DRIVEN CHEMISTRY AND INCREASINGLY COMPLEX FORMS OF DYNAMIC ORDER

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ABSTRACT
We review some of the main advances in our understanding of the living state of matter as well as its stability. We cite some of the main ideas related to the production of self-organizing chemical order. We present preliminary, experimental results on two topics. In the first part we discuss prebiotic broths, following the ideas of Miller Urey. We show that by studying the dynamics of their molecular organization, temporal patterning emerges as a coherent feature of driven, random organic soups. In the second part, we consider coexisting self-reproducing agents. We ask how the self-reproducing agents attain a robust evolution towards increased complexity, rather than getting stuck by producing a single "winner". By simplifying the system to one dimension, we show that the system may escape from getting stuck by generating new configurations. We find, however, that the theoretical construct is difficult to realize experimentally, without detailed tuning of the parameters by hand. This may well explain why, to our knowledge, a one dimensional molecular ecosystem did not emerge as a result of evolution.

BACKGROUND

In his famous lecture series "What is Life?"[1], Erwin Schrödinger asked how the events in space and time that take place within the spatial boundary of a living organism, can be accounted for by physics and chemistry. Among many interesting and far-reaching reflections, Schrödinger insisted that living matter creates order from disorder. Living matter evades the decay to thermodynamic equilibrium by feeding: it gains ' negentropy ' in an open system.

Chemical pattern formation: the Turing mechanism

More than 10 years later Turing was the first to propose a basic mechanism of order generating chemical pattern formation[2], in non-equilbrium conditions. He realized that two substances, which he termed morphogens, an activator (a) and an inhibitor (h), were needed to create a chemical pattern. Reaction-Diffusion (RD) equations of the type

$$\frac{\partial a}{\partial t} = D_a \nabla^2 a + S_a(a,h) - Q_a(a,h)$$
$$\frac{\partial h}{\partial t} = D_h \nabla^2 h + S_h(a,h) - Q_h(a,h)$$

can lead to chemical patterning if there are sufficiently strong non-linearities in the source (S) terms (in fact, stronger self-activation than simple autocatalysis is required). To observe pattern formation, it is also necessary that the diffusion constants D be sufficiently different (or the morphogen decay rates, given by Q). Alain Turing suggested that the reaction diffusion mechanism may explain patterning in developmental biology. In spite of its seducing simplicity, even today there are only a limited number of cases in Biology where molecules/morphogens that follow an RD scheme have been clearly determined. Turing suggested that the organism Hydra, a 1cm tall polyp, could follow an RD scheme. Hydra can regrow lost body parts, it can even reform from a random cluster of its own cells. The work of Hans Meinhardt discusses this phenomenon at length, showing that almost any experiment can be explained based on an RD mechanism. In spite of an intense experimental search, the morphogens could not be isolated, suggesting a different pattern generating mechanism on the molecular scale. We have shown that next-neighbour signalling among the participating cells can explain the symmetry breaking and axis formation very well [3]. In order to comply to experiments, we borrow a mechanism from self organized critical systems. Although pattern forming systems are a major part of biology and the same applies to chemical oscillators, in the organism these phenomena remain poorly understood from a reductionist point of view.

In chemistry the realization of pattern forming reactions has been the subject of intense research. The first self-organizing periodic reaction in homogeneous solution was discovered by Bray in 1926, while studying the reduction and oxidation properties of H_2O_2 [4]. However, even today only few examples of pattern forming chemical systems exist. The Beloussov-Zabotinsky reaction [5] is the famous realization of a reaction diffusion mechanism that leads to colorful waves in an initial homogeneous solution. Initially this type of reaction was considered impossible to realize experimentally. The Briggs Rauscher reaction [6] was inspired from BZ. It is another classic of the few examples of a spontaneously oscillating chemical reaction. The detailed reaction mechanisms of both reactions are rather involved. Complexity could be a characteristic feature of this type of behaviour. Autocatalysis is the essential element in generating the required non-linearities for the phenomenon. Often the non-linearity is enhanced through the use of metal or

halogenide compounds with a strong and broad oxidation potential.

In physics it has long been known that the state space of a nonlinear system contracts if the system dissipates: the system becomes more ordered when driven. Among well-known examples of pattern forming phenomena is the Rayleigh Bénard convection, where rising liquid, due to heating at the bottom, will produce regular convection roles. Many other examples exist: cloud formation, dunes, turbulent fluids, mode selection in a laser. The patterns occur as a result of competition of 'modes' of the nonlinear system. The strongest among them wins and creates a pattern by starving the others in terms of energy.

The hypercycle

It has been proposed that self-reproduction is the strongly growing chemical "mode" that necessarily "wins" and dominates the dynamic behaviour of biochemistry. Manfred Eigen and Peter Schuster theoretically considered simple systems of reproducing molecular information carriers in competition [7]. In simple situations they could show that the selection process is governed by the same equation as mode selection in a laser. They also addressed a more complex situation: the evolution, in competition, of multiple, self-reproducing, cyclic, mostly deterministic reactions of information carriers and enzymes, so called hypercycles. In this case, a molecule like RNA contains the information for the production of enzymes, which in turn multiply the RNA and, at the same time, read the stored information to produce more enzyme. In the system, Schuster and Eigen introduced molecular self-reproduction by hand, and the molecular pathways have reduced complexity compared to real chemistry. As a result the hypercycles do not produce evolution, although they undergo mutations. In most cases, it is the hypercycle with the largest number of elements that wins the competition and the evolution remains stuck.

However, chemistry is different from systems that physicists usually investigate, including the system studied by Eigen and Schuster. Chemistry includes the transformation of matter, and the rules of chemistry evolve with changing concentrations, and with new molecules or molecular scaffolds that appear. No satisfying statistical characterization of (organic) chemistry has been achieved so far. Experimentally it is not clear, how chemical systems can be supplied with energy so that they self-organize in a non-trivial way. Even, theoretically, a system that compares to living matter has not yet been suggested.

With their reflections on hypercycles, Eigen and Schuster revealed one of the challenging problems that have not been solved: how can a number of reproducing entities coexist in an ecosystem and undergo Darwinian evolution without one species dominating the system and ultimately halting the process?

EXPERIMENTAL AND RESULTS

We study the time evolution of a Miller Urey type random prebiotic broth [8] using mass spectroscopy. We show [9] that the system produces polymerized substances as sudden bursts. They tend to disappear and the process restarts. Thus reversibly assembling and disabling biodynamers emerge spontaneously as a result of driving chemical reactions.

We study the evolution of template based reproducing DNA strands that are allowed to double their length[10] We show that the experiment can produce filaments with increasing length only if it is helped by the experimentalist, in a narrow range of parameters. This is in contrast to theory, where a situation with several coexisting filaments lengths easily occurs as a stable dynamic result.

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