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Self-organization on surfaces/Auto-organisation sur les surfaces

Epitaxial self-organization: from surfaces to magnetic materials

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Abstract

Self-organization of magnetic materials is an emerging and active field. An overview of the use of self-organization for magnetic purposes is given, with a view to illustrate aspects that cannot be covered by lithography. A first set of issues concerns the quantitative study of low-dimensional magnetic phenomena (1D and 0D). Such effects also occur in microstructured and lithographically-patterned materials but cannot be studied in these because of the complexity of such materials. This includes magnetic ordering, magnetic anisotropy and superparamagnetism. A second set of issues concerns the possibility to directly use self-organization in devices. Two sets of examples are given: first, how superparamagnetism can be fought by fabricating thick self-organized structures, and second, what new or improved functionalities can be expected from self-organized magnetic systems, like the tailoring of magnetic anisotropy or controlled dispersion of properties. ***To cite this article: O. Fruchart, C. R. Physique 6 (2005).***

Résumé

Auto-organisation épitaxiale : des surfaces aux matériaux magnétiques. Alors que l'auto-organisation est un domaine maintenant consacré pour les semi-conducteurs, il est en émergence pour les matériaux magnétiques, avec une activité soutenue les cinq dernières années. Un panorama des contributions de l'auto-organisation au magnétisme est proposé ici, avec pour but de montrer les possibilités nouvelles offertes, notamment par rapport à la lithographie. Une première catégorie d'études concerne la mesure et la compréhension de phénomènes magnétiques en basse dimensionnalité, qui existent dans les matériaux applicatifs mais ne peuvent y être étudiés quantitativement du fait de leur complexité : mise en ordre magnétique, anisotropie magnétique, superparamagnétisme. Une seconde catégorie concerne la perspective de l'utilisation directe de systèmes auto-organisés. Des exemples sont donnés pour combattre le superparamagnétisme en fabriquant des structures auto-organisées épaisses, ou établir des fonctionnalités nouvelles, notamment le contrôle de l'anisotropie et de la dispersion de propriétés. ***Pour citer cet article : O. Fruchart, C. R. Physique 6 (2005).***

Keywords: Self-organization; Self-assembly; Magnetism; Magnetic anisotropy; Micromagnetism; Superparamagnetism

Mots-clés : Auto-organisation ; Auto-assemblage ; Magnétisme ; Anisotropie magnétique ; Micromagnétisme ; Superparamagnétisme

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1. Introduction

It was the semiconductor community who first drew attention to deposition processes yielding spontaneously nanostructures at surfaces. This alternative approach to lithography, a so-called *bottom-up* approach, may be called either self-assembly (SA) or self-organization (SO).[1] This topic was initiated in the mid-eighties with the prospect of fabricating high-efficiency lasers with quantum dots [1,2] and is still a hot topic today [3], with however modified prospects [4–6]. The first demonstrations of SA and SO for magnetic materials date back to the early nineties. The topic has become very active only in the past few years, motivating the present overview of the contribution of SA and SO to the advancement of magnetism.

Mostly single-element metallic systems have been demonstrated for magnetic materials, with, however, recent reports on oxydes [7–9], metallic alloys [10] and metals on molecular templates [11]. Some publications aimed at showing that these growth phenomena occur for magnetic material, e.g., SO Ni, Fe and Co dots on Au(111) [12–14], SO mono-atomic Fe stripes on Au(788) [15], SO Fe and Co dots on reconstructed N–Cu(001) [16,17], Ni stripes on Cu(110)–$(2\sqrt{2} \times \sqrt{2})R45°$–O [18], Fe dots and wires on $H_2O/Si(100)(2 \times n)$ [19], SA Fe dots on flat NaCl [20]. Some other publications reported magnetic measurements in these structures, however, with no specific purpose, e.g., for SO Co dots on Au(111) [21], SO Fe dots and wires on facetted NaCl [22], SA TM-RE dots on Nb(110) [23]. Neither these growth nor magnetic studies will be reviewed here. As the field is becoming riper, specific uses are being sought for SA and SO magnetic systems, with the question whether they might be useful for fundamental science and/or for applications. In other words, the question is: can one reconcile surface-science growth and magnetic investigations, with materials' knowledge and applications?

The use of magnetic SA and SO can be classified into two categories. In the first category such structures 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, size, ...). In this case SA–SO systems are used as objects of very high quality to serve as model systems for analyzing fundamental issues of magnetism, preferably to lithography or microstructured materials. The major investigated issues are magnetic order and thermal excitations in reduced dimension (Section 2), the crossover from bulk towards single atoms for spin, orbital momentum and magnetic anisotropy energy (MAE) (Section 3), and finally micromagnetism (Section 4). These studies are of applied interest, because devices require the use of ever smaller nanostructures, whose properties must be understood and ultimately tailored. The second category consists in investigating whether SO and SA systems might be used directly for applications. One fundamental obstacle forbidding this is the loss of most magnetic functionalities at room temperature for very small systems due to thermal excitations. This motivated the development of growth processes that replicate vertically initially flat SO structures to increase their volume with no compromise on their lateral size (Section 5). Also, examples are given were SA–SO can be used to achieve materials with specific magnetic properties (Section 6).

For these two categories SA or SO systems might be used, although SO generally receives more attention. For a broad public the order and its fascinating beauty certainly account for this. From a scientific point of view, an organized system is associated with a low dispersion of size and shape, and thus of physical properties. This benefits both to fundamental investigations because one can measure large assemblies and consider macroscopic measurements as the amplified signal of a single entity, and to applications were small dispersions are usually required, e.g., for magnetic recording media.

Complementary information on magnetism may be found in other reviews about magnetic nanostructures [24–29]. Finally, notice that this manuscript does neither cover magnetic clusters fabricated by chemical routes [30] and their self-organization at surfaces, nor the use of pre-patterned substrates to align these clusters along certain features [31].

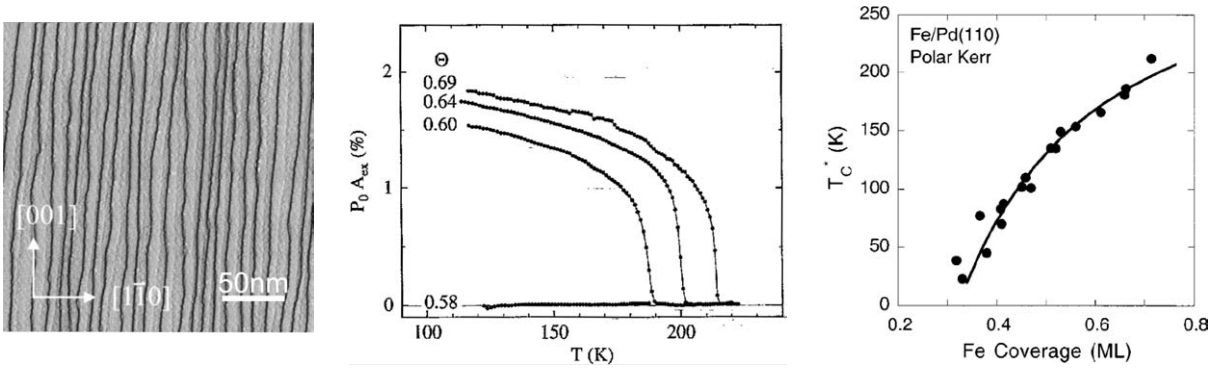
2. Magnetic order and thermal excitations in reduced dimension

2.1. Ferromagnetic ordering

Continuous ultrathin films were used in the past as model systems to study magnetic ordering in two dimensions (2D). More recently SA and SO were used to fabricate systems with reduced lateral dimensions, and thus study the cases of 1D (stripes and wires) and 0D (dots).

Spontaneous magnetic ordering in low dimensions relies on the presence of MAE. Already in two dimensions long-range magnetic ordering occurs only for the Ising model [34]. An xy system is able to order in a finite two-dimensional system [35],

[1] In the case of the spontaneous formation of nanostructures upon deposition at a surface we call SA the case where the nanostructures display no, or only short-range positional order, and SO the case where the nanostructures display a positional order with at least a middle-range order, typically over at least 5–10 items. Notice that this vocabulary is not universally used in the literature, and may also vary from one community to another. We will abusively use the acronym SO (or SA when appropriate) to stand for different expressions like self-organization, self-organize, self-organized.



(a) (b) (c)

Fig. 1. (a) Differentiated STM image of SO Fe/W(110) stripes with 0.5 AL coverage [32]; (b) in-plane remanent magnetization revealing a dramatic rise of *apparent* T_C (in reality, T_B, see text) upon percolation of SA Fe/W(110) dots around 0.6 AL; (c) finite-size scaling for SO Fe/Pd(110) stripes. The line is a fit with Eq. (1) [33].

whereas a truly isotropic Heisenberg system cannot establish long-range order at finite temperatures [36]. The MAE also influences the values of critical exponents. The rise of spontaneous magnetization per unit volume M_s close below T_C follows the scaling law $M_s \propto (1 - T/T_C)^{\beta}$, with $\beta \approx 0.125$ for 2D-Ising [34] and $\beta \approx 0.23$ for 2D-xy [35]. These exponents have been measured in many continuous ultrathin films and were found to agree with theoretical predictions of the Ising or xy models, depending on the type of MAE, either uniaxial (perpendicular or uniaxial in-plane) or easy-in-plane [24].

The fabrication of 1D systems by SO mostly relies on step-decoration of vicinal surfaces in the step-flow growth regime with sub-atomic-layer (AL) amounts of material [33,37–42]. The evolution from 2D towards 1D was pioneered by Elmers et al., who fabricated SO Fe(110) stripes on vicinal W(110) [37,38] and evidenced the finite-size scaling law [43]

$$T_C(n)/T_C(\infty) \propto 1 - (n_0/n)^{\lambda} \tag{1}$$

with $\lambda = 1.03$ in excellent agreement with $\lambda = 1$ predicted for an Ising system. n is the width of the stripes and n_0 the width for vanishing ordering at zero temperature, tentatively extrapolated to four atoms although (1) is in principle not applicable for small n. $\lambda = 1.2 \pm 0.3$ and $n_0 = 3$ was found for SO Fe stripes on vicinal Pd(110) [33] (Fig. 1(c)). The temperature dependance of magnetization was also carefully measured in the latter case, revealing enhanced magnetization decay for decreasing width. In strictly one dimension, even for an Ising system magnetic ordering is not predicted at finite temperature, which is consistent with $n_0 \simeq 1$. However hysteresis loops performed on SO Co/Pt(997) chains of monoatomic width and high uniaxial MAE, displayed remanence and coercivity below 15 K [44]. This apparent contradiction is easily lifted by noticing that ferromagnetic order can formally be defined only for systems of infinite size and under thermodynamic equilibrium. In experiments the length of the wires and the duration of the measurement are finite, so that single-domain states may occur when the correlation length exceeds the physical length of the wire and no major thermal excitation occurs during the measurement. Coercivity and remanence were also reported on flat finite-size dots [45,46]. Provided that their lateral size is smaller than all micromagnetic length scales and that the correlation length exceeds the dot's size, a nearly uniform magnetization state is expected. This is a so-called near single-domain state that behaves like a macrospin of moment $\mathcal{M} = M_s \times V$ were V is the volume of the system. Such dots may be classified as 0D as they are fully described by the degree(s) of freedom of the resulting macrospin only.

Apart from MAE, dipolar interactions also affect ferromagnetic transitions in low dimension. Positive interactions can stabilize ferromagnetism because of their long range, despite being much smaller in magnitude than exchange. The microscopic picture is the following. Exchange forces establish large blocks of parallel spins, owing to their strength at low range. However, in low dimension these blocks may fluctuate on a large scale. The blocks can freeze thanks to the long range of dipolar forces, despite their small strength, because what is to be compared with thermal energy in the dipolar energy of large blocks, not of individual spins. This was confirmed, again on an array of SO Fe/W(110) stripes [32] (Fig. 1(a)). In contrast with the case of non-interacting stripes [37] the signature of the dipolar forces here was a sharp transition of $M_s(T)$ around T_C despite a significant dispersion of stripes' width (see the finite-size scaling above), and the absence of relaxation even just below the freezing temperature. Because of the analogy with superparamagnetism, this effect was named *dipolar superferromagnetism*. This brings us naturally to the next subsection.

2.2. Superparamagnetism

So far we have discussed experimental results in terms of a ferromagnetic transition, which is a thermodynamic equilibrium concept. We have thus ignored kinetic effects, that we examine here, first theoretically, then experimentally.

Let us call ferromagnetic with Curie temperature T_C a system whose dimensions are all smaller than the correlation length for $T < T_C$. If a snapshot was experimentally feasible, it would reveal essentially uniform magnetization, with a direction lying along an easy direction of magnetization. At $T > 0$ K thermal fluctuations allow the system to explore its entire phase space. Thus after a lapse of time τ, $\mathcal{M}$ would statistically have overcome the energy barrier $E = KV$ associated with a hard axis, and would have settled in another easy axis direction. Using a simple Arrhenius law based on the Boltzmann probability of states occupancy we have:

$$\tau = \tau_0 \exp \beta K V \tag{2}$$

where $\beta = 1/kT$ and $\tau_0 \approx \times 10^{-10}$ s [48,49] is the so-called attempt period. Let us assume that the system is observed for a duration τ. For $T < T_B$, with $T_B = KV/[k_B \ln(\tau/\tau_0)]$ called the *blocking temperature*, the system has not switched over time τ and is said to be in a *blocked state*. For $T > T_B$ the system switches spontaneously in the interval of time τ. A magnetization measurement performed over that time scale reveals zero apparent magnetization, although the magnitude of the microscopic moment is still $\mathcal{M}$ at any time. This is the so-called *superparamagnetic* state. Analysis of remanent-less superparamagnetic magnetization loops can be done with *Brillouin-like* functions with $\mathcal{M}$ as an argument and allows one to infer $\mathcal{M}$. From this value V can generally be inferred because M_S is known. Notice that owing to the high MAE of most nanosized systems, in the range $T_B < T < 5T_B$ the function most fit to describe magnetization loops of magnetically-textured systems is closer to the Brillouin $1/2$ function than to the Langevin function [45,50–52]. Using the latter leads to an overestimation of V by a factor 3. It is also well known that neglecting the size distribution leads to an overestimation of the average V [45,52]. As a second step the measurement of T_B is sometimes used to deduce K. Let us emphasize that when the system's size is larger than the wall

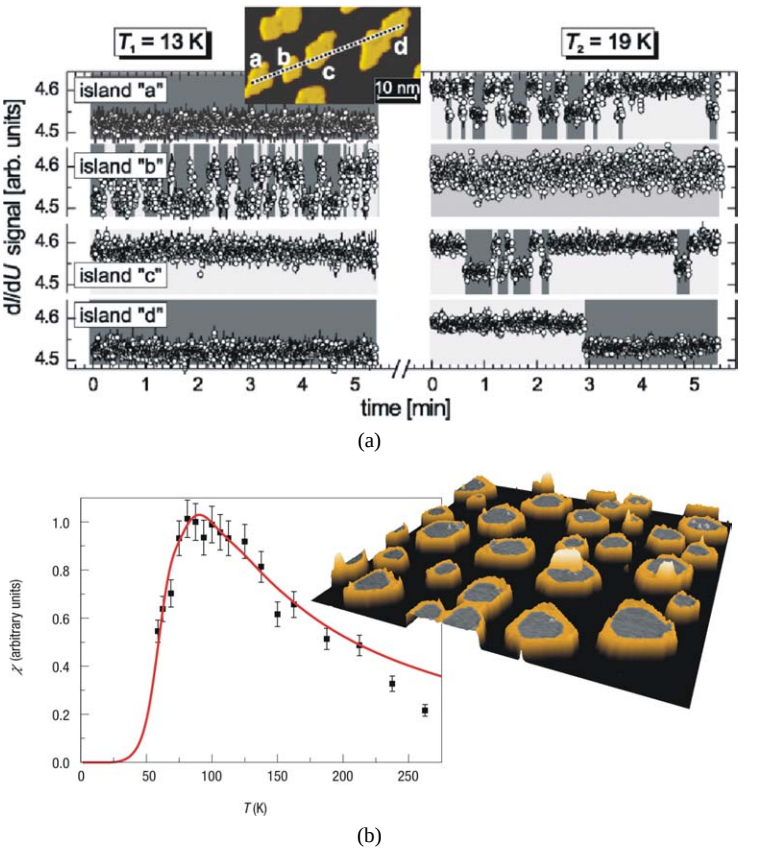


Fig. 2. (a) Spontaneous magnetization switching of SA Fe/Mo(110) dots (see inset) close to T_B, measured with Sp-STM [47]; (b) T_B of SA Co rings fabricated around Pt(111) dots (inset) determined with susceptibility measurements [45].

width λ, magnetization reversal is *not* coherent. In such a case the volume involved in the determination of T_B is close to that of a nucleation volume [53,54], of dimensions close to a wall width, not directly related to the total volume of the system. This can lead to an important overestimation of K.

A first consequence of what is said above is that $T_B < T_C$, and what can be measured practically in nearly all experiments is T_B, not T_C (notice, however, that attempts have been made to estimate T_C from the value of $\mathcal{M}$ inferred in the superparamagnetic regime, associated with the coherence length and then compared to the volume of the system [44].) This is the case of the 1D system of Co/Pt(997) mono-atomic wires reported above. The fact that $T_B < T_C$ was clearly illustrated in the case of in-plane magnetized Fe/W(110) dots self-assembled in the sub-AL range, and of lateral size 5–10 nm. These show an abrupt occurrence of spontaneous magnetization up to 190 K upon physical percolation of the dots into a film for coverage $\Theta = 0.6$ AL, whereas no spontaneous magnetization was observed for $\Theta = 0.58$ AL, down to 115 K. This effect cannot be explained by a finite-size scaling of the dots, and was attributed to superparamagnetism [38] (Fig. 1(b)). This abrupt transition was also observed for Co/Au(111) SO dots ordered on a rectangular array, percolating from 0D to 1D and then from 1D to 2D [55]. Recently small Fe/Mo(110) dots with perpendicular magnetization were magnetically imaged with spin-polarized STM (Sp-STM) (for the technique see [56]), revealing a blocked state below approximately 20 K [47] (Fig. 2(a)). The observation of telegraph noise between two states of high magnetization confirms that the macroscopic vanishing spontaneous moment is related to T_B, not to T_C. Notice that thermal activation of single nanosized objects was reported before and analyzed quantitatively with, e.g., micro-SQUID, a highly precise technique [57]. Direct information about the measured nanostructure was, however, lacking, which can now be brought with nanostructures grown at surfaces. The correlation between structure and magnetic properties is a traditional cornerstone of material improvement, and the same statement is expected to hold for nanomagnetism. An advantage of SA combined with a high-resolution direct imaging technique is that subtle changes in T_B can be associated with the shape of dots, more compact dots displaying a higher T_B, explained by a non-uniform magnetization reversal process. Systems with a strongly non-compact shape were measured, like Co rings fabricated by step-decoration of Pt(111) dots (Fig. 2(b)).

3. Orbital moment and anisotropy, from bulk to single atoms

In 1954 Néel predicted that symmetry breaking at the interfaces of magnetic thin films would induce an extra contribution to the MAE, named interface magnetic anisotropy, or Néel anisotropy. It yields a $1/t$ dependence of the total MAE density of a film with thickness t [58]. The experimental confirmation of this law was given in 1968 [59], with the foreseen possibility to overcome dipolar energy and yield perpendicular magnetization.[2] MAE was then predicted to scale with the anisotropy of the orbital momentum [60], which was checked experimentally on perpendicularly magnetized Au/Co/Au(111) [61]; see [62] for a review.

SA and SO opened the possibility to study this phenomenon quantitatively in even lower dimensions, i.e., the anisotropy associated with atomic edges (1D) and kinks (0D). This issue is of direct interest for applied nanomagnetism, as in ever smaller grains the main source of MAE will arise from surfaces, edges and kinks. A widely used technique is X-ray magnetic circular dichroism for its ability to separate spin from orbital momentum through the use of sum rules [62], and its high sensitivity, well below one atomic layer [63,64]. Pioneering work was done on SO Co/Au(111) dots [65,66]. Spin and orbital momentum were recorded as a function of the dot's diameter L. The $1/L$ dependance normalized per atom was then interpreted as an edge contribution. The spin contribution was reported to be mostly unaffected at edges in all publications. The orbital momentum was found to be non-affected at edges by Koide et al., whereas an increase of orbital momentum was reported by Dürr et al. In this publication if one uses the relevant function for fitting superparamagnetic curves to assess the volume of dots, i.e., Brillouin $1/2$ instead of Langevin used in the publication (see Section 4), we can estimate the ratio of edge over surface atoms for each measurement. Then we deduce that edge atoms bear an extra orbital moment of $0.5 \pm 0.2\ \mu_B$ as compared to bulk Co. A rise of orbital momentum of $0.5\ \mu_B$ was also determined in SO Fe/Au(111) dots [67], in quantitative agreement with the value of enhanced magnetization at steps on thin films [68,69]. However, in this case the interpretation is more difficult owing to the transition of Fe from a high-spin to a low-spin phase as a function of dot's size.

The real breakthrough came from the study of Co/Pt systems. First, the SO of Co was optimized on the vicinal Pt(997) surface, yielding stripes of width seven atoms for 1 AL coverage, down to mono-atomic width, i.e., wires [41]. Orbital momentum, its anisotropy, and the MAE could be measured as a function of the stripe width. The phenomenological contribution of edge atoms was then extracted [42,44]. An oscillatory behavior of MAE was also found [70], an effect similar to oscillatory behaviors well known in 2D. Similar studies were performed on small flat dots of Co/Pt(111) from one to some tens of atoms. The control of the size of the dots was achieved by first depositing at low temperature (15 K), where the motion of adatoms is hindered

[2] In most cases both true Néel-type and bulk magneto-elastic anisotropy contribute to this enhancement, which we will not discuss here. The resulting anisotropy will be abusively called *surface* anisotropy in this manuscript.

Table 1
Orbital momentum and magnetic anisotropy energy (MAE) of Co atoms on Pt as a function of coordination (after [44,64])

	bulk	mono-layer	bi-atomic wire	mono-atomic wire	two atoms	single atom
Orbital momentum (μ_B/at)	0.14	0.31	0.37	0.68	0.78	1.13
MAE (meV/at)	0.04	0.14	0.34	2.0	3.4	9.2

and thus only single atoms are found on the surface, followed by careful step-like annealings to increase the mean dot's size by diffusion-limited aggregation [52,64] (Notice that single atoms were first probed by XMCD prior to these experiments, for 3d atoms on alkali surfaces [63].) The outcome of various experiments from bulk to single atoms is summarized in Table 1. The orbital momentum, essentially quenched in the bulk, rises progressively above 1 μ_B in single atoms, along with the MAE. It is found that, from the point of view of orbital momentum and MAE, a cluster of two atoms (a bi-atomic wire, resp.) behaves closer to a wire (a mono-atomic slab, resp.) than to a single atom (a mono-atomic wire, resp.) (Table 1). This reminds us that the concept of dimensionality depends on the magnetic property studied: two atoms is closer to a wire when orbital momentum is concerned, but is closer to a single atom when magnetic ordering is concerned, see Section 2.1.

Model SO and SA systems like the Co/Pt structures presented above, are desirable to extract quantitatively fundamental properties like MAE. This knowledge being established, the question arises as to what extent it can be used to tailor the MAE of more versatile systems suitable for applications. It is sometimes argued that the increase of MAE in low dimensions will help to overcome superparamagnetism. The evaluation of the energy of nucleation volumes (Section 2.2) allows one to draw general trends, revealing that this is not the case. Let us assume that the MAE in any system is dominated by the contribution of lowest dimensionality, i.e., surface K_s for films and edges K_e for stripes. One finds that for continuous films of thickness t ($D = 2$) T_B is essentially independent of K_s and $T_B \propto t$; Concerning $D = 1$, for wires of more or less round section $L \times L$, $T_B \propto L^{3/2}$, while for flat wires of width L, $T_B \propto L^{3/4}$. Finally for $D = 0$, $T_B \propto L^2$ for compact clusters and $T_B \propto L$ for flat dots. Thus, in all cases a *decrease* of T_B is expected upon decrease of the system's size. The only hope resides in increasing the number of interfaces in a system of fixed size. Engineering SA to produce lateral superlattices via sequential deposition is a promising way. It has been demonstrated for 1D single rings for magnetic materials [45,52] (more generally, multiple concentric rings and lateral stripes of Ge/Si [71] were reported). This concept applies also to the case of spontaneously ordered lateral multilayers of FeAg and CoAg [72,73], see Section 6. Ultimately no strict borderline exists between this approach and the fabrication of high-anisotropy ordered alloys like FePt [74] or CoPt [10]. Tailoring the anisotropy with arrays of interfacial dislocations is another approach, as reported for Fe/W(001) nanostructures [75,76].

4. Model systems for micromagnetism

Micromagnetism is the field of study of magnetic domains and domain walls, both statically and under magnetization reversal or magnetic excitations, that typically spans in the range 50 nm–10 μm. Lithography is then the most relevant fabrication technique because of its resolution and versatility [25,78]. Nevertheless, UHV-fabricated nanostructures have niche applications, where they are better suited: (i) specific techniques can be applied that require a UHV-compatible surface. This is the case of Sp-STM, that allows both the highest available magnetic resolution, down to the atomic level [56], and the application of an external field of arbitrary value in any direction of space, which can mostly not be done with high-resolution microscopies based on electrons (SEMPA or spin-SEM, Lorentz, X-PEEM and SPLEEM); (ii) surfaces and edges can be fabricated avoiding defects arising during lithography and/or etching (amorphisation, oxidation, etching or resist-related loss of resolution), thus yielding high-quality nanostructures for model investigations; (iii) UHV fabrication may be more reliable than lithography, e.g., for nanometer-sized features [79] or, on the reverse, objects with a high vertical aspect ratio [80]. All these aspects are illustrated in Fig. 3.

The core of magnetic vortices could be imaged by Sp-STM in flux-closure states [77] (Fig. 3). Magnetic vortices had previously been detected by magnetic force microscopy in dots made by lithography [81]. However, UHV measurements with the high-resolution Sp-STM technique yielded the true width of the vortex core at the surface, 9 ± 1 nm for Fe(8 nm)/W(110), in good agreement with micromagnetic predictions [78]. The size of the core was found to expand or shrink upon application of an external field parallel or antiparallel, respectively, to the magnetization in the core, again in good quantitative agreement with micromagnetic predictions. Another topic is geometrical constrictions. Bruno predicted that domain walls may be compressed in geometrical constrictions, because the increase of exchange energy can be overcompensated by a decrease of the length of the wall, thus of its total energy [82]. The shrinking of domain walls in constrictions was confirmed in SO double-layers (DLs) Fe/W(110) stripes, e.g., from 6 nm in smooth stripes to 2 nm for a constriction ~ 1 nm wide and long [83]. Going to ever smaller scale, since conventional micromagnetism is a continuum theory, it breaks down at the atomic level. Discrete models

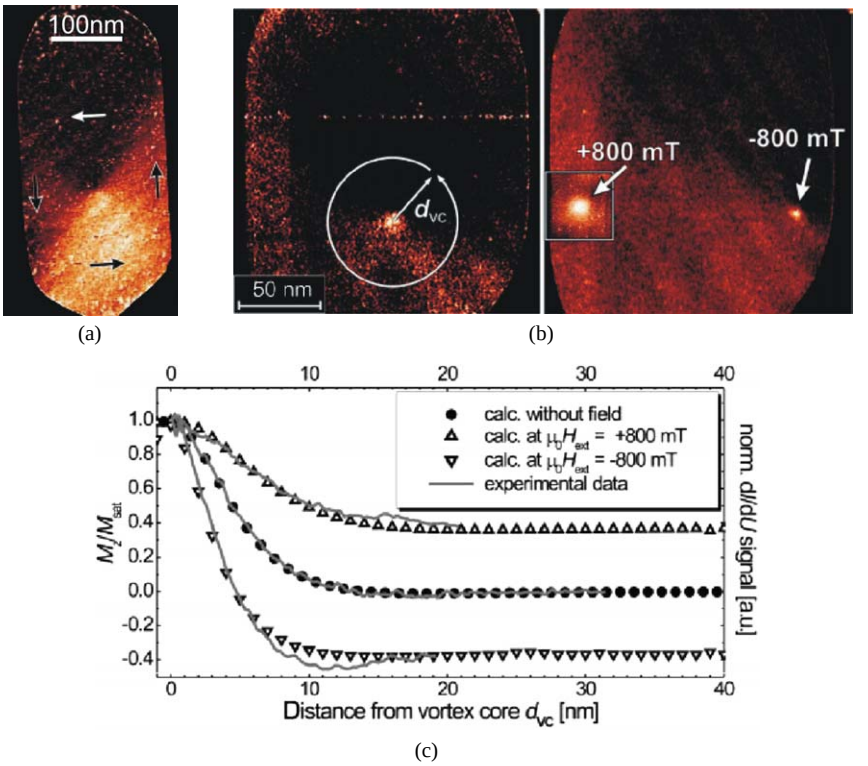


Fig. 3. Micromagnetism with Sp-STM using an Fe/W(110) dot [77]. (a) Overview of the flux-closure domain state, in-plane surface magnetization; (b) out-of-plane component for zero-field (left), and in-perpendicular-field (right, main for negative field, inset for positive field); (c) surface profile of the vortex core.

may be substituted, e.g., to describe extremely narrow domain walls in hard magnetic materials [84]. Such narrow domain walls could be imaged with Sp-STM on Fe(1 AL)/W(110), revealing a width of 0.6 nm using the asymptote of the cosine angle, a value that may still be limited by the spatial resolution of the technique. In such high anisotropy materials the wall profile depends solely on exchange A and MAE K. Thus, based on values of K measured by techniques like ferromagnetic resonance or torque-oscillatory magnetometer that can be applied down to the monolayer [85], values of the exchange could be extracted. On a larger scale, around 100 nm, geometrically-constrained domain walls were studied in cross-paths of SA Fe/W(001) wires using SEMPA, revealing the arrangements for the four possible topological incoming magnetic fluxes from the four arms [76].

The above-mentioned reports concern very small length scales. Another direction of research in micromagnetism consists in studying ever larger systems, to try to bridge the gap between nanostructures now understood quantitatively, and macroscopic materials still described phenomenologically. The use of SA is justified as larger systems become increasingly complex, so that it is essential to study model nanostructures to avoid an extra interplay of extrinsic effects related to defects. Studies at zero field concern the direct observation of the transition from the single-domain to vortex-state [86] in SA Fe/W(001) dots with a more or less square shape [87], more generally the evolution from single-domain to a variety of flux-closure domain states like vortex, Landau and diamond [78] in SA elongated Fe/W(110) dots [80,88,89]. Thermally-activated switching between metastable states was investigated in Co/Ru(0001) SA dots, namely single-domain and the so-called V-state [90]. A further step consists of the quantitative understanding of magnetization reversal in such multi-domain states. Nucleation and annihilation fields of magnetic vortices and walls were measured with a micro-SQUID in a single dot 30 nm-thick as a function of the in-plane direction of the external field. Discontinuities were demonstrated as a function of angle. Such jumps are sometimes thought to result from defects. Here, they could be ascribed, with the help of simulations, to intrinsic bifurcations related to the direction of the external field with respect to edges [91]. Very few experiments are available for thicker – thus more bulk-like – dots [92] whereas such systems can now be tackled with simulations [93]. Such studies are now in progress, revealing features that cannot be understood with 2D micromagnetics only [94].

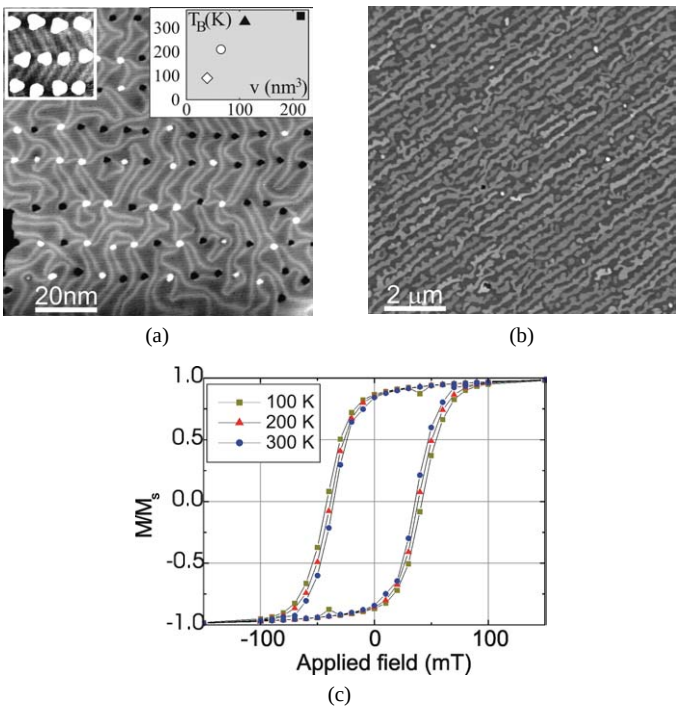


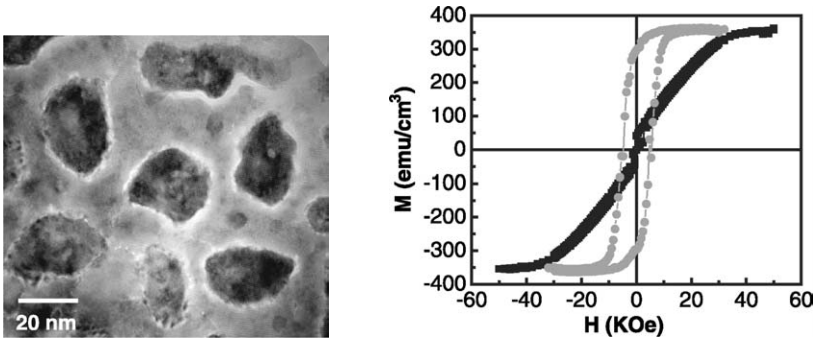
Fig. 4. (a) STM image of a SO Co/Au(111) system during the fabrication of pillars. Insets: conventional SO Co/Au(111) dots in the sub-AL range (left) and blocking temperature of pillars as a function of their volume [51] (right); (b) SO Fe/W/Mo(110) stripes, 4.3 nm-thick [95]; (c) magnetization loops of Fe/W(110), 5.5 nm-thick, along in-plane [001].

5. Thick self-organized systems: from surfaces to materials

It is still unclear whether epitaxial SA or SO of magnetic systems will bring any new useful functionality. Above all, there exists at the moment a fundamental obstacle against their use in devices. These systems have small lateral dimensions and are generally one or two ALs high only, so that they have an extremely small volume. Thus, they do not provide enough material for applications, and most important, they are all superparamagnetic at room temperature [21,40,44,55,64], see Section 3. We have discussed that the rise of MAE in low dimension is overbalanced by the decrease of volume, so that the issue becomes ever more acute for smaller nanostructures as the anisotropy barrier KV shrinks. At first sight V cannot be increased much, as for conventional deposition processes upon increasing the amount of material deposited percolation into a continuous film occurs [38,44,55]. The key may lie in engineered growth processes that were demonstrated to yield SO nanostructures much thicker than atomic layers while avoiding percolation, thus significantly increasing V without compromise on the lateral density, see below.

The first process was demonstrated for SO Co/Au(111) dots. Inspired by the vertical stacking of multilayers of quantum dots [96] sequential deposition of AL-fractions of Au and Co was performed. When Au just fills up the empty space between the Co dots, the dots from the next layer grow atop the existing dots, thereby increasing their height by one AL. This process is driven by immiscibility and a large lattice mismatch. Multilayers thus yielded pillars of height up to 8 nm and diameter in the range 3–5 nm [97,98] (Fig. 4(a)). Magnetization is essentially perpendicular, T_B increases monotonically with V and could be brought up to 350 K [51].

Another process makes use of strain fields arising in the vicinity of atomic steps. Upon deposition of Fe on vicinal cc(110) (cc = Mo,W) at the temperature of layer-by-layer deposition, a smooth film is formed except above the buried atomic steps of cc were trenches are formed. Then, upon annealing unwetting of the substrate occurs in registry with the trenches, yielding an ordered array of stripes with heights up to 5 nm [95] (Fig. 4(b)). Magnetically soft or hard (Fig. 4(c)) stripes were obtained by tuning the interface anisotropy. Thus the two main functional properties of ferromagnets could be demonstrated at room temperature, first coercivity-remanence, second the ability to break down into (stable) domains, whereas conventional Fe/W(110) stripes have $T_C = 179$ K [32]. Reports of trenches atop buried steps on other systems [99,100] suggest that this process might be quite general.



(a) (b)

Fig. 5. SO ferrimagnetic $CoFe_2O_4$ columns in a $BaTiO_3$ ferroelectric matrix: (a) plane view (b) magnetization loops performed at 300 K for perpendicular (shaded) and in-plane (dark) field [8].

6. Self-organization for material design

Systems reviewed in Sections 2 through 4 were used to study fundamental properties of nanosized objects. Concerning potential applications, SO and SA systems are more liable to be used as a whole, to achieve a material with specific properties, rather than for the direct use of each nanostructure independently. For instance, the anisotropy of continuous films can be tailored by deposition on patterned surfaces like step-bunched [101] and facetted [102] surfaces, on surfaces grooved in arbitrary directions after grazing-incidence ion sputtering [103], by shadow deposition on facetted surfaces [22,104,105].

Several types of systems with tailored magnetic properties can be fabricated by SO. Concerning SO from the deposit, arrays of parallel nanometer-sized Fe–Ag stripes and Co–Ag stripes are formed by co-deposition on Mo(110) [72]. These lateral superlattices display a high in-plane MAE along the wires, and magneto-transport with cpp geometry while the current flows in-the-plane [73]. The highest achieved T_B in these systems is currently 220 K, with an in-plane anisotropy field around 0.5 T [106]. The growth of an FeIr/Ir(001) lateral superlattice with $\sim$1 nm-period was also reported [107]. We have already noticed that there exists no strict borderline between nanoscale superlattices fabricated by SO and layered ordered alloys of high MAE-like L_10 phases [74]. Concerning these alloys, SA can be used to lower the ordering temperature owing to enhanced adatom mobility along the facets of dots [10]. This initial stage of nucleation can be used to fabricate at moderate temperature and upon percolation a grainy continuous film, partially ordered and thus with perpendicular anisotropy, found below 8 nm [108].

In Section 5 I have not included columnar growth because no clear borderline exists between SA and continuous films like CoCr-based columnar recording media, where SA can indeed be used to create a proper microstructure to adjust extrinsic properties like coercivity. Recent developments are columnar growth for oxides, e.g., $La_{0.8}Sr_{0.2}MnO_3/LaAlO_3$ [109] and $CoFe_2O_4$ ferrimagnetic columns with high perpendicular anisotropy due to the strain imposed by a $BaTiO_3$ ferroelectric matrix [8] (Fig. 5). One order of magnitude higher density could be readily achieved for hard-disk media with present technology if monodisperse media were available with no ordering defects, motivating demonstrations for such patterns of dots, like for Co/Au(788) [46,110]. An interesting area to follow in the future is the combination of prestructuring, e.g., using lithography, and self-assembly, an already ripe domain for semiconductors [111] but emerging for metals [31,112]. This would combine the versatility of lithography with the model features of UHV-deposited nanostructures.

7. Conclusion

This manuscript proposed a short overview of the use of self-organization (SO) and self-assembly (SA) for magnetic purposes. The studies performed fall into two categories. In the first category SO and SA are used to measured and analyze quantitatively low-dimensional magnetic phenomena. Beyond fundamental knowledge, these phenomena occur in systems of interest for application. However, they cannot be studied directly in these because of their extreme complexity, in terms of distributions, defects and microstructure. Thus, the idea is to get fundamental knowledge from surfaces, to be used to further tailor the properties of functional materials. The advantages of SO and SA over lithography are twofold. First, the high surface quality and cleanness allows intrinsic phenomena to be recorded, and also demanding techniques such as spin-polarized scanning tunneling microscopy can be applied. Second, the achievable size, down to the atomic level, lies much below that of any existing lithography technique. Issues that were investigated and reported here are magnetic ordering in 1D or 0D, spin and orbital momentum at edges and kinks, superparamagnetism in 1D or 0D. In the second category studies aim at bridging the gap between surfaces and materials, i.e., at showing that SO and SA systems might be useful directly in devices. These include the

fight against superparamagnetism, mainly by demonstrating processes for vertical growth of SA and SO nanostructures, and the tailoring of magnetic properties like anisotropy and the distribution of properties.

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