



**UvA-DARE (Digital Academic Repository)**

**Dynamical and structural self-organization : a study of friction, liquid-crystal nucleus growth, and supramolecular polymers through simple models**

Huisman, B.A.H.

[Link to publication](#)

*Citation for published version (APA):*

Huisman, B. A. H. (2008). Dynamical and structural self-organization : a study of friction, liquid-crystal nucleus growth, and supramolecular polymers through simple models

**General rights**

It is not permitted to download or to forward/distribute the text or part of it without the consent of the author(s) and/or copyright holder(s), other than for strictly personal, individual use, unless the work is under an open content license (like Creative Commons).

**Disclaimer/Complaints regulations**

If you believe that digital publication of certain material infringes any of your rights or (privacy) interests, please let the Library know, stating your reasons. In case of a legitimate complaint, the Library will make the material inaccessible and/or remove it from the website. Please Ask the Library: <http://uba.uva.nl/en/contact>, or a letter to: Library of the University of Amsterdam, Secretariat, Singel 425, 1012 WP Amsterdam, The Netherlands. You will be contacted as soon as possible.

# Summary

## Self-organization

Randomly throwing a lot of bricks and mortar in your back yard might result in something that separates your property from that of your neighbor, it does not however, result in a nicely ordered brick wall. For that you will need some tools, some masonry experience, and a lot of effort. No wonder that many see an intelligent designer in the order of the things around us. But while walls do not build themselves, many naturally occurring structures do. Ice, for instance, is not meticulously crafted by someone putting all the water molecules in the right place, liquid water “just” freezes below a certain temperature. And while we put a lot of effort into making objects behave in an organized fashion, many naturally occurring processes seem to take no effort at all to become, and stay, organized—be it in space or with some regularity in time.

All these processes in which the organization of a system with many, often very similar, elements increases in complexity or regularity without any external guidance, are called self-organizing. In other words, the self-organization of the constituent elements is only driven by the interactions between the elements. That is not to say that these interactions instantly draw the elements to their most stable configuration, as if pulled by many invisible strings. The elements usually explore many configurations before reaching the one that is most stable. So even if the bricks in our pile could attract each other very specifically, they would need to move around a lot more in their pile for them to find their wall configuration.

These self-organizing systems are the subject of many fields of science. The structure of almost all materials is achieved by self-organization, patterns found in dynamical systems with many coupled elements often arise through it, and even quite complex biological structures emerge from this process. The reasons behind why the individual elements behave the way they do may be very different, and may constitute a field of science all of its own. However, the art of studying self-organization is to model these elements and their interactions in the simplest way possible, while still retaining the complexity of the structures and patterns they form. Here I have tried to do just that.

In this thesis I used and studied three simple models that describe a self-organizing system, or that exhibit self-organization. Each of these three subjects required a different numerical approach. I studied a simple block and spring model representing multicontact friction, which I did by integrating large sets of ordinary differential equations. To study slow relaxation processes in this model, I studied a simplified version of these equations by means of a probabilistic automaton. As a second example

of a self-organizing system I determined the influence of latent heat on the dynamics of an isotropic-to-nematic phase transition in liquid crystals, that involved integrating a coupled set of partial differential equations. Finally, to study the linear supramolecular self-organization of discotic molecules into polymers, and their subsequent bundling into fibers, I performed Monte Carlo simulations of patchy particles.

## Multicontact friction

In chapter 2 I introduced the Burridge-Knopoff (BK) model of multicontact friction. This is a model for two slowly moving, macroscopic objects that are pressed together by an external force. Although these objects are in relative motion, the surface separating the objects consists of many small contacts that move only occasionally and remain motionless the rest of the time. The BK model describes these rather complex sliding systems as a collection of blocks connected to one of the objects by springs of strength  $k_1$  and to the other object by a simple, velocity dependent friction force. The blocks interact with neighboring blocks via springs of constant  $k_2$ . Depending on the ratio  $k_2/k_1$  of the stiffness of the springs, and depending on the type of friction law that is used, this model can be applied to sliding systems of hugely different length scales. On the high end of the scale it has been used to describe the motion of the interface between two tectonic plates in an earthquake fault, where strong interaction between blocks causes avalanche type sliding events. On the other end of the scale it has been proposed to describe the interface between two atomically flat sliding surfaces, lubricated by a locally solidifying liquid only a few molecular layers thick. In this case  $k_2$  is about the same size as  $k_1$ .

I studied a version of the BK model suitable to describe the latter situation of boundary lubricated friction, i.e. where  $k_2 \sim k_1$ , in Chapter 3. For this model an ordered mode of motion arises, that we have called the solitary state, in which every block only moves when its direct neighbors do not. This type of motion disappears if the blocks interact too strongly. I studied the properties and stability of this solitary state, both computationally and theoretically and showed that the solitary state is stable against boundary effects and small temperature fluctuations.

In Chapter 4 I then used the properties of this solitary state of motion to obtain a better description of the relaxation of the average stress on the contacts at finite temperature, after the sliding objects suddenly stop moving. To study this very slow relaxation process I have devised a very efficient probabilistic automaton model, and I simplified this automaton model further to obtain an analytical expression for the stress relaxation in the BK model. I find that the stress relaxation in the BK model is very similar to the relaxation of hierarchically constrained glassy systems, showing that neither hierarchical constraints nor highly complex dynamics are necessary for slow, glassy relaxation.

---

## The influence of latent heat on nematic liquid-crystal nucleus growth

I introduced the concept of a first order phase transition by means of nucleus growth, applied to the growth of orientational order in a liquid crystal in Chapter 5. In this phase transition an undercooled isotropic liquid, consisting of anisotropic liquid crystal molecules suddenly aligns locally into an orientationally ordered, nematic, domain. If this nucleated domain is large enough it grows, so that eventually all of the molecules are aligned. Latent heat caused by the change of order leads to a small temperature change at the growing domain interface, which might influence the growth dynamics. Experimentally a transition from diffusive domain growth to linear domain growth has been reported, as a function of the undercooling temperature.

In chapter 6 I tried to answer the question whether the diffusion of latent heat causes this transition from diffusive to linear domain growth. For the time and length scales I am interested in, I cannot keep track of the organization of the individual elements in this transition. Instead, I studied the evolution of an order parameter that describes the *amount* of ordering at each position during the transition. For this order parameter I solved a partial differential equation in time and space, called the time dependent Landau-Ginzburg equation, that models the evolution of the order parameter as a function of temperature. By coupling this equation to a diffusive heat equation I calculated the effect latent heat has on domain growth dynamics. Because the time and length scales involved in orientational ordering and in thermal diffusion are so different in liquid crystals, I devised a semi-analytical approach to integrate the coupled equations efficiently. I showed that the diffusion of latent heat away from a growing domain boundary can cause a transition from diffusive to linear growth in very large systems. However, comparison with experiments shows that the diffusion of latent heat is too fast to explain the diffusive growth measured in the lab.

## The self-assembly of molecules into polymers and of polymers into bundles

In the last part of this thesis I addressed supramolecular self-assembly, a type of self-organization found in nature to result in highly complex biological structures such as virus capsids, membranes and fibers. Studying this type of self-assembly may not only lead to a better understanding of how these structures are formed, but may also lead to the design of new materials. In Chapter 7 I introduced the patchy particle, a promising approach to model the subunits that self-assemble into these supramolecular structures. Dealing with each individual atom in each individual molecule, and calculating all the interactions between all the atoms quickly becomes too costly for the large assemblies that are formed. Therefore the patchy particle model separates the shape of a molecule from the interactions between the molecules.

The shape of the molecule is modeled by one, or only a few, simple beads and the interactions between the molecules are modeled by only a few patches of interaction at the surface of these beads.

In Chapter 8 I described a discotic molecule by a single bead and two directionally dependent patches, and I studied the linear self-assembly of these patchy particles numerically. I showed that a small sideways interaction between the beads in different polymers causes these polymers to undergo a transition to a solid bundle, which may be a first step to understanding why many naturally forming polymers self-assemble further into bundles. I compare our numerical results to theoretical predictions based on the Wertheim thermodynamic perturbation theory, and propose a simple thermodynamic model of polymer sublimation to describe the bundling transition. Although this patchy particle model does not lead to bundles with a limited bundle thickness as often found in nature, it may be a first step towards understanding bundling in these systems as well.