Chemical organization theory: Towards a theory of constructive dynamical systems

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Complex dynamical networks consisting of many components that interact and produce each other are difficult to understand, especially, when new components may appear. In this paper we outline a theory to deal with such systems. The theory consists of two parts. The first part introduces the concept of a chemical organization as a closed and self-maintaining set of components. This concept allows to map a complex (reaction) network to the set of organizations, providing a new view on the system's structure. The second part connects dynamics with the set of organizations, which allows to map a movement of the system in state space to a movement in the set of organizations.

Our world is changing, qualitatively and quantitatively. The characteristics of its dynamics can be as simple as in the case of a friction-less swinging pendulum, or as complex as the dynamical process that results in the creative apparition of novel ideas or entities. We might characterize the nature of a dynamical process according to its level of novelty production. For example, the friction-less swinging pendulum implies a process where the novelty is only quantitative. Whereas the process of biological evolution is highly creative and generates qualitative novelties, which then spread in a quantitative way.

Fontana and Buss (1) called processes and systems that display the production of novelty, constructive (dynamical) processes and constructive (dynamical) systems, respectively. Constructive systems can be found on all levels of scientific abstraction: in nuclear physics, where the collision of atoms or subatomic particles leads to the creation of new particles; in molecular chemistry, where molecules can react to form new molecules; or in social systems, where communication can lead to new communication (2). As a result of a combinatorial explosion, it is easy to create something that is new, e.g., a molecule or a poem that is unique in the whole known universe. Nevertheless, it should be noted that novelty is relative. Whether something is considered to be new or not, depends on what is already there. Thence it follows that whether a system appears as a constructive dynamical system is usually context dependent.

Despite the fact that a large amount of interesting dynamical processes are constructive, classical systems theory does not, conveniently, take novelty into account. Classical systems theory assumes a given static set of components. For example, a classical systems analysis would first identify all components and their relations; then would identify a state space with fixed dimensionality, e.g., the state of our pendulum could be described by a two-dimensional vector specifying angle and angular velocity.

The lack of a theory for constructive dynamical systems has been identified and discussed in detail by Fontana and Buss (1) in the context of a theory for biological organiza-tion. As a solution, they suggested the important concept of a (biological) organization as an operationally closed and dynamically self-maintaining system. Taking this idea further, we suggested to divide the theoretical approach in a static and a dynamic analysis (3). Here, we present the results of our study for general reactions systems, integrating stoichiometry and the concept of mass-maintenance in the definition of organizations, which is a prerequisite for a broad applicability.

Reaction Systems. The theory described herein aims at understanding *reaction systems.* A reaction system consists of reactants, which we refer to as *molecules.* An interaction among molecules that lead to the appearance or disappearance of molecules is called a *reaction.* In particular, reaction systems are used to model chemical processes, but in general, they can be applied to virtually any domain where elements interact to influence the production of other elements, e.g., population dynamics, evolution of language, economy, or social dynamics.

We have to distinguish between a reaction system as an abstract description of all possible molecules (and their reactions), and a reaction vessel, which contains concrete instances of molecules from the set of all possible molecules. In general, the description of a reaction system can be subdivided into three parts: (1) the set of all possible molecules $\mathcal{M}$, (2) the set of all possible reactions among all the possible molecules $\mathcal{R}$, and (3) the dynamics, which describes how the reactions are applied to a collection of molecules inside a reaction vessel.

Static Analysis

Algebraic Chemistry. In the first part of the paper, we are only concerned with the static structure of a reaction system, that is, the molecules and the reactions. And instead of considering a state (e.g., a concentration vector), we limit ourself to the analysis of the set of molecules present in that state. We introduce the concept of an algebraic chemistry, which is a reaction network, including stoichiometric information, from which we will derive the organizational structure of the system.

Definition 1 Given a set $\mathcal{M}$ of elements (called molecules) and a set of reaction rules given by the relation $\mathcal{R}$: $\mathcal{P}_M(\mathcal{M}) \times \mathcal{P}_M(\mathcal{M})$. We call the pair $\langle \mathcal{M}, \mathcal{R} \rangle$ an *algebraic chemistry.*

$\mathcal{P}_M(C)$ is the set of all multisets with elements from C. A multiset differs from a set in the fact that the same element can appear more than one time. The frequency of occurrence of an element a in a multiset A is denoted by $\#(a \in A)$. For simplicity, we adopt a notion from chemistry to write reaction rules. Instead of

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writing $(\{s_1, s_2, \ldots, s_n\}, \{s'_1, s'_2, \ldots, s'_{n'}\}) \in \mathcal{R}$ we write: $s_1 + s_2 + \cdots + s_n \to s'_1 + s'_2 + \cdots + s'_{n'}$. Given the left hand side molecules $A = \{s_1, s_2, \ldots, s_n\}$ and the right hand side molecules $B = \{s'_1, s'_2, \ldots, s'_{n'}\}$, we write $(A \to B) \in \mathcal{R}$ instead of $(A, B) \in \mathcal{R}$. $A \to B$ represents a chemical reaction equation where A is the multiset of molecules on the left hand side (also called *reactants*) and B the multiset of molecules on the right hand side (also called *products*).

Input and Output. There are many processes that give rise to an inflow and outflow, such as, incident sunlight, decaying molecules, or a general dilution flow. In this paper we handle input simply by adding a reaction rule $\emptyset \to a$ to the reaction rules $\mathcal{R}$ for every molecule a that is in the inflow. $\emptyset$ denotes the empty set. Equivalently, for an output molecule a (e.g., a molecule that is decaying) we add the rule $a \to \emptyset$ to the reaction rules $\mathcal{R}$.

Semi-Organization. In classical analysis, we study the movement of the system in state space. Instead, here, we consider the movement from one set of molecules to another. As in the classical analysis of the dynamic of the system, where fixed points and attractors are considered more important than other states, some sets of molecules are more important than others. In order to find those sets, we introduce some properties that define them, namely: closure, self-maintenance, semi-organization, mass-maintenance, and finally the organization.

All definitions herein refer to an algebraic chemistry $\langle \mathcal{M}, \mathcal{R} \rangle$. The first requirement, called closure, assures that a set of molecules contains all molecules that can be produced by reactions among those molecules.
Definition 2 A set $C \subseteq \mathcal{M}$ is *closed*, if for all reactions $(A \to B) \in \mathcal{R}$, with A a multiset of elements in C ($A \in \mathcal{P}_M(C)$), B is also a multiset of elements in C ($B \in \mathcal{P}_M(C)$).

Given a set $S \subseteq \mathcal{M}$, we can always generate its closure $G_{CL}(S)$ according to the following definition:
Definition 3 Given a set of molecules $S \subseteq \mathcal{M}$, we define $G_{CL}(S)$ as the smallest closed set C containing S. We say that S *generates the closed set* $C = G_{CL}(S)$ and we call C the *closure* of S.

The closure implies a union and an intersection operator on the closed sets $\mathcal{O}_{CL}$ of an algebraic chemistry. Given two closed sets U and V, the closed set generated by their union ($U \sqcup_{CL} V$) and intersection ($U \sqcap_{CL} V$) is defined as: $U \sqcup_{CL} V \equiv G_{CL}(U \cup V)$, and $U \sqcap_{CL} V \equiv G_{CL}(U \cap V)$, respectively. Trivially, closed sets form a lattice $\langle \mathcal{O}_{CL}, \sqcup_{CL}, \sqcap_{CL} \rangle$, which is a common algebraic structure (a poset in which any two elements have a greatest lower bound and a least upper bound, see supl. mat.). The property of closure is important, because the closed set represents the largest possible set that can be reached from a given set of molecules. Furthermore a set that is closed cannot generate new molecules and is in that sense more stable than a set that is not closed.

The next important property, called self-maintenance, assures that every molecule that is used-up within a set, is produced within that set.
Definition 4 A set of molecules $S \subseteq \mathcal{M}$ is called *semi self-maintaining*, if all molecules $s \in S$ that are used-up within S are also produced within that set S.
More precisely, we say that a molecule $k \in \mathcal{M}$ is *produced* within a set $C \subseteq \mathcal{M}$, if there exists a reaction $(A \to B)$, with $A \in \mathcal{P}_M(C)$, and $\#(k \in A) < \#(k \in B)$. In the same way, we say that a molecule $k \in C$ is *used-up* within the set C, if, within the set C, there is a reaction $(A \to B)$ with $A \in \mathcal{P}_M(C)$, and $\#(k \in A) > \#(k \in B)$.

Taking closure and self-maintenance together, we arrive at the important concept of a semi-organization.
Definition 5 A semi-organization $O \subseteq \mathcal{M}$ is a set of molecules that is closed and semi self-maintaining.

Organization. In a semi-organization, all molecules that are used-up are produced; but, this does not guarantee that the total amount of mass can be maintained. A small, but important example is the reversible reaction in a flow reactor: $\mathcal{M} = \{a, b\}, \mathcal{R} = \{a \to b, b \to a, a \to \emptyset, b \to \emptyset\}$, where both molecules also decay. $O = \{a, b\}$ is a semi-organization, because the set is closed, a is produced by the reaction $b \to a$, and b is produced by the reaction $a \to b$. But, obviously, the system $\{a, b\}$ is not stable, because both molecules decay and are not sufficiently reproduced, so that they will finally vanish. The solution to this problem is to consider the overall ability of a set to maintain its total mass. We call such sets self-maintaining:

Definition 6 Given an algebraic chemistry $\langle \mathcal{M}, \mathcal{R} \rangle$ with $m = |\mathcal{M}|$ molecules and $n = |\mathcal{R}|$ reactions, and let $\mathbf{M} = (m_{i,j})$ be the ($m \times n$) stoichiometric matrix implied by the reaction rules $\mathcal{R}$, where $m_{i,j}$ denotes the number of molecules of type i produced in reaction j. A set of molecules $C \subseteq \mathcal{M}$ is called *self-maintaining*, if there exists a flux vector $\mathbf{v} \in \mathbb{R}^n$ such that the three following condition apply: (1) for all reactions $(A \to B)$ with $A \in \mathcal{P}_M(C)$ the flux $v_{(A \to B)} > 0$; (2) for all reactions $(A \to B)$ with $A \notin \mathcal{P}_M(C)$, $v_{(A \to B)} = 0$; and (3) for all molecules $i \in C$, $f_i \geq 0$ with $(f_1, \ldots, f_m) = \mathbf{Mv}$.

$v_{(A \to B)}$ denotes the element of $\mathbf{v}$ describing the flux of reaction $A \to B$. For the example above, the stoichiometric matrix becomes $\mathbf{M} = ((-1, 1), (1, -1), (-1, 0), (0, -1))$, and we can see that there is no positive flux vector $\mathbf{v} \in \mathbb{R}^4$, such that $\mathbf{Mv} \geq \mathbf{0}$. In fact, only the empty semi-organization $\{\}$ is self-maintaining. In case a and b would not decay, $\mathcal{R} = \{a \to b, b \to a\}$, the set $\{a, b\}$ would be (as desired) mass-mainataing, because threre is a flux vector, e.g., $\mathbf{v} = (1.0, 1.0)$, such that $\mathbf{Mv} = \mathbf{0} \geq \mathbf{0}$ with $\mathbf{M} = ((-1, 1), (1, -1))$. Now, closure and mass-maintenance together lead to the central definition of this work:

Definition 7 A set of molecules $O \subseteq \mathcal{M}$ that is closed and self-maintaining is called an *organization*.

An organization represents an important combination of molecular types, which are likely to be observed in a reaction vessel on the long run. A set of molecules that is not closed or not self-maintaining would not exists for long, because new molecules can appear or some molecules would vanish, respectively. The condition "self-maintaining" is stronger and more difficult to compute than the condition "semi self-maintaining". In fact, the former implies the latter, because in a mass-maintaing set, a molecule i that is used-up within that set must also be produced within that set in order to achieve a non-negative production rate f_i. Thus we can say:

Lemma 1 Every organization is a semi-organization.

Example 1 In order to illustrate the new concepts, we consider a small example where there are just four molecular species $\mathcal{M} = \{a, b, c, d\}$, which react according to the following reaction rules $\mathcal{R} = \{a + b \to a + 2b, a + d \to a + 2d, b + c \to 2c, c \to b, b + d \to c, b \to \emptyset, c \to \emptyset, d \to \emptyset\}$, where b, c, d decay spontaneously and a is a permanent catalyst. Although the reaction system is small, its organizational structure is already difficult to see when looking at the rules or their graphical representation (Fig. 1). In Fig. 1, all 16 possible sets of molecules are shown as a lattice. There are 9 closed sets, 8 semi self-maintaining sets, 7 self-maintaining sets, and 7 semi-organizations, 6 of which are organizations. The organizations are the only combination of molecules that can reside in a reaction vessel for a long time. We can also see immediately that a reactor containing $\{b, c, d\}$ must have a transient dynamics "down" where molecular types are lost, and that a reactor containing $\{a, b, d\}$ will have a transient where a new molecular type will appear, and so on...
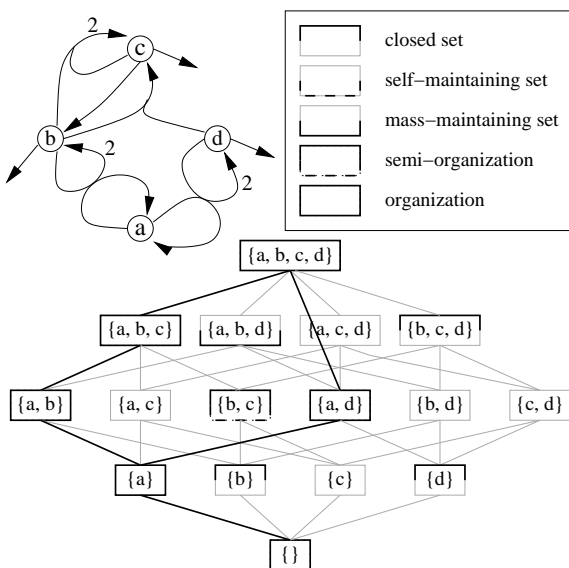
Figure 1: Example with four species. Reaction network (a "2" means that two molecules are produced) and graphical representation of the lattice of all sets. The vertical position is determined by the set size. The solid lines depict the lattice of organizations.

Different reaction systems

Finding all organizations of a general reaction system appears to be computationally difficult. One approach is to find the semi-organizations first, and then check, which of them are also self-maintaining. The property of the set of organizations and semi-organizations depends strongly on the type of system studied. We will present here four types of systems and analyze their properties with respect to the newly introduced concepts.

Catalytic Flow System . In a *catalytic flow system* all molecules are used-up by first-order reactions of the form $\{k\} \to \emptyset$ (dilution) and there is no molecule used-up by any other reaction. So, each molecule k decays spontaneously, or equivalently, is removed by a dilution flow. Apart from this, each molecule can appear only as a catalyst (without being used-up). Examples of *catalytic flow system* are the replicator equation(4), the hypercycle (5), the more general *catalytic network equation* (6), or AlChemy (7). Furthermore some models of genetic regulatory networks and social system (8) are *catalytic flow systems* .
Lemma 2 In a *catalytic flow system* , every semin-organization is an organization.

In a *catalytic flow system* we can easily check, whether a set O is an organization by just checking whether it is closed and whether each molecule in that set is produced by that set. Furthermore, given a set A, we can always generate an organization by adding all molecules produced by A until A is closed and than removing molecules that are not produced until A is semi self-maintaining. With respect to the intersection and union of (semi-)organizations the set of all (semi-) organizations of a *catalytic flow system* forms an algebraic lattice (see below), which has already been noted by Fontana and Buss (1).

Reactive Flow System . In a *reactive flow system* all molecules are used-up by first-order reactions of the form $\{k\} \to \emptyset$ (dilution). But as opposed to the previous system, we allow arbitrary additional reactions in $\mathcal{R}$. This is a typical situation for chemical flow reactors or bacteria

that grow (9). In a *reactive flow system* , semi-organizations are not necessarily organizations, as in the reversible reaction example shown before. Nevertheless, both, the semi-organizations and the organizations form a lattice $\langle \mathcal{O}, \sqcup, \sqcap \rangle$, Moreover, the union ($\sqcup$) and intersection ($\sqcap$) of any two organizations is an organization (see below).

Consistent Reaction System . In a *consistent reaction system* there are two types of molecules: persistent molecules P and non-persistent molecules. All non-persistent molecules $k \in \mathcal{M} \setminus P$ are used-up (as in the two systems before) by first-order reactions of the form $\{k\} \to B$ with $k \notin B$; whereas a persistent molecule $p \in P$ is not used-up by any reaction at all. An example of a *consistent reaction system* is Example 2, where a is a persistent molecule. The *consistent reaction system* is the most general of the four systems where the semi-organizations and organizations always form a lattice, and where the generate organization operator can properly be defined (see below). As in a *reactive flow system* , not all semi-organizations are organizations.

General Reaction System . A *general reaction system* consists of arbitrary reactions. Examples are planetary atmosphere chemistries (10). In a *general reaction system* , neither the set of organization nor the set of semi-organizations necessarily form a lattice. Because of the lack of properties there is an actual difficulty in studying those systems. The analysis gets much easier, if we are able to transform the system into something else, e.g., a *consistent reaction system* . This could be done by introducing a small outflow for each molecule.

Common Properties of Consistent Reaction System
Consistent reaction systems (including *reactive flow system* and *catalytic flow system*) possess some comfortable properties that allow us to present a series of useful definitions and lemmas. In a *consistent reaction system* , given a set of molecules C, we can uniquely *generate* a semi self-maintaining set, a semi-organization, a self-maintaining set, and an organization in a similar way as we have generated a closed set. And like for closed sets, we can define the union and intersection on semi self-maintaining sets, semi-organizations, self-maintaining sets, and organizations, respectively. Furthermore, each, the semi self-maintaining sets, semi-organizations, self-maintaining sets, and organizations form a lattice together with their respective union and intersection operators. This does not generalize to *general reaction system* , because for a *general reaction system* we cannot uniquely generate a semi self-maintaining set, a semi-organization, a self-maintaining set, nor an organization as in the case of a *consistent reaction system* .
Definition 8 Given a set of molecules $C \subseteq \mathcal{M}$, we define $G_{SSM}(C)$ as the biggest semi self-maintaining set S contained in C. We say that C *generates the semi self-maintaining set* $S = G_{SSM}(C)$.

In order to calculate the semi self-maintaining set generated by C, we remove those molecules that are used-up and not produced within C, until all molecules used-up are also produced, and thus reaching a semi self-maintaining set. The operator G_{SSM} (generate semi self-maintaining set) implies the union $\sqcup_{SSM}$ and intersection $\sqcap_{SSM}$ on semi self-maintaining sets: Given two semi self-maintaining sets S_1 and S_2, the semi self-maintaining sets generated by their union ($S_1 \sqcup_{CL} S_2$) and intersection ($S_1 \sqcap_{CL} S_2$) are defined as: $S_1 \sqcup_{SSM} S_2 \equiv G_{SSM}(S_1 \cup S_2)$, and $S_1 \sqcap_{SSM} S_2 \equiv G_{SSM}(S_1 \cap S_2)$, respectively. And as already mentioned, in a *consistent reaction system* , $\langle \mathcal{O}_{SSM}, \sqcup_{SSM}, \sqcap_{SSM} \rangle$ forms a lattice, where $\mathcal{O}_{SSM}$ is the set of all semi self-maintaining sets of an algebraic chemistry. Finally note that, if S

is semi self-maintaining, its closure $G_{CL}(S)$ is semi self-maintaining, too (in *consistent reaction systems*).

There are many ways in which we can generate a semi-organization from a set. We will present here the simplest one, which implicitly assumes that molecules are produced quickly and vanish slowly. This assumption leads to the largest possible semi-organization generated by a set:

Definition 9 Given a set of molecules $C \subseteq \mathcal{M}$, we define $G_{SO}(C)$ as $G_{SSM}(G_{CL}(S))$. We say that C *generates the semi-organization* $O = G_{SO}(C)$.

In the same way as before, the generate semi-organization operator $G_{SO}(C)$ implies the union $\sqcup_{SO}$ and intersection $\sqcap_{SO}$ on semi-organizations, namely $O_1 \sqcup_{SO} O_2 \equiv G_{SO}(O_1 \cup O_2)$, and $O_1 \sqcap_{SO} O_2 \equiv G_{SO}(O_1 \cap O_2)$, respectively, which implies the lattice of semi-organizations.

Definition 10 Given a set of molecules $C \subseteq \mathcal{M}$, we define $G_{SM}(C)$ as the biggest self-maintaining set S' contained in C. We say that C *generates the self-maintaining set* $S = G_{SM}(C)$.

For *consistent reaction systems* , $G_{SM}(C)$ is always defined, because the union $(\cup)$ of two self-maintaining sets is self-maintaining; and further, every set is either self-maintaining, or it contains a unique biggest self-maintaining set. Thus from every set we can generate a self-maintaining set. Note that self-maintaining sets are also semi self-maintaining, $G_{SM}(G_{SSM}(S)) \equiv G_{SM}(S)$, which is a useful property, because $G_{SSM}(S)$ is easier to compute. As usual, the union $\sqcup_{SM}$ and intersection $\sqcap_{SM}$ of self-maintaining sets S_1, S_2 are defined as $S_1 \sqcup_{SM} S_2 \equiv G_{SM}(S_1 \cup S_2)$, $S_1 \sqcap_{SM} S_2 \equiv G_{SM}(S_1 \cap S_2)$, respectively. Thus also the set of all self-maintaining sets $\mathcal{O}_{SM}$ forms a lattice $\langle \mathcal{O}_{SM}, \sqcup_{SM}, \sqcap_{SM} \rangle$. If S is self-maintaining, its closure $G_{CL}(S)$ is self-maintaining, too (again, not valid for *general reaction systems*).

Finally, in *consistent reaction systems* , we can also generate uniquely an organization, (here, again, the largest organization that can be generated from a set) according to the following definition:

Definition 11 Given a set of molecules $C \subseteq \mathcal{M}$, we define $G(C)$ as $G_{SM}(G_{CL}(C))$. We say that C *generates the organization* $O = G(C)$.

Equivalently $G(C) = G_{SM}(G_{SSM}(G_{CL}(C)))$, which allows to compute the organization generated by a set more easily in three steps. Following the same scheme as before, the union $\sqcup$ and intersection $\sqcap$ of two organizations U and V is defined as the organization generated by their set-union and set-intersection: $U \sqcup V \equiv G(U \cup V)$, $U \sqcap V \equiv G(U \cap V)$, respectively. Thus, for *consistent reaction system* , also the set of all organizations $\mathcal{O}$ forms a lattice $\langle \mathcal{O}, \sqcup, \sqcap \rangle$. This important fact should be emphasized by the following lemma:

Lemma 3 Given an algebraic chemistry $\langle \mathcal{M}, \mathcal{R} \rangle$ of a *consistent reaction system* and all its organizations $\mathcal{O}$, then $\langle \mathcal{O}, \sqcup, \sqcap \rangle$ is a lattice.

Knowing that the semi-organizations and organizations form a lattice, and that we can uniquely generate an organization for every set, is a useful information. In order to find the whole set of organizations, it is impractical just to check all the possible sets of molecules. Instead, we can start by computing the lattice of semi-organizations, and then test only those sets for mass-maintenance. Furthermore, if the semi-organizations form a lattice, we can start with small sets of molecules and generate their semi-organizations, while the $\sqcup_{SO}$ operator can lead us to the more complex semi-organizations.

As a summary, from a practical point of view, we calculate first the set of semi-organizations. If our system is a *catalytic flow system* , we automatically obtain the lattice of organizations. Otherwise we have to check for each semi-organization whether it is self-maintaining or not. If we have a *consistent reaction system* , then we are assured to obtain a lattice, where there is a unique smallest and largest organization, and where we can easily obtain the intersection

and union of two organizations from the graphical representation of the lattice (see examples). For a *general reaction system* , the set of organizations does not necessarily form a lattice. Nevertheless, this set of organizations represent the *organizational structure* of the reaction network, which can be visualized and which can provide a new view on the dynamics of the system by mapping the movement of the system in state space to a movement in the set of organizations, as will be shown in the following section.

Dynamic Analysis

The static theory deals with molecules $\mathcal{M}$ and their reaction rules $\mathcal{R}$, but not with the evolution of the system in time. To add dynamics to the theory, we have to formalize the dynamics of a system. In a very general approach, the *dynamics* is given by a *state space* X and a formal definition (mathematical or algorithmical) that describes all possible movements in X. Given an initial state $\mathbf{x}_0 \in X$, the formal definition describes how the state changes over time. For simplicity, we assume a deterministic dynamical process, which can be formalized by a phase flow $(X, (T_t)_{t \in \mathbb{R}})$ where $(T_t)_{t \in \mathbb{R}}$ is a one-parametric group of transformations from X. $T_t(\mathbf{x}_0)$ denotes the state at time t of a system that has been in state $\mathbf{x}_0$ at $t = 0$.

Connecting to the Static Theory A state $\mathbf{x} \in X$ represents the state of a reaction vessel that contains molecules from $\mathcal{M}$. In the static part of the theory we consider just the set of molecular species present in the reaction vessel, but not their concentrations, spatial distributions, velocities, and so on.

Now, given the state $\mathbf{x}$ of the reaction vessel, we need a function that maps uniquely this state to the set of molecules present. Vice versa, given a set of molecules $A \subseteq \mathcal{M}$, we need to know, which states from X correspond to this set of molecules. For this reason we introduce a mapping ϕ called *abstraction*, from X to $\mathcal{M}$, which maps a state of the system to the set of molecules that are present in the system being in that state. We require this mapping to be monotonically increasing on the number of molecules. In other words, if we have two states $\mathbf{x}_1$ and $\mathbf{x}_2$, and the concentration of the molecule m_1 is higher in $\mathbf{x}_2$ than in $\mathbf{x}_1$; and if $m_1 \in \phi(\mathbf{x}_1)$ then $m_1 \in \phi(\mathbf{x}_2)$. The exact mapping can be defined precisely later, depending on the state space, on the dynamics, and on the actual application.

The concept of *instance* is the opposite of the concept of abstraction. While $\phi(\mathbf{x})$ denotes the molecules represented by the state $\mathbf{x}$, an instance $\mathbf{x}$ of a set A is a state where exactly the molecules from A are present according to the function ϕ.

Definition 12 We say that a state $\mathbf{x} \in X$ is an instance of $A \subseteq \mathcal{M}$, iff $\phi(\mathbf{x}) = A$

In particular, we can define an instance of an organization O (if $\phi(x) = O$) and an instance of a generator of O (if $G(\phi(x)) = O$). Loosely speaking we can say that $\mathbf{x}$ *generates organization* O. Note that a state $\mathbf{x}$ of a *consistent reaction system* , *reactive flow system* , and *catalytic flow system* is *always* an instance of a generator of one and only one organization O. This leads to the important observation that a lattice of organizations partitions the state space X, where a partition X_O implied by organization O is defined as the set of all instance of all generators of O: $X_O = \{\mathbf{x} \in X | G(\phi(\mathbf{x})) = O\}$. Note that as the system state evolves over time, the organization $G(\phi(\mathbf{x}(t)))$ generated by $\mathbf{x}(t)$ might change (see below, Fig. 2 and 3).

Fixed Points are Instances of Organizations. Now we describe a theorem that relates fixed points to organizations, and by doing so, underlining the relevancy of organizations. We will show that, given an ODE of a form that is commonly used to describe the dynamics of reaction systems, every fixed point of this ODE is an instance of an organization. We therefore assume in this section that $\mathbf{x}$ is a concentration vector $\mathbf{x} = (x_1, x_2, \ldots, x_{|\mathcal{M}|})$, $X = \mathbb{R}^{|\mathcal{M}|}, x_i \geq 0$ where x_i denotes the concentration of molecular type i in the reaction vessel, and $\mathcal{M}$ is finite. The dynamics is given by an ODE of the form $\dot{\mathbf{x}} = \mathbf{M}\mathbf{v}(\mathbf{x})$ where $\mathbf{M}$ is the stoichiometric matrix implied by the algebraic chemistry $\langle \mathcal{M}, \mathcal{R} \rangle$ (reaction rules). $\mathbf{v}(\mathbf{x}) = (v_1(\mathbf{x}), \ldots, v_n(\mathbf{x})) \in \mathbb{R}^{|\mathcal{R}|}$ is a flux vector depending on the current concentration $\mathbf{x}$, where $|\mathcal{R}|$ denotes the number of reaction rules. A flux $v_j(\mathbf{x})$ describes the rate of a particular reaction j. For the function v_j we require only that $v_j(\mathbf{x})$ is positive, if and only if the molecules on the left hand side of the reaction j are present in the state $\mathbf{x}$, and otherwise it must be zero. Often it is also assumed that $v_j(\mathbf{x})$ increases monotonously, but this is not required here. Given the dynamical system as $\dot{\mathbf{x}} = \mathbf{M}\mathbf{v}(\mathbf{x})$, we can define the abstraction of a state $\mathbf{x}$ formally by using a (small) threshold $\Theta \geq 0$ such that all fixed points have positive coordinates greater than Θ.

Definition 13 Given a dynamical system $\dot{\mathbf{x}} = f(\mathbf{x})$ and let $\mathbf{x}$ be a state in X, then the abstraction $\phi(\mathbf{x})$ is defined by

$$\phi(\mathbf{x}) = \{i | x_i > \Theta, i \in \mathcal{M}\}, \quad \phi : X \to \mathcal{P}(\mathcal{M}), \quad \Theta \geq 0 \quad (1)$$

where x_i is the concentration of molecular type i in state $\mathbf{x}$, and Θ is a threshold chosen such that it is smaller than any positive coordinate of any fixed point of $\dot{\mathbf{x}} = f(\mathbf{x}), x_i \geq 0$.

Setting $\Theta = 0$ is a safe choice, because in this case ϕ always meets the definition above. But for practical reasons, it makes often sense to apply a positive threshold greater zero, e.g., when we take into consideration that the number of molecules in a reaction vessel is finite.

Theorem 1 **Hypothesis:** Let us consider a *general reaction system* whose reaction network is given by the algebraic chemistry $\langle \mathcal{M}, \mathcal{R} \rangle$ and whose dynamics is given by $\dot{\mathbf{x}} = \mathbf{M}\mathbf{v}(\mathbf{x}) = f(\mathbf{x})$ as defined before. Let $\mathbf{x}' \in X$ be a fixed point, that is, $f(\mathbf{x}') = \mathbf{0}$, and let us consider a mapping ϕ as given by Def. 13, which assigns a set of molecules to each state $\mathbf{x}$.

Thesis: $\phi(\mathbf{x}')$ is an organization.

Proof: We need to prove that $\phi(\mathbf{x}')$ is closed and self-maintaining: (a) Closure: Let us assume that $\phi(\mathbf{x}')$ is not closed, then there exist a molecule k such that $k \notin \phi(\mathbf{x}')$ and k is generated by molecules contained in $\phi(\mathbf{x}')$. Since, $\mathbf{x}'$ is a fixed point, $f(\mathbf{x}') = \mathbf{M}\mathbf{v}(\mathbf{x}') = 0$. Now we decompose the stoichiometric matrix $\mathbf{M}$ into two matrices $\mathbf{M}^+$ and $\mathbf{M}^-$ separating all positive from the negative coefficients, respectively, such that $\mathbf{M} = \mathbf{M}^+ + \mathbf{M}^-$ and $\mathbf{M}^+\mathbf{v}(\mathbf{x}') \geq \mathbf{0}$, and $\mathbf{M}^-\mathbf{v}(\mathbf{x}') \leq \mathbf{0}$ (note that $\mathbf{v}(\mathbf{x}')$ is always non-negative by definition). Let $\dot{x}_k^+$ and $\dot{x}_k^-$ be the k-th row of $\mathbf{M}^+\mathbf{v}'(\mathbf{x})$ and $\mathbf{M}^-\mathbf{v}'(\mathbf{x})$, respectively, which represent the inflow (production) and outflow (destruction) of molecules of type k. Note that $\dot{x}_k^+ + \dot{x}_k^- = \dot{x}_k' = 0$ (fixed point condition). Since we assumed that k is produced by molecules from $\phi(\mathbf{x}')$, $\dot{x}_k^+$ must be positive, $\dot{x}_k^+ > 0$ and thus $\dot{x}_k^- < 0$. But this leads to a contradiction. In order to show this we have to differentiate the following two cases: (i) Assume that $x_k' = 0$, then $\dot{x}_k^-$ must be zero, too (by definition; and intuitively, because a molecule not present cannot vanish). (ii) Assume that $0 < x_k' \leq \Theta$ ($x_k' > \Theta$ needs not to be considered, because in that case k would be contained in $\phi(\mathbf{x}')$.). Because

we assumed that x_k' is a coordinate of a fixed point, Θ must be smaller than x_k', which is again a contradiction.

(b) self-maintaining: We have to show that $\phi(\mathbf{x}')$ is self-maintaining. Since $\mathbf{x}'$ is a fixed point $\mathbf{M}\mathbf{v}(\mathbf{x}') = \mathbf{0}$, which fulfills condition (3) of the definition of self-maintaining (Def. 6). From the requirements for the flux vector $\mathbf{v}$, it follows directly that $v_{(A \to B)}(\mathbf{x}') > 0$ for all $A \in \mathcal{P}_M(\phi(x'))$, which fulfills condition (1) of Def. 6. Following the same contradictory argument as before in (ii), x_k' must be zero for $k \notin \phi(\mathbf{x})$, and therefore $v_{(A \to B)}(\mathbf{x}') = 0$ for $A \notin \mathcal{P}_M(\phi(\mathbf{x}))$, which fulfills the remaining condition (2) of self-maintaining. q.e.d.

From this theorem it follows immediately that a fixed point is an instance of a closed set, a semi self-maintaining set, and of a semi-organization. Let us finally mention that even if each fixed point is an instance of an organization, an organization does not necessarily possess a fixed point. Further note that given an attractor $A \subseteq X$, there exists an organization O such that all points of $\bar{A}$ are instances of a generator of O. In fact, it might be natural to suppose that all points of an attractor are actually instances of O, yet it is not clear if this is true for all systems or just for some.

Movement from Organization to Organization

ODEs and Movement in the Set of Organizations Not all system can be studied using ODEs. In particular a discrete system is usually only *approximated* by an ODE. In a discrete dynamical system, the molecular types that are present in the reaction vessel can change in time, e.g, as the last molecule of a certain type vanishes. In an ODE instead, this does not generally happen, where molecules can *tend to* zero as time *tends to* infinity. So, even if in reality a molecule disappears, in an ODE model it might still be present in a tiny quantity. The fact that every molecule ends up being present in (at least) a tiny quantity, generally precludes us to notice that the system is actually moving from a state where some molecules are present, to another state where a different set of molecules is present. Yet this is what happens in reality, and in this respect, an ODE is a poor approximation of reality. A common approach to overcome this problem is to introduce a concentration threshold Θ, below which a molecular type is considered not to be present. We use this threshold in order to define the abstraction ϕ, which just returns the set of molecules present in a certain state. Additionally, we might use the threshold to manipulate the numerical integration of an ODE by setting a concentration to zero, when it falls below the threshold. In this case, a constructive perturbation has to be greater than this threshold.

Downward Movement Not all organizations are stable. The fact that there exits a flux vector, such that no molecule of that organization vanishes, does not imply that this flux vector can be realized when taking dynamics into account. As a result a molecular type can disappear. Each molecular type that disappears simplifies the system. Some molecules can be generated back. But eventually the system can move from a state that generates organization O_1 into a state that generates organization O_2, with O_2 always below O_1 ($O_2 \subset O_1$). We call this spontaneous movement a *downward movement*.

Upward Movement Moving up to an organization above requires that a new molecular type appears in the system. This new molecular type cannot be produced by a reaction among present molecules (condition of closure). Thus moving to an organization above is more complicated then the movement down and requires a couple of specifications that

describe how new molecular types enter the system. Here we assume that new molecular types appear by some sort of random perturbations or purposeful interference. We assume that a small quantity of molecules of that new molecular type (or a set of molecular types) suddenly appears. Often, in practice, the perturbation (appearance of new molecular types) has a much slower time scale than the internal dynamics (e.g., chemical reaction kinetics) of the system.

Example 2 Assume a system with two molecular species $\mathcal{M} = \{a, b\}$ and the reactions $\mathcal{R} = \{a \rightarrow 2a, b \rightarrow 2b, a + b \rightarrow a, a \rightarrow, b \rightarrow\}$. All combinations of molecules are organizations, thus there are four organizations (Fig. 2). Assume further that the dynamics is governed by the ODE $\dot{x}_1 = x_1 - x_1^2, \dot{x}_2 = x_2 - x_2^2 - x_1 x_2$ where x_1 and x_2 denote the concentration of species a and b, respectively. We map a state to a set of molecules by using a small, positive threshold $\Theta = 0.1 > 0$ (Def. 13). Now, assume that the system is in state $\mathbf{x}_0 = (0, 1)$ thus in organization $\{b\}$. If a small quantity of a appears (constructive perturbation), the amount of a will grow and b will tend to zero. The system will move upward to organization $\{a, b\}$ in a transient phase, while finally moving down and converging to a fixed point in organization $\{a\}$. This movement can now be visualized in the lattice of organizations as shown in Fig. 2.
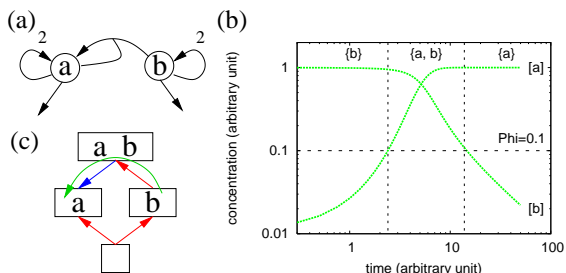

Figure 2: *Example of an up-movement caused by a constructive perturbation, followed by a down-movement. (a) reaction network, (b) concentration vs. time plot of a trajectory, (c) lattice of organizations including trajectory.*

Visualizing Possible Movements in the Set of Organizations In order to display potential movements in the lattice or set of organizations, we can draw links between organizations. As exemplified in Fig. 2 and Fig. 3, these links can indicate possible downward movements (down-link, blue) or upward movements (up-link, red). A neutral link (black line) denotes that neither the system can move spontaneously down, nor can a constructive perturbation move the system up. Whether the latter is true depends on the definition of "constructive perturbation" applied. For the example of Fig. 3 we defined a constructive perturbation as inserting a small quantity of *one* new molecular type.

The dynamics in between organizations is more complex than this intuitive presentation might suggest, for example in some cases it is possible to move from one organization O_1 to an organization O_2, with O_2 above (or below) O_1 without passing through the organizations in between O_1 and O_2. In Fig. 3 this is the case for an upward movement from organization $\{a\}$ to organization $\{a, b, c\}$ caused by a constructive perturbation where a small quantity of c has been inserted.
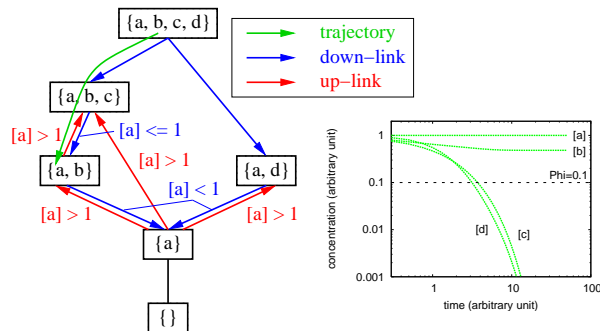

Figure 3: *Lattice of organizations of Example 1, including up-links and down-links. Furthermore a trajectory is shown starting in organization $\{a, b, c, d\}$ moving down to organization $\{a, b\}$.*

Organizations in Real Systems In preliminary studies (11) we have shown that artificial chemical reaction networks that are based on a structure-to-function mapping (e.g., ref. (1; 12)) possess a more complex lattice of organization than networks created randomly. From this observation we can already expect that natural networks possess non-trivial organization structures. Our investigation of planetary photo-chemistries (10) and bacterial metabolism (which will be published elsewhere), revealed lattices of organizations that vanish when the networks are randomized, indicating a non-trivial structure. Here, in order to give an impression, we show a lattice of organizations obtained from a model of the central sugar metabolism of *E.coli* by Puchalka and Kierzek (9). The model consists of 92 species and 197 reactions, including gene expression, signal transduction, transport, and enzymatic activities. Figure 4 shows the lattice of organizations resulting from the original model, ignoring inhibiting interactions. The smallest organization, O_1, contains 76 molecules including the glucose metabolism and all input molecules. The input molecules, chosen according to ref. (9), include the external food set (Glcex, Glyex, Lacex) and all promoters (see suppl. material). Two other organizations, O_3 and O_4, contain the Lactose and Glycerol metabolism, respectively. Their union results in the largest organization O_5 that contains all molecules. In summary, we can conclude, that the organizations found are biological meaningful, indicating a promising potential for future applications of this theory.
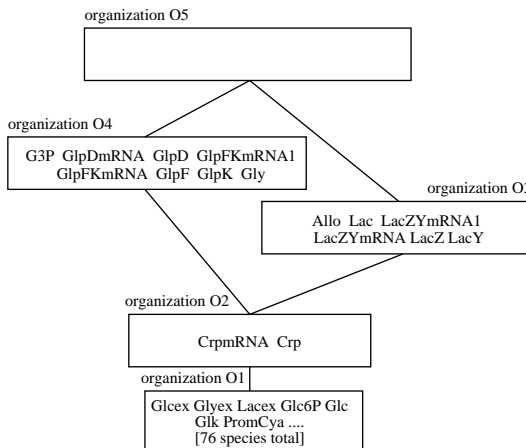

Figure 4: *Lattice of organizations of a model of the central sugar metabolism of E.coli. (9). In an organization, only names of new molecular species are printed that are not present in an organization below.*

Conclusion We presented a general way to define organizations in reaction systems, and proved a theorem that showed that fixed points are instances of those organizations. We also investigated the relative structure of the organizations, opening up the door to visualize the dynamical movement of the system through organizations. Much work still needs to be done, and we believe that this line of research is just in its infancy. Reaction systems with inhibiting interactions need to be investigated, as well as the structure inside organizations (i.e. the relation between attractive states and organizations). In fact the whole issue of movement among sets of molecules could only be introduced here. The concepts of attractive and stable organizations, which play a fundamental role in dynamical systems, has still to be formally defined. Yet we believe that our work represents a step forward towards a formal study of constructive dynamical system.

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