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C. R. Physique 6 (2005) 3–9
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Self-organization on surfaces/Auto-organisation sur les surfaces

Self-organization on surfaces: foreword

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Available online 12 January 2005

Abstract

After decades of work, the growth of continuous thin films, i.e., two-dimensional structures, is progressively becoming a technological issue more than a field of fundamental research. Incidentally self-organization of nanostructures on surfaces is now an important field of research, i.e., structures of dimensionality one or zero, with a steep rise of attention in the past five years. Whereas self-organization was initially motivated by potential applications, it has up to now essentially contributed to the advancement of fundamental science in low dimensions, as model systems could be produced that could not have been fabricated by lithography. This Special Issue aims at giving a cross-community timely overview of the field. The Issue gathers a broad panel of articles covering various self-organization mechanisms, specific structural characterization, physical properties, and current trends in extending the versatility of growth. The materials mostly covered here are semiconductors and magnetic materials. ***To cite this article: O. Fruchart, C. R. Physique 6 (2005).***

Résumé

Auto-organisation sur les surfaces : préface. Après plusieurs décennies d'intense activité, la croissance des couches minces, c'est-à-dire de systèmes de dimension deux, tend progressivement à devenir plus une question de maîtrise technologique qu'un sujet de recherche fondamentale. Dans le même temps l'auto-organisation aux surfaces de nanostructures, c'est-à-dire de dimensionnalité un voire zéro, prend une importance grandissante, tout spécialement depuis cinq ans. Alors que l'auto-organisation était initialement motivée par des perspectives d'applications, elle a pour l'instant essentiellement contribué à l'acquisition de nouvelles connaissances de physique fondamentale en basse dimension, puisque des systèmes modèles ont pu être fabriqués, qui n'auraient pu être obtenus par la technique conventionnelle de structuration qu'est la lithographie. Ce dossier thématique a pour objectif de proposer un aperçu actuel de l'auto-organisation, en essayant de dépasser les frontières de communautés. Ainsi sont rassemblées des contributions traitant de divers mécanismes physiques de l'auto-organisation, de techniques de caractérisation spécifiques, de propriétés physiques, et des nouvelles approches poursuivies pour augmenter la versatilité des procédés de croissance. Les matériaux couverts ici sont essentiellement les semiconducteurs et les matériaux magnétiques. ***Pour citer cet article : O. Fruchart, C. R. Physique 6 (2005).***

Keywords: Self-organization; Self-assembly; Bottom-up; Low-dimensionality; Dots; Wires; Nanostructures

Mots-clés : Auto-organisation ; Auto-assemblage ; Bottom-up ; Basse dimensionnalité ; Plots ; Fils ; Nanostructures

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1631-0705/$ – see front matter
doi:10.1016/j.crhy.2004.11.009

1. Introduction

A draft definition of self-organization (SO) could be: *a situation were structures with a certain degree of order appear spontaneously*, or for the restricted case of growth at a surface: *a growth process during which the spontaneous formation of nanostructures occurs*. For decades growth parameters and the combination of materials have been optimized to yield continuous thin films of the highest structural quality and smoothness upon growth at surfaces; lithography was then used when circuits and other patterns of finite lateral dimensions were required. On the other hand, materials and growth parameters can be optimized with an opposite goal, that of producing spontaneously nanostructures on a surface. This is self-organization, which appears as an alternative route to lithography. Self-organization can be viewed as the generalization of growth processes to dimensions lower than two. Indeed thin films growth makes use of the aggregation of material at a surface (2D), while self-organization makes use of aggregation along lines (1D) and points (0D) to yield stripes and dots. It is the purpose of this Special Issue to examine the current status of self-organization at surfaces and its uses.

The first motivation put forward to study self-organization is often the potential for hi-tech applications. Indeed, the exponential increase of integration and performance of devices is accompanied by a similar increase of the cost of investment for building the fabrication facilities. Self-organization, that allows the spontaneous fabrication of nanostructures upon a single and cheap growth procedure has attracted attention for its potential to become an alternative route to lithography to push further the limits of integration at a reasonable cost. The increase of density of lithographically-defined patterns is called a *top-down* approach, were scientists and engineers work on improving the concepts in use for today-lithography. Alternative approaches are called *bottom-up*, where one tries to tailor the growth of individual entities, like atoms, clusters, molecules, ..., up to the spontaneous fabrication of patterns. However, although some serious prospects still exists, there has been no breakthrough for applications after nearly two decades of research. Intense work is nonetheless still going on in the field, pushed by the potential for fabricating model systems for fundamental studies, that could not be achieved by any other existing technique. We will see that the effort in the field has lead to the advancement of physics in low dimensions. Notice that technology and fundamental research in the nano-world are nonetheless closely related, as new effects are expected at the scale of nanometers. As such effects cannot be predicted from the properties of larger objects, it would not be desirable to have investigations of self-organization guided solely by the need to fabricate in the low-nanometer scale patterns whose functionality are desired and known at a larger scale. Fundamental research is a mandatory step to explore the field in all directions with no a priori prospect, and is a sufficient justification for the current effort in the field of self-organization.

Why is today an overview of SO timely? SO first came in the mid-eighties from the semiconductor community, with the technological prospect of high-efficiency lasing in quantum dots [1,2]. The rapid expansion of the field was facilitated by the incidental introduction at the same time of scanning probe microscopies like scanning tunneling microscopy and atomic force microscopy. Now SO is a broad and established field of epitaxial growth in many communities, both in terms of materials and physics. In the past five years many new ramifications emerged beyond the study of growth and of specific properties like luminescence for semiconductors, magnetic anisotropy for magnetic materials, etc. For example, X-ray characterization techniques specific to SO were developed. At the same time the growth engineering was still being pushed further. Investigated directions include a bottom-up approach to fabricate truly 3D materials now that the structure at the nano-level is controlled at surfaces, or combine a first versatile step of pre-patterning, e.g., lithography, with overgrowth. It is therefore timely to propose an overview of what has been achieved, and of current prospects of technological and fundamental interest. The fact that SO is now spread in many communities has motivated a broad overview of the field in this Special Issue, rather than a set of very specialized reviews on a narrow subject. Although some specialized reviews about SO were available [3–8], mainly devoted to growth, a broad set of reviews was not available to date. Indeed, as self-organization requires the engineering of growth processes, and also accurate characterization techniques, there is often a lot to gain at looking at other communities than our own. It is also essential to understand what kind of new physical functionalities might arise from SO systems to tailor the growth in the good direction. With these ideas, I hope that this Issue will help the reader to gain an overview of the field of SO.

Let us first discuss a semantic issue. We will see that two prototypical situations are encountered concerning the spontaneous formation of nanostructures upon growth: the nanostructures may either display only weak nearest-neighbor correlations, or display a mid- or long-range positional order. It seem therefore desirable to use different names to distinguish these two situations. Incidentally two names are found in the literature to describe the spontaneous formation of nanostructures on surfaces, i.e., *self-organization* (SO) and *self-assembly* (SA).[1] However there is no agreement in the community as to the exact use of these two terms, that are sometimes used interchangeably, or whose meaning may depend on the community. To reconcile this lack of consensus with the need for two distinct names stressed above, I propose to call *self-organization* the prototypical case of nanostructures displaying a mid- or long-range positional order, and *self-assembly* the prototypical case of nanostructures displaying

[1] We will abusively use the acronym SO (or SA when appropriate) to stand for different expressions like self-organization, self-organize, self-organized.

only weak nearest-neighbor positional correlations. Of course, in real situations there may be no strict borderline between these two situations. If one forgets about this issue of positional order, the use of SO and SA nanostructures can be classified into two categories. In the first category they are used to gain information about fundamental phenomena that occur in materials and systems that may be of interest for applications, but cannot be understood directly, because they are too complex (owing to microstructure, defects, large size . . .). Beyond the advancement of our understanding of physics in low-dimension, these studies are of applied interest, because devices require the use of ever smaller nanostructures, whose properties must be understood and ultimately tailored. In this case SO systems are used preferably to lithography or microstructured materials as objects of very high quality to serve as model systems for analyzing fundamental issues of physics. Indeed, these are characterized by a very high resolution, ultimately down to the atomic size [9,10], much better than existing and presumably any forthcoming lithography technology. Besides, the quality of the interfaces can be potentially controlled at this scale, with presumably much less defects than for lithography (roughness, amorphisation, oxydation or other gas adsorbtion, . . .). Thus, they possess the required features to study fundamental physical phenomena in low dimensions, as the use of a model system maximizes the chances to elucidate the relevant phenomena quantitatively. The second category of studies consists in investigating whether SO systems might be used directly for applications. This still remains an open question after twenty years of research: the physical properties of these systems are often appealing, but technological barriers remain to be overcome. These two categories of studies will be illustrated in this Special Issue. Finally, notice that SO receives often more attention than SA. This partly stems from the fascination for an order that can arise spontaneously. However from scientific and technological points of view the advantage of SO most often is not the order itself, but the fact that in ordered patterns the size distribution is very narrow, because the area of capture of all objects during growth is similar for all the nanostructures. For fundamental science this gives researchers the opportunity to assess the properties of nano-sized systems by measuring large assemblies of these, and assuming that all entities contribute in a similar way. From a technological point of view, reducing the size dispersion is important to reduce the dispersion of physical properties of use in a device, like wavelength in a quantum-dot laser or coercive field in a magnetic recording media. The sketch of the Issue is given in the following.

The first article, by Olivier Pierre-Louis, is a review of the literature work about *steps on surfaces* [11]. As crystals can never be polished exactly along surfaces of low Miller indices, steps are always found on surfaces to compensate for the miscut angle. Thus the study of steps on surfaces is a long-standing issue in surface physics [12], and steps have been observed to self-assemble or self-organize. Experiments were first made possible by electron microscopies like grazing incidence electron microscopy, or by LEEM, and then revived by the introduction of scanning probe microscopies. Experimentally, parameters like the miscut angle and growth conditions provide theoreticians with sets of data to help understanding both thermodynamic and kinetic processes. Despite the large number of models and simulations released especially in the last fifteen years, the subject is still timely. The current trend is the increasing overlap of phenomenological models with microscopic aspects like strain and electronic structure in a multi-scale approach. The knowledge of the base models in this well-established field is worth, as the microscopic ingredients governing the interaction between steps are also often those who are responsible for the SO and SA of nanostructures deposited on surfaces.

Another many-decades-old issue on crystalline surfaces is the classification of growth modes [13–15], that was first rationalized by Bauer in 1958 [16] in the growth modes now called Volmer–Weber, Stranski–Krastanov, and Frank-van-den-Merwe. For reviews, see for instance [17–19]. However, Nature does not like drawers and boxes, and models need always to be refined to describe different systems, physical effects, and dimensions of the patterns. Thus there is still a significant activity going on to explain and then tailor the growth of nanostructures, especially concerning the conditions of occurrence and the shape of dots [20] in the Stranski–Krastanov growth mode. In this context Henri Mariette proposes an interesting contribution: *Key parameters for the formation of self-assembled quantum dot induced by the Stranski–Krastanov transition*: *a comparison for various semiconductor systems* [21]. A surprisingly simple model allows one to understand quantitatively the occurrence of semiconductor quantum dots, by taking into account both the strain decrease in the dots and conventional surface energy arguments occurring in the growth modes recalled above. Such understanding is essential to increase the versatility of SA. An original aspect of the model is to explain the effect of parameters other than just material, substrate, temperature and amount of material deposited, namely the use of surfactants to manipulate the surface free energy and thus control the formation and density of quantum dots.

The advantage of SO over SA was mentioned in the introduction: the dispersion of size is greatly reduced, thus either easing fundamental studies over assemblies of nearly mono-disperse objects, or improving materials for applications by reducing the dispersion of physical properties (the order may also allow one to apply the powerful analytical methods of diffraction, see the contribution by Metzger et al.). SO generally does not arise during deposition on crystalline surfaces, because once nucleated nanostructures are often immobile, as substrate-deposit interactions are much smaller than interactions between neighboring nanostructures (this contrasts with ordered arrays of clusters fabricated by chemical means, see below the contribution of B. Chaudret). Thus in most cases the organization of deposited nanostructures relies on the nucleation in an ordered fashion, in registry with a pre-existing regular pattern on the growth surface. The way this pre-existing pattern gets organized should in principle be the step that should first be called self-organized. Sylvie Rousset et al. propose an overview of the issues related to

self-organization of surfaces including steps, intrinsic and adsorbate-driven reconstructions, as well as the use of such surfaces for self-organized overgrowth, mainly illustrated by results of their group [22].

New fabrication processes often trigger the need for new characterization techniques. The most widely reported studies of SO systems are performed in real space, using electron or scanning probe microscopies. A complementary approach is X-ray scattering. Indeed, thanks to coherence lengths of typically a few micrometers, information on electron density fluctuations on a larger length scale than atomic spacings can be obtained with a high precision using powerful analytical tools. In principle, the entire mesoscopic range can be covered by X-ray scattering, from the order of nanostructures down to their atomic structure. Besides, X-ray techniques are non-destructive and can be performed in situ. Mainly small-angle scattering [23] and satellite diffraction studies are reported in the literature, as reported in recent reviews [24–26]. An example of such studies is reported by Till Metzger et al., entitled *X-ray characterization of self-organized semiconductor nanostructures* [27]. These authors present an original combination of grazing incidence techniques, i.e., grazing incidence diffraction, with anomalous dispersion effects, i.e., at energies close to an adsorbtion edge of one of the constituent elements, to add chemical sensitivity to the technique. Applied to the case of SiGe quantum dots, this allows the chemical composition and strain to be determined, a so-called *iso-strain-scattering* technique. Such studies are essential as physical properties like luminescence critically depends on composition and strain.

Semi-conductor nanostructures play a central role in SA and SO. As mentioned above these triggered the first growth investigations with the prospect of high-efficiency lasing in nanometer-sized dots [1,2]. Since that time the prospects have been refined. Quantum wires or quantum dots will certainly not replace quantum wells because their uniformity in size is not sufficient. Foreseen applications rather consist of niche applications, like single-photon sources, or quantum-dot lasers with a wavelength in a range not achievable with quantum wells. Several recent reviews are available on the subject [28–30] and no contribution was included in the present Issue.

Magnetism is another field that has benefited from SO and SA systems. The studies started later than for semiconductors, with the first growth demonstrations published at the beginning of the nineties. As the field is becoming riper, systems are now not solely grown for demonstration, but their properties are investigated for specific purposes since a few years, with a clear rising interest and number of reports. This interest is stimulated by the fact that the further increase of density of magnetic recording media seems to come to an end using conventional film media. Similarly, magnetic elements are now integrated in magneto-electronic devices like magnetic random access memories (MRAMs), for which the trend of miniaturization raises similar issues as for recording media. Olivier Fruchart proposes a review article entitled *Epitaxial self-organization: from surfaces to magnetic materials* [31]. This consists of an overview of the use for magnetic purposes that has been done up to now of SO and SA, with an emphasis on aspects that could not have been addressed with nanostructures fabricated by lithography. A first set of issues concerns the quantitative study of low-dimensional magnetic phenomena (1D and 0D) that also occur in functional materials but cannot be studied because of their complexity: magnetic ordering, magnetic anisotropy, superparamagnetism. A second set of issues concerns the possible direct use of self-organized systems in devices. Examples are given how superparamagnetism can be fought, and what new or improved functionalities can be expected.

A magnetic achievement made possible by SO and SA is presented in more detail in the manuscript by Pietro Gambardella et al., who report on *magnetic anisotropy from single atoms to large monodomain islands of Co/Pt(111)* [32]. Magnetic anisotropy is an essential property because it allows the freezing of magnetic systems in a given direction, and therefore is a keystone of magnetic recording. Understand how this anisotropy is modified and can be manipulated in nano-sized systems is a challenge, that can be tackled using SO and SA to produce model systems for fundamental investigations. These authors studied model systems consisting of large flat islands, stripes narrow down to mono-atomic wires, and controlled clusters down to isolated atoms. Thus, the evolution of magnetic anisotropy from bulk crystals to isolated atoms was fully covered, i.e., from 3D to 0D. The physical conclusion is that magnetic anisotropy mainly arises from the atoms that posses the lowest dimensional coordination in the system: surfaces for ultrathin films, which was already predicted [33], checked experimentally [34], understood microscopically [35] with again experimental confirmation [36]. Here the conclusion is extended to edges for OD and 1D objects. This new piece of knowledge opens the perspective to control the magnetic anisotropy independently from the size of the system by engineering the area of interfaces in a compact cluster, or of the length of edges in a flat structure, in an analogous approach to the control of the dimension in fractal structures. This is illustrated by an original example of engineered growth, were flat rings of Co were fabricated by step-decoration of non-magnetic Pt flat islands.

A prerequisite for the use of spontaneously-ordered systems in devices is to increase the versatility of patterns that can be achieved, and also the quality of the order. Growth studies in this direction are presented in the next two manuscripts. First Günther Springholz proposes a literature review of the *three-dimensional stacking of self-assembled quantum dots in multilayer structures* [37]. It was indeed shown that dots from successive layers can display correlations, so that a three-dimensional ordered superstructure can be fabricated upon deposition of many layers of dots and spacer layer [38–40], although the dots from the first layer are only self-assembled. The manuscript reviews the various physical effects that determine the occurrence of this order, the range of geometrical and material parameters required for the ordering, the type of stackings that can result, and ways to control the super-lattice parameters. As stressed in the introduction, ordering is interesting because it reduces the

dispersion of size, thus of physical properties. SO three-dimensional stackings could be used in a very general way: if a stacking is inert with respect to the physical properties that are sought, it could be used as a template for the overgrowth of the material of physical interest, i.e., the stacking could be a building block of a more complex stacking [41]. New physical properties may also emerge because of interactions between dots through the spacer layers.

Although stackings increase the versatility of SO, still only a restricted number of patterns can be obtained, that are all regular. In order to lift this limitation but still benefit from the advantages of spontaneous fabrication under ultra-high vacuum, a new direction of research consists in combining a first step of artificial structuring to impose a pattern, not necessarily regular, with a second step consisting of growth self-organized in registry with the pre-defined patterns. The main approaches followed are reviewed by their developers in the manuscript of Joël Eymery et al., entitled *nanometric artificial structuration of semiconductor surfaces for crystalline growth* [42]. These consist of the use of arrays of buried dislocations produced by controlled wafer bonding (fast and with a low pitch, however not more versatile than regular arrays), lithography patterning, or ion implantation patterning. These studies initiated in the field of semiconductors about five years ago, and were eased by the existing technological procedures to prepare again clean epitaxial surfaces after the pre-patterning step. More recently a few reports appeared for the control of nucleation of metals [43,44], or clusters [45,46].

The Special Issue is concluded by a manuscript that does not relate epitaxial systems, but nanoparticles synthesized by chemical means. Physical chemistry is indeed a promising route to yield both model systems for fundamental studies, and materials for applications. Many approaches have been developed to yield nanoparticles by chemical means [47], and let them self-organize in 2D or 3D arrays. A first advantage of chemistry over physical deposition is the lower magnitude of interactions between molecules than between epitaxial nanoparticles, which favors the ordering. A second advantage is the nearly infinite possible combination of ligands, that give many more degrees of freedom than the choice of elements for epitaxial growth. A popular chemical route for the fabrication of magnetic nanoparticles is inverse micella [48–50]. In this Issue Bruno Chaudret presents an original route recently opened, i.e., an *organometallic approach to nanoparticles synthesis and self-organization* [51]. As detailed in the article, this approach presents several advantages. First, it benefits from the existing know-how in metal-organic chemistry, yielding metals, semi-conductors and oxides, each with a great variety of materials, in pure clusters or core-shell structures. Second and probably the most original feature of this report, the shape of the clusters can be controlled, e.g., to yield cubes. In SO 2D and 3D supra-crystals of these clusters, this anisotropic shape can induce the alignement of the crystallographic axes of all the clusters along a given direction, owing to nearest-neighbor interactions via ligands. It is of prime importance when the physical properties of interest of the particles are anisotropic, because in this case alignement of all major axes in given directions can be required for integration into devices. This is for example the case for the high-magnetic-anisotropy FePt nanoparticles that may be used in future generations of magnetic recording media. The alignement of all axes reduces the distribution of magnetic switching field mandatory to achieve sharp bit transitions thus reducing the number of grains per bits to its minimum, as well as provide a nearly 100% remanence, desirable for maximizing the read-out signal and the SNR. The anisotropic shape easily obtained is explained by the anisotropic role of the ligands during growth, while only the anisotropic surface energy of the crystalline clusters plays a role in inverse micella. The organo-metallic approach is also complementary to inverse micella because it allows the synthesis of smaller clusters, typically a few nanometers in diameter against several tens of nanometers, respectively.

To conclude, self-assembly and self-organization on surfaces has become an established field in the community of growth, with a steep rise of attention in the last five years. Whereas self-organization was initially motivated by potential applications, up to now it has essentially contributed to the advancement of fundamental science in low dimensions, for example in the fields of semiconductors and magnetism, as model systems could be produced, that could not have been fabricated by lithography. However, there are still breakthroughs under way in extending the versatility of self-organization, with a great hope in the emerging field of the combination of first an artificial pre-patterning, followed by self-organized growth in registry with the artificial pattern. This lifts the limitations of self-organization regarding the regularity of the pattern, while keeping model nanostructures. Thus, self-organized systems are still under active development and may anyhow find technological applications in the future.

Acknowledgements

I would like to express my gratitude to Jacques Villain, who has given me the opportunity to coordinate this Special Issue.

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