 (2000) 2506-2509
- [5] H. Mahara, T. Yamaguchi, Entropy balance in distributed reversible Gray-Scott model, Physica D 239 (11) (2010) 729–734. [6] Y. Hidaka, Y. Hosoakwa, N. Oikawa, K. Tamura, R. Anugraha, S. Kai, Nonequilib-
- rium temperature and fluctuation theorem in soft mode turbulence, Physica D 239 (11) (2010) 735-738
- [7] M.O. Vlad, D.H. Rothman, J. Ross, Random channel kinetic for reactiondiffusion systems, Physica D 239 (11) (2010) 739–745.
- [8] G. Ertl, Reactions at surfaces: From atoms to complexity (Nobel lecture), Angew. Chem. Int. Ed. 47 (2008) 3524-3535.
- [9] S. Wehner, S. Karpitschka, Y. Burkov, D. Schmeißer, J. Küppers, H.R. Brand, Stochastic aspects of pattern formation during the catalytic oxidation of CO on Pd(111) surfaces, Physica D 239 (11) (2010) 746-751.
- [10] K. Rácz, M. Burger, Z. Ungvarai-Nagy, Comparison of the oxidation of two porphyrin complexes by bromate with respect to wave propagation, Physica D 239 (11) (2010) 752-756.
- [11] K. Kovacs, M. Leda, V.K. Vanag, I.R. Epstein, Front propagation in the bromatesulfite-ferrocyanide-aluminum(III) system: Autocatalytic front in a buffer system, Physica D 239 (11) (2010) 757-765.
- [12] G. Bordyugov, N. Fischer, H. Engel, N. Manz, O. Steinbock, Anomalous dispersion in the Belousov-Zhabotinsky reaction: Experiments and modeling, Physica D 239 (11) (2010) 766-775.
- J. Horváth, I. Szalai, P. De Kepper, Pattern formation in the thiourea-iodatesulfite system: Spatial bistability, waves, and stationary patterns, Physica D 239 (11) (2010) 776-784.
- [14] M. Tinsley, A.F. Taylor, Z. Huang, F. Wang, K. Showalter, Dynamical quorum sensing and synchronization in collections of excitable and oscillatory catalytic particles, Physica D 239 (11) (2010) 785–790. [15] C. Echeverria, R. Kapral, Autocatalytic reaction dynamics in systems crowded
- by catalytic obstacles, Physica D 239 (11) (2010) 791-796.
- [16] V. Zykov, G. Bordyugov, H. Lentz, H. Engel, Hysteresis phenomenon in the dynamics of spiral waves rotating around a hole, Physica D 239 (11) (2010)
- [17] O. Steinbock, S.C. Müller, Chemical spiral rotation is controlled by lightinduced artificial cores, Physica A 188 (1992) 61-67.
- [18] Z. Jiménez, B. Marts, O. Steinbock, Pinned scroll rings in an excitable system, Phys. Rev. Lett. 102 (2009) 244101.
- [19] H. Miike, K. Miura, A. Nomura, T. Sakurai, Flow wave of hierarchical pattern formation induced by chemical waves: The birth, growth and death of hydrodynamic structures, Physica D 239 (11) (2010) 808-818
- [20] J. D'Hernoncourt, A. De Wit, Influence of heat losses on nonlinear fingering dynamics of exothermic autocatalytic fronts, Physica D 239 (11) (2010) 819-830.
- [21] T. Rica, E. Pópity-Tóth, D. Horváth, Á. Tóth, Double-diffusive cellular fingering in the horizontally propagating fronts of the chlorite-tetrathionate reaction, Physica D 239 (11) (2010) 831-837.
- [22] V.G. Viner, J.A. Pojman, D. Golovaty, Effect of phase change materials on the frontal polymerization of a triacrylate, Physica D 239 (11) (2010) 838-847.
- [23] Y. Zhai, I.Z. Kiss, H. Kori, J.L. Hudson, Desynchronization and clustering with pulse stimulations of coupled electrochemical relaxation oscillators, Physica D 239 (11) (2010) 848-856.
- [24] S. Métens, S. Villain, P. Borckmans, Mechano-chemical oscillations of a gel bead, Physica D 239 (11) (2010) 857–865.
- [25] O. Steinbock, E. Kasper, S.C. Müller, Complex pattern formation in the polyacrylamide-methylene blue-oxygen reaction, J. Phys. Chem. A 103 (1999) 3442-3446.
- [26] L. Zimanyi, P. Khoroshyy, Th. Mair, A chemometric method to identify enzymatic reactions leading to the transition from glycolytic oscillations to waves, Physica D 239 (11) (2010) 866-872.
- [27] Th. Mair, S.C. Müller, Traveling NADH and proton waves during oscillatory glycolysis in vitro, J. Biol. Chem. 271 (1996) 627-630.
- [28] S. Takagi, T. Ueda, Annihilation and creation of rotating waves by a local light pulse in protoplasmic droplet of the *Physarum* plasmodium, *Physica D* 239 (11) . (2010) 873–878.
- [29] H. Ševčíková, J. Čejková, L. Krausová, M. Přibyl, F. Štěpánek, M. Marek, A new traveling wave phenomenon of Dictyostelium in the presence of cAMP, Physica D 239 (11) (2010) 879-888.
- [30] M. Dahlem, R. Graf, A.J. Strong, J.P. Dreier, Y.A. Dahlem, M. Sieber, W. Hanke, K. Podoll, E. Schöll, Two-dimensional wave patterns of spreading depolarization: Retracting, re-entrant, and stationary waves, Physica D 239 (11) (2010) 889-903.

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