

# Flows of information in spatially extended chemical dynamics

Kristian Lindgren<sup>1</sup>, Anders Eriksson<sup>1</sup> and Karl-Erik Eriksson<sup>2</sup>

<sup>1</sup>Department of Physical Resource Theory, Chalmers University of Technology and Göteborg University, SE-41296 Göteborg, Sweden

<sup>2</sup>Division of Engineering Sciences, Physics and Maths, Karlstad University, SE-65188 Karlstad, Sweden  
frtkl@fy.chalmers.se

## Abstract

A continuity equation for information is presented, involving flows in both space and scale in chemical pattern formation systems. The flows are connected to thermodynamic properties of the system. Information leaves the system (or is destroyed) at the smallest length scales, which corresponds to entropy production. Information enters the system at the largest length scales when the system is open to an inflow of Gibbs free energy. The continuity equation describes how information can be aggregated at different scales and positions during the pattern formation process. The formalism is applied to the Gray-Scott model, exhibiting self-replicating spots in a spatially extended system.

The aim of the present paper is to give a physically consistent picture on how spatial structure emerge in chemical systems. This picture should illustrate how aggregation of information in the form of spatial patterns can be connected to the physical constraints of the studied system posed by thermodynamics, in particular the driving forces in terms of flows of free energy. Our ambition is to contribute towards a theoretical framework, that may be used to find necessary conditions for the formation of higher-order structures from lower-level components and mechanisms – a framework to encompass both biological life and artificial life.

Pattern formation in chemical systems is an example of a process where the connection between information theory, statistical mechanics, and thermodynamics is useful for a description and characterisation of the dynamics. Patterns can be characterised by their information content - a characterisation that may include how information is distributed in the system, both with respect to position and scale of resolution (Eriksson and Lindgren, 1987). At the same time, the flows of Gibbs free energy that drives the pattern formation process, or maintains the spatial structures that have been formed, can be given an information-theoretic interpretation. By combining these information-theoretic perspectives with the general reaction-diffusion type of chemical dynamics, we derive a continuity equation for information in open chemical systems. Information flows in both space and scale; flows across system boundaries are due to inflow of Gibbs free energy, and at the microscopic level to entropy

production in chemical reactions and in the diffusion process. The results presented here is a generalisation of the equation derived earlier for closed systems (Eriksson et al., 1987).

The formalism is exemplified by an analysis of the Gray-Scott model (Gray and Scott, 1984), exhibiting self-reproducing “spots” in a spatially extended system (Lee et al., 1993; Pearson, 1993). In this model, two chemical components U and V react according to  $U + 2V \rightarrow 3V$ ,  $V \rightarrow G$ , where G is not reacting with U or V.

The formalism is based on the information-theoretic concept of relative information or Kullback information (Kullback, 1959), defined by

$$K[P^{(0)};P] = \sum_i p_i \log \frac{p_i}{p_i^{(0)}} \geq 0,$$

where  $P$  and  $P^{(0)}$  are normalised probability distributions. This measure quantifies the information one gains when one learns that the a priori distribution  $P^{(0)}$  was not correct but that the system is described by  $P$ . An advantage with the use of this quantity as a basis for the information-theoretic analysis of the spatial structure in a chemical system is the strong connection to statistical mechanics and thermodynamics (Jaynes, 1957). This will make it possible to relate our information-theoretic analysis to thermodynamic properties of the system.

The exergy  $E$ , or available energy, for a system of volume  $V$ , characterised by pressure  $p$ , temperature  $T$ , and chemical potentials  $g_i$  (for  $M$  different components), in an environment characterised by the corresponding intensive variables ( $p_0$ ,  $T_0$ , and  $g_{i0}$ ), can be written (Reif, 1985)

$$E = S(T - T_0) - V(p - p_0) + \sum_{i=1}^M N_i(g_i - g_{i0}),$$

where  $S$  is the thermodynamic entropy of the system and  $N_i$  is the number of molecules of the different types. The total number of molecules is  $N = \sum_i N_i$ .

Assuming  $T = T_0$  and  $p = p_0$ , and ideal gas expression for the chemical potential  $g_i = k_B T_0(C + \ln c_i)$ , with normalised

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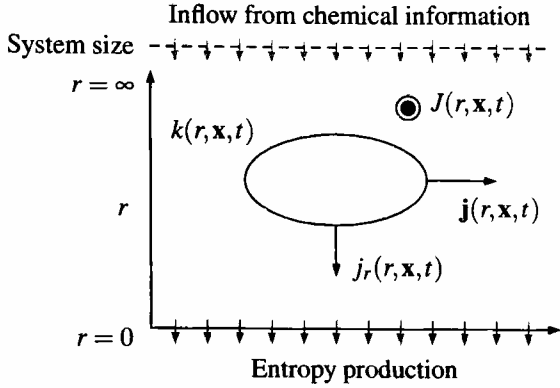


Figure 1: A schematic picture of the flows of information in a chemical pattern formation system. The pattern is characterised by an information density  $k(r, \mathbf{x}, t)$  distributed over spatial dimensions as well as over different length scales  $r$ . Information flows both in space and in scale, where the flow is destroyed when it gets down to the microscopic level. Here information disappears into microscopic degrees of freedom due to entropy production. Information enters the system at the very large scale due to a diffusion-controlled inflow of chemical information or Gibbs free energy. A pattern is formed when information that flows downwards in scale is aggregated at certain positions as described by the continuity equation.

concentrations  $c_i = N_i/N$  and corresponding concentrations  $c_{i0}$  in the environment, we get a Kullback information expression for the exergy, or the Gibbs free energy, of the system

$$E = k_B T_0 N \sum_{i=1}^M c_i \ln \frac{c_i}{c_{i0}} = k_B T_0 N K[c_0; c]$$

or, in general, when spatially dependent concentrations are allowed,

$$\begin{aligned} E &= k_B T_0 n \int_V d\mathbf{x} \sum_{i=1}^M c_i(\mathbf{x}) \ln \frac{c_i(\mathbf{x})}{c_{i0}} \\ &= k_B T_0 n \int_V d\mathbf{x} K[c_0; c(\mathbf{x})] = k_B T_0 n K, \end{aligned}$$

where  $n = N/V$ . This relation between exergy  $E$  and information  $K$  allows us to connect the information-theoretic analysis to the thermodynamic properties of the system. The integrated Kullback information,  $K$ , between actual and equilibrium concentrations, is our starting point for the analysis of spatial structure. First, by using the average concentration the information can be decomposed into a structural part  $K_{\text{struct}}$  and a chemical part  $K_{\text{chem}}$ . The structural information measures deviation from a homogenous state, and the chemical information measures the deviation of average

concentrations  $\bar{c}_i$  from chemical equilibrium:

$$\begin{aligned} K &= \int_V d\mathbf{x} \sum_i c_i(\mathbf{x}) \ln \frac{c_i(\mathbf{x})}{\bar{c}_i} + V \sum_i \bar{c}_i \ln \frac{\bar{c}_i}{c_{i0}} \\ &= K_{\text{struct}} + K_{\text{chem}} \end{aligned}$$

In order to get a more detailed description, we continue with a decomposition of the structural information into contributions  $k(r, \mathbf{x}, t)$  from different positions  $\mathbf{x}$  as well as from different length scales  $r$ . In order to do this, we introduce a resolution dependent concentration distribution. We define the concentration  $\tilde{c}_i(r, \mathbf{x}, t)$  at resolution  $r$  as

$$\tilde{c}_i(r, \mathbf{x}, t) = \exp\left(\frac{1}{2} r^2 \nabla^2\right) c_i(\mathbf{x}, t),$$

where  $\exp(\frac{1}{2} r^2 \nabla^2)$  is the resolution operator in  $d$  dimensions, defined by the convolution with the kernel  $(2\pi r^2)^{-d/2} \exp[-\mathbf{x}^2/(2r^2)]$ . As can be seen in the Fourier space, this operator suppresses structures at length scales  $\lesssim r$ . At  $r = 0$ , the two concentrations coincide since  $\tilde{c}_i(0, \mathbf{x}, t) = c_i(\mathbf{x}, t)$ . If  $r$  is much larger than the system size, the concentration is approximately homogenous,  $\tilde{c}_i(\infty, \mathbf{x}, t) = \bar{c}_i(t)$ .

This means that the structural information can be written

$$K_{\text{struct}} = \int_0^\infty dr/r \int d\mathbf{x} k(r, \mathbf{x}, t),$$

with a local information density at position  $\mathbf{x}$  and scale (resolution)  $r$ ,

$$k(r, \mathbf{x}, t) = \sum_i \tilde{c}_i [r \nabla \ln \tilde{c}_i]^2 \geq 0.$$

In the following we present an information-theoretic description of how information is flowing in the system that connects to the thermodynamic loss of information due to entropy production. We will assume that the system evolves according to the reaction - diffusion equation,

$$\dot{c}_i = D_i \nabla^2 c_i + F_i(c).$$

This will be formulated in a continuity equation for information density  $k$ , taking into account flows both in scale ( $r$ ) and in space ( $\mathbf{x}$ ), see Figure 1. There may also be sources or sinks due to the fact that we allow for an open system. By assuming that the following continuity equation holds, we can derive the terms for the flow in the scale direction  $j_r(r, \mathbf{x}, t)$ , the spatial flow  $\mathbf{j}(r, \mathbf{x}, t)$  and the sources/sinks term  $J(r, \mathbf{x}, t)$ ,

$$\dot{k}(r, \mathbf{x}, t) = r \frac{\partial}{\partial r} j_r - \nabla \cdot \mathbf{j} + J.$$

In order to properly define the flows, we also require (i) that the flows are rotation-free, (ii) that the system is spatially closed, i.e., the chemical flows across the system

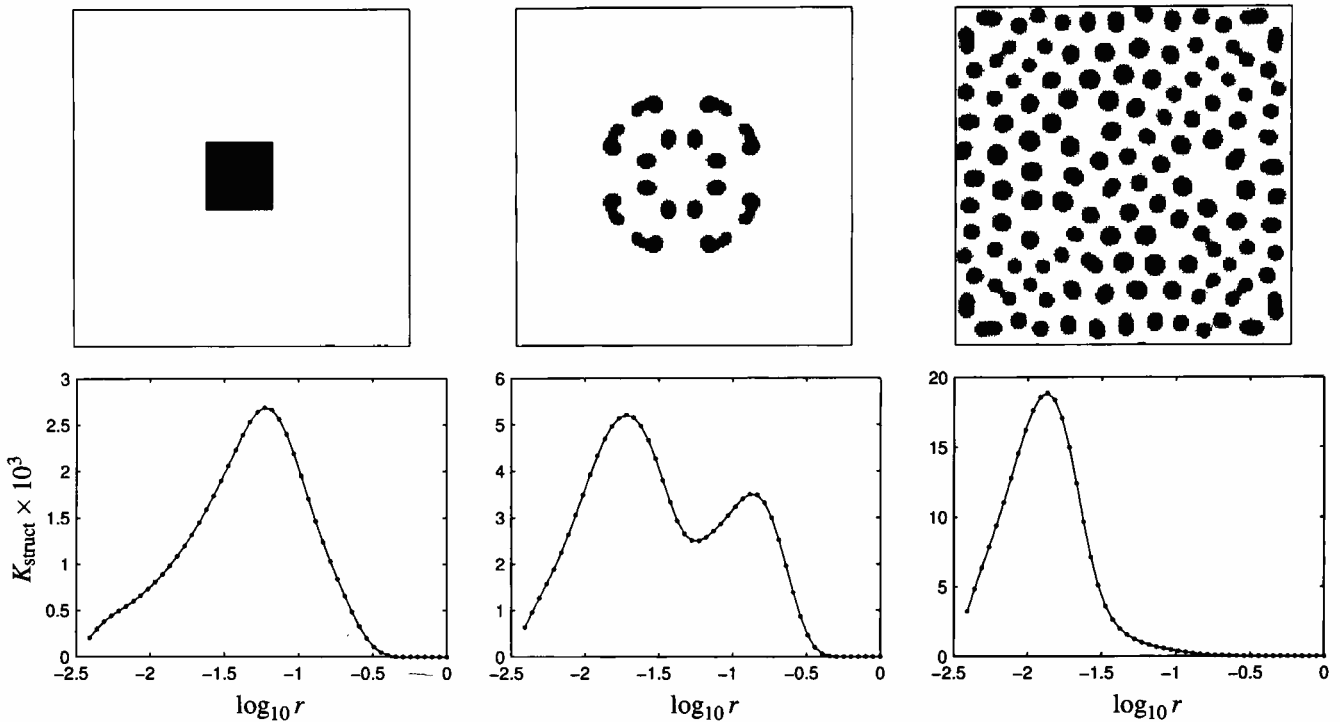


Figure 2: **Top row** The concentration of the chemical V in the system at three times;  $t = 0$ ,  $t = 1000$ , and  $t = 7000$  steps. White corresponds to zero concentration, and black corresponds to a concentration of one half. **Bottom row** The structural information, integrated over the system, as a function of the resolution  $r$ . The length of the system is 1,  $D_u = 2D_v = 0.05$ ,  $\hat{D} = 0.02$ ,  $k = 0.058$ .

boundaries are in a different direction (in which the system has no extension), (iii) that the information content of the inflow and outflow corresponds to the net flow of Gibbs free energy into the volume, and (iv) the destruction of information corresponds to the entropy production.

In the limit of  $r \rightarrow \infty$  we cannot distinguish any spatial structure, but the chemical information  $K_{\text{chem}}$  is still present, unaffected by the resolution parameter. In a decomposition of the total information this part can therefore be considered as present at the  $r \rightarrow \infty$  limit. The chemical information will be consumed by the chemical reactions and we should therefore expect that a proper definition of information flow shows how information will flow in the direction towards smaller length scales  $r$ . If the system gives rise to spatial structure, that should be captured in the continuity equation, resulting in a temporal accumulation of structural information.

The entropy production is determined by one term corresponding to the entropy produced due to diffusion in the system and one term given by the reactions that tend to even out the chemical non-equilibrium in the system. The entropy production certainly leads to a decay of the information in the system – decay of structural information as well as of chemical information.

It is reasonable to assume that information is leaving the system, through the thermodynamic entropy production, at the smallest length scales of the system, i.e., at  $r = 0$ . At this point information disappears from the macroscopic description of our system, and the information is spread out on microscopic degrees of freedom.

Therefore, we define the information flow  $j_r$  in the direction of smaller  $r$ , at the border  $r = 0$ , to be equal to the chemical entropy production. To define this flow for general resolution values  $r$ , we generalise by introducing the resolution operator into the expression for entropy production,

$$j_r(r, \mathbf{x}, t) = \sum_i D_i \frac{[\nabla \tilde{c}_i]^2}{\tilde{c}_i} + J_{\text{pot}}$$

$$J_{\text{pot}}(r, \mathbf{x}, t) = \left( \ln \frac{\tilde{c}_i}{c_{i0}} \right) \exp\left(\frac{r^2}{2} \nabla^2\right) F_i(\mathbf{c}(\mathbf{x}, t))$$

Now we use the continuity equation for the closed system, in which  $J = 0$ , to define the spatial flow by  $\mathbf{j}(r, \mathbf{x}, t) = r^2 \nabla J_{\text{pot}}$ .

In the case of an open system, the flow of chemical information through the system (typically an inflow of a substance acting as a fuel and an outflow of waste products) affects the pattern formation process in two opposite ways. If the flow across the system boundary is controlled by diffusion, the direct effect is a decrease of the spatial structure

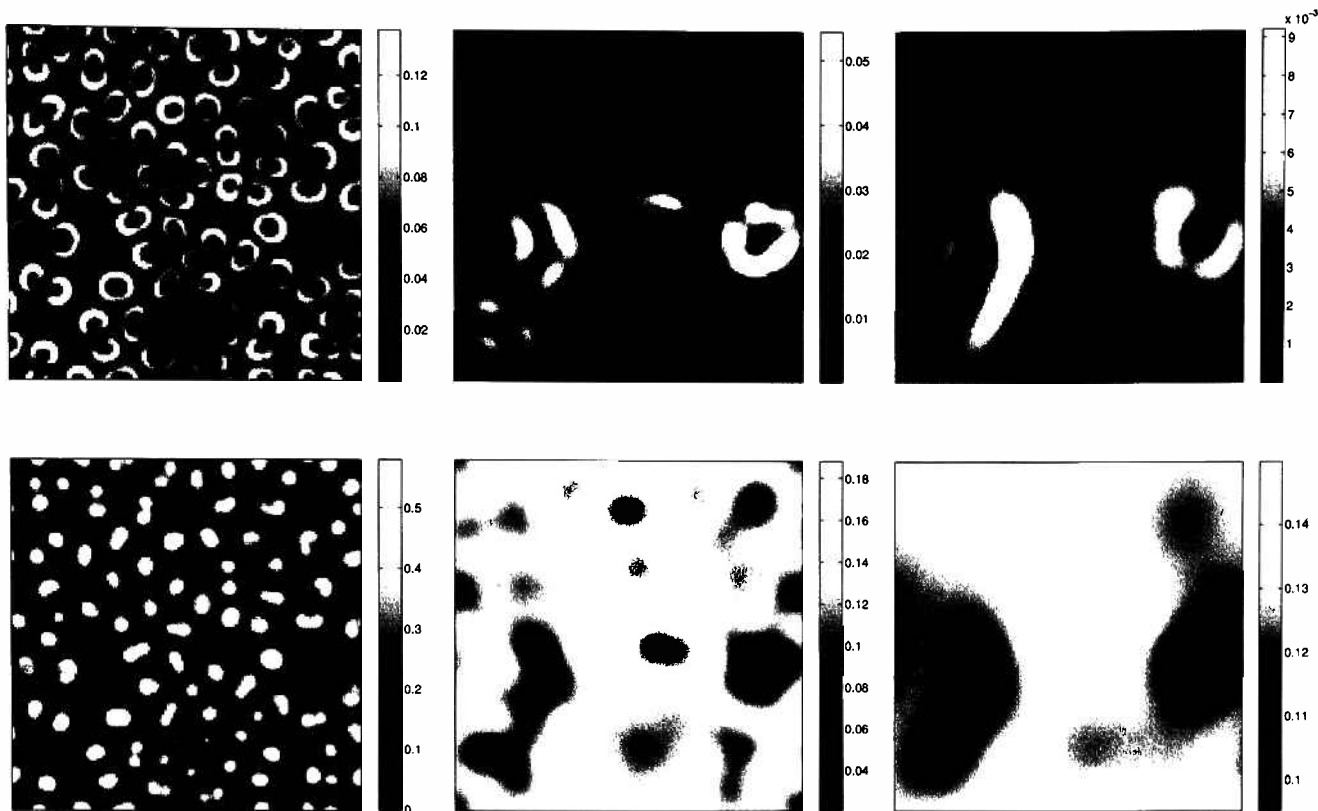


Figure 3: **Top row** The structural information in the system at  $t = 10000$  steps, for three values of the resolution  $r$ ;  $r = 0.01$ ,  $r = 0.05$ , and  $r = 0.1$ . **Bottom row** The information flow  $j_r(r, \mathbf{x}, t)$ . The length of the system is 1,  $D_u = 2D_v = 0.05$ ,  $\hat{D} = 0.02$ ,  $k = 0.058$ .

spatial differences. The opposite effect is that the flow may lead to an increase in chemical information, i.e., the system is pushed away from equilibrium. This is of course what makes it possible to keep the driving information flow for the pattern formation processes in the system. The change in chemical information takes the following form

$$\dot{k}_{\text{chem}}(t) = -j_r(\infty, \mathbf{x}, t) + \sum_i \bar{X}_i \ln \frac{\bar{c}_i}{c_{i0}},$$

where  $\bar{X}_i$  is the average net inflow of component  $i$ . Thus, in a stationary situation, the information flow towards smaller length scales,  $j_r$ , is balanced by the inflow of exergy (or Gibbs free energy) in units of  $k_B T_0$ , represented by the last term.

The negative effect from the diffusion over the system boundary is captured by the sink term  $J$  in the continuity equation. In a diffusion controlled flow,  $X_i$  is given by  $\hat{D}_i(\hat{c}_i - c_i)$ , where  $\hat{D}_i > 0$  and  $\hat{c}_i$  is the concentration outside the system, chosen so that  $\hat{c}_i > c_i$  for inflow and  $\hat{c}_i = 0$  for an outflow of  $i$ , respectively. This results in the following expression for  $J$ ;

$$J(r, \mathbf{x}, t) = - \sum_i \hat{D}_i (c_i + \hat{c}_i) [r \nabla \ln \bar{c}_i]^2 \leq 0,$$

which shows that  $J$  always is a sink term.

We apply the formalism to the pattern formation of the “self-replicating spots” system (Gray and Scott, 1984; Lee et al., 1993; Pearson, 1993),  $U + 2V \rightarrow 3V$  and  $V \rightarrow G$ , with the dynamics

$$\begin{aligned} \dot{c}_u &= D_u \nabla^2 c_u - (c_u - k_{\text{back}} c_v) c_v^2 + \hat{D}(1 - c_u) \\ \dot{c}_v &= D_v \nabla^2 c_v + (c_u - k_{\text{back}} c_v) c_v^2 - k c_v - \hat{D} c_v. \end{aligned}$$

We have introduced a very slow back reaction ( $k_{\text{back}} = 10^{-5}$ ) in order to get the relationship between equilibrium concentrations of  $U$  and  $V$  defined by the reactions. In Figure 2, the dynamics is illustrated starting from an initial state (left) with a square of high concentration of  $V$ . As the system evolves four concentration peaks (spots) emerges from the square, and these spots reproduce by growing and splitting until the system is filled with spots (middle and right). In the process, spots may disappear, which leaves space for other spots to reproduce. In the lower part of the figure, the decomposition of the information in the pattern with respect to scale is plotted for the three snapshots above (at time 0, 1000, and 7000, respectively). It is clear that the initial state has a longer characteristic length as detected by the information density. When the system produces the spots at the

significantly shorter length scale, information is found both at the old length, now due to the size of the cluster (middle), and at the length scale of the spots. When the square distribution has been completely decomposed into spots, no information is left at the initial length scale.

In Figure 3, we show the information density over the system for three different length scales after long time (upper part), and the information flow in scale,  $j_r$ , for the same state (lower part). At low resolution, or large  $r$  (right), the information density is low and captures structures of longer lengths, while at finer resolution, small  $r$  (left), the information density is large and reflects the pattern of spots. Note that each spot is seen as a circle in the information density picture, since the information is sensitive to gradients in the pattern. The information flow in scale,  $j_r$ , is close to homogeneous for large  $r$  (right), but when information moves on to finer scales of resolution, the spatial flow  $\mathbf{j}$  redistributes the flow so that a higher flow  $j_r$  is obtained at the concentration peaks. At finest resolution,  $r = 0$ , this flow leaves the system as entropy production, which is mainly located to the concentration peaks where the chemical activity is high.

In this paper we have presented an information-theoretic perspective on pattern formation in chemical systems. The continuity equation, with the corresponding information flows, connects to thermodynamic flows and thus to thermodynamics restrictions of the system. It is clear that the second law of thermodynamics leads to a destruction of information at the finest levels of resolution. Is there a second law of information-dynamics in thermo-dynamical systems, which makes information flow in the direction of smaller scales? Under what circumstances is this law valid? These are questions for further investigation.

In the Gray-Scott model (Gray and Scott, 1984), we detect an aggregation of information on macroscopic length scales. This illustrates how information theory can be used in order to determine whether spatial structure emerges on length scales much larger than those of the individual components and their interaction. It should be noted that there is also an information flow in the opposite direction (i.e. from micro scale to macro scales) due to noise, which govern the formation of spatial configurations. This flow is several orders of magnitude smaller, but may still control where in space the information from the free energy will be aggregated. The interplay between the driving and the controlling information flows will be investigated in a forthcoming paper.

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