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# Self-organization on surfaces : an overview

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Brasov (Romania), Sept. 2003

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## 1 Introduction

A short overview of the field of condensed matter self-organization on surfaces is proposed. The different mechanisms responsible for self-organization will be covered, with examples taken from semiconductors, oxides and metals.

In this document I will call self-assembly (SA) the process by which nanostructures are fabricated spontaneously by deposition on a surface. These nanostructures might be dots, stripes, wires, tubes etc., and in general they display no long range positional order. I will call self-organization (SO) a special case of SA, that where the nanostructures display a long-range position order. We will see that in most cases the order is caused by self-organization of the substrate surface itself before deposition, not to phenomena related to growth. Note that this definition of SA and SO is not universally admitted in the literature.

## 2 Self-assembled epitaxial growth

### 2.1 Growth modes in epitaxy

The first parameter acknowledged to influence growth modes is surface energy [6], a parameter also occurring at the macroscopic scale, that favors wetting and therefore the growth of continuous films for materials of low surface energy deposited on substrates of high surface energy [7] (Franck van den Merwe growth mode, FvdM). In the reverse case the formation of *dots* (also called *islands* or *clusters*) on an otherwise bare surface is favored (Volmer-Weber growth mode, VW). Another parameter, found only at the microscopic scale, is the lattice mismatch between substrate and deposit. Lattice mismatch induces the formation of dislocations at the interface, whose energy cost favors again the formation of dots. This leads to a third growth mode, consisting of wetting of a few atomic layers related to surface energy arguments, followed by dots formation due to lattice mismatch and dislocations formation (Stranski-Krastanov growth mode, SK) [8]. This thermodynamic picture may be hindered by kinetic aspects. SO and SA generally make use of SK and VW growth modes.

### 2.2 Shape of dots

At the macroscopic scale and down to some hundreds of nanometers, the lattice symmetry and parameters of supported dots is relaxed to their bulk values. In this case their shape is only determined by the minimization of surface plus *interface* energies. It can be predicted straightforwardly using a geometrical construction named Wulff-Kaichev's theorem, based on an extension of the century-old Wulff's theorem for free dots (Ref.9 and included references). For smaller dots' size strain come into play, and one should use either a modified Wulff-Kaichev's continuum approach [9] or an atomistic calculation.

### 2.3 Parameters to play with

The parameters accessible to the experimentalist to tailor the shape of SA nanostructures are :

- *Temperature*, that influences the areal density of nanostructures on a defect-free surface. This parameters may not be effective in the case of SO deposition on a pre-patterned surface.
- *Amount of deposited material*, that influences the size of nanostructures after the nucleation stage.
- *Symmetry* of both substrate and deposited material, influencing the dot's shape and facetting.

## 2.4 Potential applications of self-assembled dots

SO and SA cannot compete with lithography because the order and versatility are not sufficient for most industrial requirements. However they are competitive for producing large-area assemblies of dots in the low nanometer range, at or below the limits of lithography. Beyond the argument of low-cost, the advantage is to produce high-quality dots when one needs to develop physical properties that depend critically on the structure down to the atomic scale (in terms of microstructure and surface/interface quality). This is the case for semiconductors where defects can localize charge carriers, magnetism where interface defects induce irreproducible events, and catalysis that deals with surfaces.

The present prospects of potential applications concern SA deposits, where each dot is used independently, and the positional order plays no role. The deposit is taken as a whole, and considered as a material. This includes catalysis for metals deposited on oxides, and quantum dots for semiconductors. In the latter case there are serious prospects for high-efficiency laser, and more futuristic ideas of single-electron devices.

## 3 Self-organized epitaxial growth

Generally SO does not arise from growth-related phenomena, but occurs thanks to a self-organized pattern pre-existing on the substrate before deposition. SO will be competitive and of interest when the formation of structures intermediate between atomic size and the lithography resolution limit is achievable, *i.e.* around 10 nm. The interest in SO structures is not necessarily the order itself, but the improved monodispersion of structures' size as compared to SA. Different types of SO strategies to fulfill this criterium are listed below.

### 3.1 Arrays of atomic steps

A vicinal surface of a single-crystal is a free surface whose orientation is close to that of a low-Miller-index surface. Then, one or two Miller indices are small, the other(s) are high, for example (1 1 13), (1 17 0), (1 17 13), etc. Vicinal surfaces are usually obtained by polishing a single crystal slightly off a low-Miller indices direction, by an angle called the *miscut angle*. When suitably prepared, at the atomic scale a vicinal surface consists either of a regular array of parallel *atomic steps* separating atomically flat *atomic terraces*, or an array of parallel accumulations of atomic steps separated by flat atomic terraces, a phenomenon called *step bunching* [3]. Step bunching can be viewed as a periodic micro-facetting of a surface. A 'suitable preparation' usually consists of annealing, or a series ion etching/annealing, or (homo)epitaxial overgrowth, or a combination of several of these procedures.

Vicinal surfaces can be used as a template for further deposition ; this SO process is called *step decoration*. The idea is that the nucleation of adatoms along atomic steps is favored due to a higher coordination number. The growth temperature must be chosen high enough so that the mobility of adatoms is sufficient to reach atomic steps before nucleating clusters on terraces<sup>1</sup>. It is also advisable to choose a temperature not too high, so that alloying or intermixing can be avoided, and lower than the *two-dimensional evaporation temperature*<sup>2</sup>, otherwise growth instabilities may develop [10]. Step decoration can result in the fabrication of dots clustered along steps [11], stripes (thin and large) [12, 13] or wires (thin and narrow down to the size of one atom) [14].

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<sup>1</sup>In other words, the distance between steps must be smaller than the mean distance  $\ell(T)$  between islands that would be nucleated on an infinite flat terrace.

<sup>2</sup>Two-dimensional evaporation means that the adatoms stay on the surface, but can leave steps to wander again on terraces

### 3.2 Surface reconstructions

At a free surface the coordination number of atoms is smaller than in the bulk. A surface is said to be *reconstructed* when the lateral positions of the surface atoms differ—spontaneously— from a bulk atomic plane of identical Miller indices<sup>3</sup>, resulting in an increase of the surface primitive cell. For semiconductors broken bonds often lead to the grouping of surface atoms, for example pairing to form rows, or more complex structures like for Si( $7 \times 7$ ). For metals the density of states is increased at the surface, which favors smaller in-plane inter-atomic distances. In some instances, this results in the topmost plane being denser than in the bulk, inducing a long-range wavy structure to accommodate this plane with bulk ones, like in Au(001) or Au(111). The reconstruction of Au(111) is complex but useful for SO, for it has a cell size well above atomic distances, around 10 nm [15, 16]. A surface reconstruction often implies breaking of surface-symmetry.

Surface reconstructions can be used as a template for growth. The adatoms can be trapped in a reconstruction cell, like for Si(111) [17], or nucleate via an atomic site-exchange process like for Ni, Co and Fe on Au(111) [18]. Note that the transfert of SO from the template to the overgrowth is not granted, and can be achieved only for selected systems and suitable growth conditions.

### 3.3 Reconstructions due to adsorbates

Surface reconstructions can also be induced by the adsorption of sub-atomic-layer amounts of gases. Such reconstructions are more versatile than *intrinsic* reconstructions—*i.e.* without adsorbates— because they may exist whereas intrinsic reconstructions do not occur, or may be tailored changing the type of adsorbate or the adsorbed ratio. Besides, by adsorbing an amount of gas atoms smaller than that required to form a continuous reconstruction, patches of reconstructed areas are formed. Then, owing to short-range attractive forces holding gas atoms together, and long-range repulsive forces elastically mediated through the substrate [19], these patches may form a SO pattern with a period much larger than that of atoms, like for N/Cu or O/Cu with various orientations [20, 21], and therefore suitable for the growth of nanostructures.

The growth of metals, among which Ni and Fe, have thus been used to fabricate arrays of dots or stripes on Cu(001) and Cu(110).

### 3.4 Overlayer dislocations

It is known from the growth of continuous thin films that a periodic lattice of dislocations may be found at the materials interface for hetero-epitaxial growth. Such a lattice can be used as a template for SO. In a first step a hetero-epitaxial ultrathin film is deposited at temperature high enough to yield a flat surface, with a thickness just above the critical strain relaxation thickness to maximize the lateral modulation of chemical potential at the free surface. In a second step, the temperature is lowered so that newly-deposited adatoms remain trapped in the cells fenced by the dislocations, so that one dot is formed per cell. This has been achieved for Ag(sub-AL,110 K)/Ag(2 AL,400 K, annealing at 800 K)/Pt(111).

### 3.5 Wafer bonding and smart-cut

The type of array of dislocations described previously can be obtained without growth, by wafer bonding an ultra-thin plate on a thick substrate, making use of a smart-cut-like process. The geometry, the type of dislocations and the period of the interface dislocation network can be tailored by controlling the misorientation between the two single crystals. Then, if the top ultrathin layer is thinner than about the dislocation array period, a substantial adatoms chemical potential lateral modulation is induced at the

<sup>3</sup>A vertical displacement of the topmost atoms, *i.e.* a modification of the last inter-atomic distance, is always observed, usually a contraction. This effect is not called a reconstruction

surface resulting from the modulation of strain at the surface. This modulation can be used directly to achieve SO, or can be enhanced by an etching process prior to deposition. These processes were proposed and are still under active development in CEA-LETI, Grenoble [22]. The primary purpose is to provide a template for quantum dots with a tunable size.

### 3.6 Interplay between steps and reconstructions

Two or more of the above-mentioned routes for SO can be combined, for instance vicinal surfaces and intrinsic reconstruction. This may result in an increase of the reconstruction cell size, along with a possible change of symmetry. In the case of Au(788) the reconstructions can be forced to run perpendicular to atomic steps, lifting the three-fold symmetry of Au(111). This was used to produce a rectangular array of Co/Au(111) dots, instead of the conventional set of three domains of Co/Au(111). Vicinal surfaces of Si were used to stabilize triangular areas of  $(7 \times 7)$  and  $(1 \times 1)$  reconstructions with size up to hundreds of nanometers [23].

### 3.7 Self-organization from the deposit

SO obtained directly during deposition is more rare because the interaction of a nanostructure with a substrate is much higher than the interaction between neighboring nanostructures. However it is sometimes observed in the case of the growth of alloys or mixtures of elements. In this case, a *lateral composition modulation* (LCM) might reduce epitaxial strain more effectively than a uniform film, analogously to the so-called *Grinfeld instability* [24] that favors the development of roughness in thick multilayers. LCM is commonly observed in the growth of thick semiconductor alloys [25]. LCM has also been evidenced for ultrathin metal deposits consisting of two immiscible elements like CoAg or FeAg, leading to nanometers-wide chemical stripes [26].

## 4 3D Self-organization via multilayers stacking

### 4.1 Examples and theory of vertical stacking

With a view to producing macroscopic amounts of quantum dots, multilayers of semiconductor SA such dots have been produced. It has then been observed that dots from successive layers have a tendency to pile up vertically [27], when the inter-layer spacing is thin enough<sup>4</sup>. This effect is explained by the strain induced by the heteroepitaxial dots and mediated to the surface via the interlayer spacing [3]. This effect is therefore close to that used in Wafer-bonding (Sec.3.4,3.5).

### 4.2 From self-assembly to self-organization

A second effect is associated with the vertical stacking of dots. Whereas the dots of the first layer are SA, *i.e.* they display only a liquid-type short-distance order, after tens of multilayers the *lateral* order is observed to have greatly improved [28]. This is again explained by theory rather simply with Marchenko's arguments [19] like in the case of partial adsorbate areas (partie 3.3) : lateral inter-dot interactions are repulsive due to strain, so that the lateral interdots spacings tend to become equal. This effect do not occur for a single deposit, as we explained above due to the strong interaction of the dot with the substrate, but can be progressively reached by tilting the 'columns' of dots through the stacking. A good organization can occur when the total height of the stacking becomes much larger than the lateral inter-dots spacing.

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<sup>4</sup>Thin must be understood when compared to the lateral size and/or nearest-neighbor distance between dots

### 4.3 Different types of stackings

Finally, different types of stackings symmetry could be obtained, depending on the orientation of the growth surface. Indeed, for highly elastically anisotropic materials, the effect of strain can be mediated in tilted directions instead of vertically, like in the case of PbTe(111) [28]. In this case it was shown by HREM and diffraction that a super-fcc array of dots could be formed. The topmost layer again displays a long-range order, with a hexagonal symmetry.

## 5 Perspectives of self-organization

### 5.1 Towards self-organization engineering

In the previous sections we have listed the occurrences of SA and SO in categories and subcategories, in a pedagogical approach. However nature does not like boxes and drawers, and sometimes real situations cannot be disentangled into idealized cases. Besides, with a view to increasing the versatility of SO, it is a good idea to try to mix different approaches and change deposition conditions during the process. For example, we have mentioned the successful association of vicinal surfaces with reconstructions (partie 3.6), or the change of temperature during growth (partie 3.4). More can be done in the art of SO engineering, with the same spirit as in the case of continuous thin films where underlayers, separation layers and cappings can be suitably chosen. For example, 3D SO can be performed with one set of materials with the purpose to create the lateral order, then switch to another material, responsible for specific physical properties [29]. In the case of magnetism, such a process might be used to improve the monodispersion of grain size in recording media.

### 5.2 Self-organization on lithographic templates

Beside combining several epitaxial SO processes, one would also like to combine different techniques, like lithography and SO, the former to fabricate a pattern, the latter to growth defect-free nanostructures. This has already been achieved using annealed semiconductor MESAs, where the nucleation selectivity of more or less rough or stepped areas is used [30], or using oxide-based UHV compatible masks, *in situ* etching, followed by selective area epitaxy (SAE) [31]. Up to now examples came only from the semiconductor community, because of the well-established cleaning procedures that allow surfaces to be switched back to epitaxy-compatibility after lithographic procedures. Combination of lithography and SO remains a challenge for metals.

### 5.3 Self-organization beyond epitaxy

3D SO is not restricted to epitaxy. The stacking of Co clusters with layers of amorphous alumina were reported to give rise to a super-fcc stacking [32]. In this case, corrugation correlations are responsible for the progressive occurrence of order.

### 5.4 Structural characterization : scattering and super-diffraction

The development of new fields of physics often triggers the need for new characterization techniques. Apart from real imaging techniques, SO systems have been investigated by Xray scattering. When the order between nanostructures is sufficiently good, scattering resembles diffraction, with scattering vectors scaling inversely with the SO mesh size, not with atomic sizes. The, small-angle 'super-diffraction' [33] as well as satellite peaks around Bragg peaks [34] can be used. Information about Size and spacing can be extracted, along with strain and chemical composition [35].

## 6 Self-organization for magnetic investigations

To date SO and SA are less developed for metals than for semiconductors, and no potential applications can be foreseen. However SO and SA systems can be used in research as model systems, to elucidate fundamental issues that cannot be disentangled from extrinsic effects in bigger or lithographic-produced nanostructures. Some examples of such studies are given below.

### 6.1 Magnetic order in reduced dimension

SO has been used to mimic archetypical models of theoretical physics, like 2D or 1D spin networks. It has been confirmed that dipolar interactions, although weak, help stabilize long-range magnetic order in stripes [13]; mono-atomic chains of Co were shown to remain non-ferromagnetic, although the magnetic correlation length may exceed the system's finite size in experiments [36]. SA with mono-atomic height have been studied during structural percolation into a continuous film, revealing the universality of critical exponents associated with the magnetic percolation, each flat dot considered as a macrospin [37].

### 6.2 Anisotropy, from bulk towards atoms : steps, kinks and single atoms

Similarly to interfaces in continuous thin films, a symmetry breaking is expected at the edges and kinks of flat dots, the consequences of which are expected to dominate over bulk *and* top/bottom interfaces at small size. Owing to the deep sub-ÅL sensitivity of dichroism, it has become possible to investigate the evolution of spin and orbital moments of clusters [38, 39], stripes and wires down to monoatomic-width [36], and eventually single supported atoms [40], and thus scan the entire range of properties from bulk to atoms. The orbital moment, essentially quenched in the bulk, rises progressively above  $1 \mu_B$  in single atoms. Concerning spin moment, that of Co is essentially independent of size and shape, whereas that of Fe may vary considerably, which is explained by the sensitivity of Fe to structural changes, like in the bulk.

### 6.3 Model systems for micromagnetism

SO and SA are also useful for fabricating and studying model micromagnetic systems. For example quantitative analysis could be conducted on geometrically constrained domain walls [41], magnetic core vortices [42] and interaction of nucleation processes with facets [43].

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