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# Magnetic Micro-Origami

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Additional information is available at the end of the chapter

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## Abstract

Microscopic origami figures can be created from thin film patterns using surface tension of liquids or residual stresses in thin films. The curvature of the structures, direction of bending, twisting, and folding of the patterns can be controlled by their shape, thickness, and elastic properties and by the strength of the residual stresses. Magnetic materials used for micro- and nano-origami structures play an essential role in many applications. Magnetic force due to applied magnetic field can be used for remote actuation of microrobots. It can also be used in targeted drug delivery to direct cages loaded with drugs or microswimmers to transport drugs to specific organs. Magnetoelastic properties of free-standing micro-origami patterns can serve for stress or magnetic field sensing. Also, the stress-induced anisotropy and magnetic shape anisotropy provide a convenient method of tuning magnetic properties by designing a shape of the micro-origami figures instead of varying the composition of the films. Micro-origami figures can also serve as building blocks for two- and three-dimensional meta-materials with unique properties such as negative index of refraction. Micro-origami techniques provide a powerful method of self-assembly of magnetic circuits and integrating them with microelectro-mechanical systems or other functional devices.

**Keywords:** micro-origami, nano-origami, self-assembly, self-rolling, magnetic micro-tubes, thin film patterns, magnetic thin films, magnetic anisotropy, residual stresses, magnetoelastic effects, magnetoelectric effects, multiferroic composites

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## 1. Introduction to origami and micro-origami

The meaning of the Japanese name “origami” is paper folding. Although the techniques of paper folding were developed in Europe and China in seventeenth and eighteenth centuries, they became the most popular in Japan. There is a vast literature on the history and practical instructions for assembly of three-dimensional objects from pieces of paper by folding [1, 2].

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Origami is primarily an art but it has also an impact on technology. The origami techniques were implemented in satellite solar cells, airbag design, and stent implants [3]. Currently, mathematical [4] and computer models such as TreeMaker [5] allow designing complex three-dimensional structures for decorative arts, as well as technological applications. Some examples of origami figures are presented in **Figure 1** [6].



**Figure 1.** Various origami figures (reproduced from Ref. [6]).

The traditional origami methods rely on folding a single rectangular piece of paper by hand. More advanced techniques take advantage of using tools, such as bone folder, clips, or tweezers. More complex designs require cutting the paper into various patterns (different from rectangle), coloring them, combining with other materials (e.g., aluminum foil), and using multiple pieces of paper, which is called a modular origami. In contrast to static designs, some origami patterns can possess movable parts, which can be activated by kinetic energy of human's hand or inflation [7]. Another interesting trend in origami is tessellation of the figures, which is in fact a method of assembly of larger figures from building origami blocks [8].

The smallest origami figures were made from the paper pieces as small as  $1 \text{ mm}^2$ , using a needle and optical microscope. At this point a question arises whether we can do even smaller micro-origami figures and use them to advance technology? The answer is "yes." It is possible to fabricate origami figures with features as small as  $2 \text{ nm}$ , actuate them, and assemble them into meta-materials. The submillimeter or submicrometer figures made from different materials using origami techniques are called micro- or nano-origami, respectively. A list of examples of applications of origami and micro-origami include bendable microelectronics, cell origami, origami nanorobot, pollen origami, origami lens, origami DNA, self-deployable origami stent, etc.

The materials used for micro- and nano-origami must be much thinner and tougher than conventional paper. Good candidates are thin foils and thin films. Although, in principle, it is

possible to use nanomanipulators and nanotweezers to bend and fold these materials, to make individual figures in microscale, this does not seem to be a practical approach for a mass production of various devices. Instead, other forces can be used which emerge at micro- and nanoscale. Adhesion forces, surface tension, or interfacial stresses start playing important roles when the size of the figures decreases below several micrometers [9].

Different materials and phenomena can be used for self-assembly and animation or actuation of origami figures. For example, shape memory alloys or polymers undergo significant deformations when heated by contact or radiation [10–13], capillary forces can deform paper structures [14], photosensitive polymers deform when exposed to laser light [15], and internal stresses produce deformations when the structure is released from the substrate.

Some of these forces can be used to create the micro-origami figures, while others can be used to produce motion of already formed structures. The microrobots made from stimuli-responsive materials can move, walk, or swim. These functions are especially useful in biomedical applications [16–22]. Magnetostrictive and multiferroic materials take a special place among the stimuli-responsive materials, because they couple magnetic, elastic, and electronic properties of the materials.

Currently demonstrated functional robots [21] have size of the order of centimeters, but biomedical applications require devices with dimensions smaller by at least two orders of magnitude. They should be made from thin films with submicrometer or nanometer thickness. Even the thinnest magnetic thin films preserve their magnetic and magnetostrictive properties in contrast to some shape memory alloys, which require certain thickness to undergo phase transition to produce their conformation. Therefore, magnetic films are good candidates for the micro- and nano-origami devices. In this chapter, we will discuss methods of fabrication of magnetic micro-origami structures, present experimental results on their physical properties, and indicate potential applications of magnetic micro-origami techniques.

## **2. Fabrication of micro-origami structures**

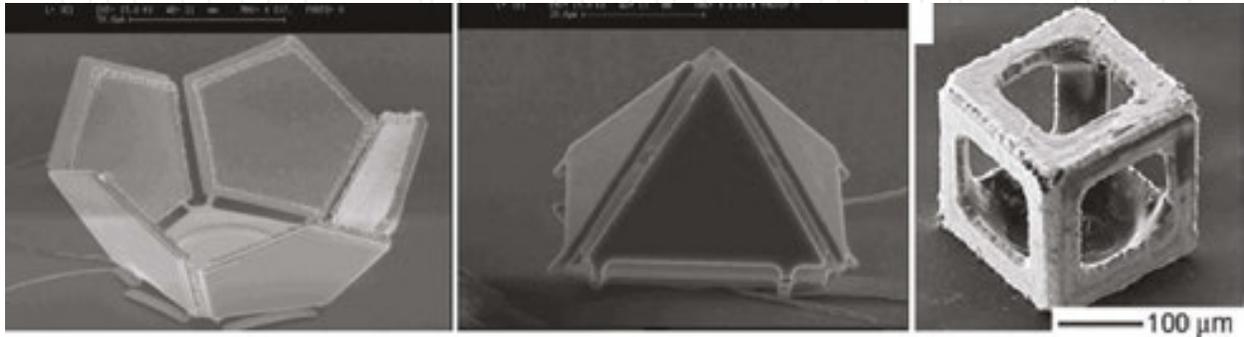
### **2.1. Forces responsible for conformations**

A large variety of forces is available to form or actuate thin film patterns into micro-origami figures. The magnitude of the force, elastic moduli, and the thickness of the film ultimately determine the curvature of the three-dimensional shapes. In addition, anisotropic stresses, shape of the pattern, and direction of etching decide about the direction of rolling or folding of the patterns.

#### *2.1.1. Surface tension*

Capillary forces or surface tension are negligible in macroscale; however, they are of primary importance in micro- and nanoscales. Gagler [21] and Gracias et al. [23–31] showed that the surface tension of evaporating drop of liquid is sufficient to pull the walls of the photolithographically defined thin film patterns together and form various figures, such as boxes or pyramids with the size of several micrometers. These patterns typically consist of more rigid

areas connected by thinner areas, which serve as hinges, when the thicker walls fold. This approach is versatile and can be applied to magnetic films or magnetic/nonmagnetic composites. Self-assembled structures can serve as building blocks for designing mesoscopic metamaterials with interesting responses to electromagnetic radiation in microwave, millimeter wave, and optical ranges [27, 28]. In some cases, the capillary forces can produce reversible conformations due to swelling and dehydrating of porous structures. Examples of micro-origami figures are presented in **Figure 2**.



**Figure 2.** Examples of self-folding micro-origami structures (left and middle picture reproduced with permission from Ref. [21] and the right picture from [23]).

At this point, it is worth noting that in some cases the surface tension may cause undesirable effects. For instance, it may oppose or prevent deformation of patterns which would otherwise deform due to internal stresses or thermal stresses.

### 2.1.2. Residual and interfacial stresses

Different types of growth of the polycrystalline materials may result in significant residual stresses in thin film materials [32, 33]. Island growth (Volmer-Weber) or Stanski-Krastanow growth of polycrystalline films may lead to significant residual stresses due to coalescence of grains. Also, growth of bilayered or multilayered films of different materials can produce interfacial stresses due to lattice mismatch between constituent phases. When the films with residual stresses are released from the substrate on which they were grown they undergo deformation.

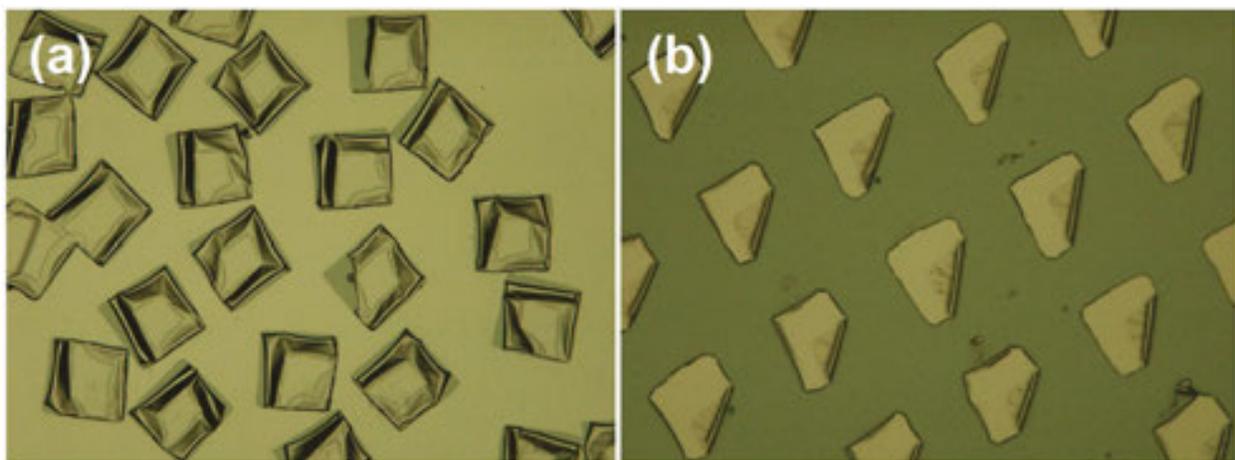
The curvature of the films depends on the internal stress level and the thickness of the film. For the simple bilayered beam, the continuum elasticity theory predicts that the curvature  $\kappa$  (inverse of radius  $r$ ) is [32].

$$\kappa = \frac{6E_1E_2(h_1 + h_2)h_1h_2\varepsilon}{E_1^2h_1^4 + E_2^2h_2^4 + 4E_1E_2h_1^3h_2 + 4E_1E_2h_2^3h_1 + 6E_1E_2h_1^2h_2^2} \quad (1)$$

where  $E_1$  and  $E_2$  are Young's moduli,  $h_1$  and  $h_2$  are thicknesses of the layers, and  $\varepsilon$  denotes the interfacial strain between the two layers. Based on this equation, it is easy to demonstrate the

general rule, which states that for the layers with similar thicknesses and Young's moduli the radius of the structures is proportional to the film thickness. Thus, the smaller origami figures we wish to design the thinner films we must use. Multilayered patterns or complex shapes require numerical solutions by the means of computer simulations to predict their conformations at certain level of residual stresses. The microstructure of thin films can vary with their thickness, which results in variation of residual stresses across the film thickness. Thus, even the single polycrystalline magnetic films can form magnetic microtubes [34] or other micro-origami figures.

If the tensile stresses in the bilayers are smaller near the substrate than on the film surface, the film released from the substrate tends to bend under and may form wrinkles rather than regular micro-origami patterns. An example of wrinkled squares of magnetic films is presented in **Figure 3**. This indicates that the sequence of the deposition of the films matters. For instance, depositing metal A on top of B will not result in the same structure as depositing B on A.



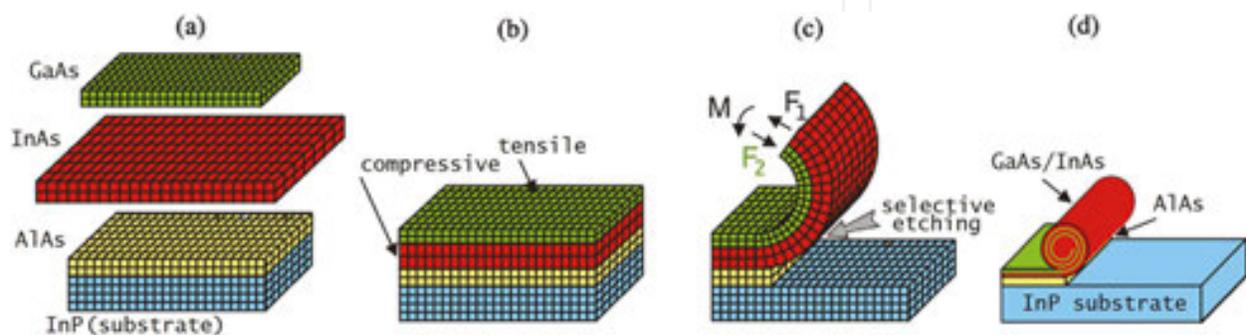
**Figure 3.** (a) Wrinkling of permalloy-Ti films and (b) correct rolling of the edges of Ti-permalloy squares.

The residual stresses due to grain coalescence depend on the deposition conditions, such as deposition method (sputtering, evaporation, or laser ablation), base pressure in the chamber, pressure of Ar during sputtering, and deposition rate or temperature of the substrate [33]. For this reason, it is difficult to control them and reproduce in different deposition systems.

Much better control of stresses can be achieved in heteroepitaxial structures. An excellent example of use of interfacial stresses is the technology developed by Prinz et al. [35–39].

They used misfit between single crystalline semiconductor films grown epitaxially on top of each other, as shown in **Figure 4**. Indium arsenide atoms arriving on the AlAs surface must reduce their interatomic distances to match the lattice of AlAs. Therefore, the InAs film on AlAs is in compressed state. On the other hand, GaAs atoms experience tensile stresses when they form single crystal film on the top of InAs with larger lattice constant. When AlAs sacrificial layer is selectively etched (with selectivity better than 1:1000), the initially compressed InAs expands and the strained GaAs shrinks resulting in bending and rolling of the

pattern of the bilayered film of InAs/GaAs which eventually forms a multiwall tube. For very thin films (a few atomic layers), the 7.5% mismatch strain between InAs and GaAs lattices results in a very small diameter of the tubes (down to 2 nm). Depending on the size of the rectangular pattern the number of turns can change between 1 and 40. The diameter of the tubes can be designed using formula (1). Here, it is important to note that ferromagnetic films such as single crystalline Fe or antiferromagnetic films such as Cr,  $\text{KCoF}_3$ , or  $\text{KFeF}_3$  can be grown epitaxially on top of GaAs [40]. In fact, a similar semiconductor template was used by the group of Mendach to prepare three-layered scrolls of InGaAs/GaAs/Au films with tunable plasma frequency in the optical range [41] and magnonic InGaAs/GaAs/NiFe films [42].



**Figure 4.** Heteroepitaxial semiconductor structures which result in formation of nanotubes of GaAs/InAs when sacrificial layer of AlAs on InP substrate is selectively etched releasing the film pattern from the substrate (reproduced with permission from Ref. [38]).

Thermal stresses in thin films provide another way to deform flat patterns after release from the substrate. Deposition of bilayers consisting of materials with markedly different expansion coefficients can be used to introduce internal strains when the bilayers are deposited at low or high temperatures and the films are released from the substrate at room temperature. This technique was used by Moiseeva et al. [43] to fabricate sophisticated cages from the patterns of magnetic films on top of thermal oxide with large compressive stresses. Some magnetic alloys called INVARs ( $\text{Fe}_{64}\text{Ni}_{36}$ ) take advantage of magnetostrictive properties to reduce their linear coefficient of thermal expansion by more than one order of magnitude as compared to other metals. On the other hand, some metals such as Zn or polymers have exceptionally large thermal expansion coefficients. Combination of these dissimilar materials can result in large strains associated with relatively small temperature range during deposition or in applications.

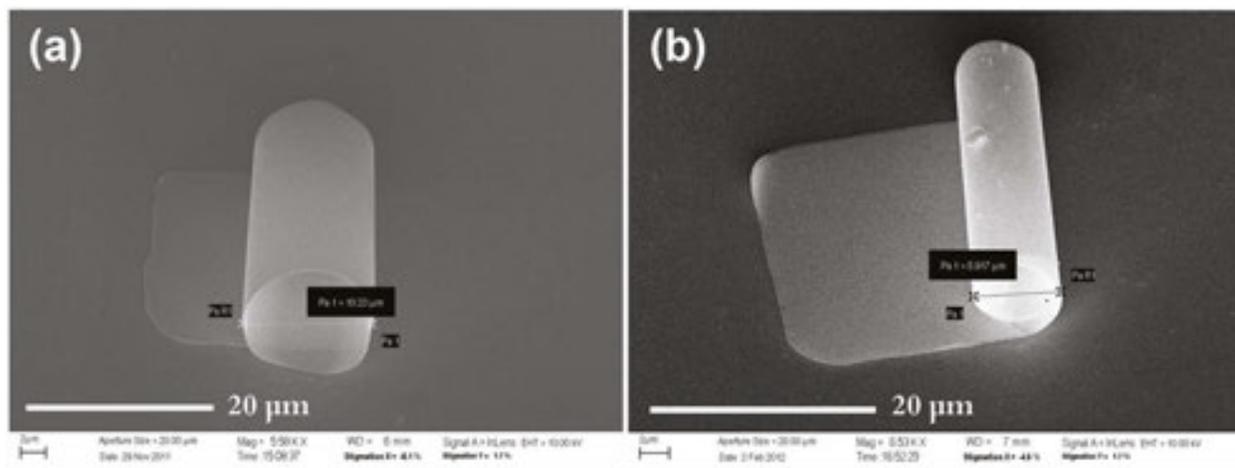
### 2.1.3. Other useful forces

There are more forces which have potential for interesting applications in micro-origami. External forces were used by Jackman et al. [44] to assemble millimeter-sized cubes. Deposition of magnetic films on strained substrates has advantage over other methods: uniaxial or biaxial stresses can be applied along different directions with respect to a pattern and the direction can be varied during deposition of subsequent layers. Application of external stresses can be realized by deposition of the films on bent or strained substrates. The stress level and distribution in the films can be predicted and highly reproducible (like in the heteroepitaxial

structures) and can be easily controlled in a broad range by adjusting the curvature of the substrate or external forces. A combination of polymers with relatively small Young's modulus, which can be easily stretched, and rigid inorganic magnetic films deposited on them seems to be suitable for this kind of micro-origami applications.

Finally, significant stresses in thin films can occur when the films undergo change of crystal structure or are chemically altered. Shape memory alloys, which deform due to reversible transition from the austenitic to the martensitic phase, fall into this category. A special place among memory alloys take magnetic memory alloys such as Ni-Mn-Ga [45], which can produce strain as large as 9% when subjected to the action of a magnetic field. Strains in giant magnetostrictive alloys are almost two orders of magnitude smaller (a small fraction of %) and are considered to be insufficient to efficiently form the micro-origami figures. However, magnetostrictive materials with even smaller magnetostriction coefficients, but large magneto-mechanical coupling, are expected to be effective in stimulating already formed structures to perform certain functions, such as vibrations.

Large changes of volume of the order of 30% occur in some materials during their oxidation [46]. This effect is analogous to swelling of polymers or other porous materials. The change of the volume is associated with the increase of the linear dimensions. To exemplify this method we demonstrate the effect of the Ti oxide formation on the diameter of self-rolled microtubes of Ti/FeGa/Au and Ti/FeNi/Au. It has been suggested by Nastaushev et al. [47] that oxidation of titanium in the process of removal of the sacrificial layer may produce stresses, which affect formation of the micro-origami patterns.



**Figure 5.** Effect of anodization of the Ti layer on the radius of the Ti/FeGa/Au film. (a) Original microtube, (b) anodized microtube. [48].

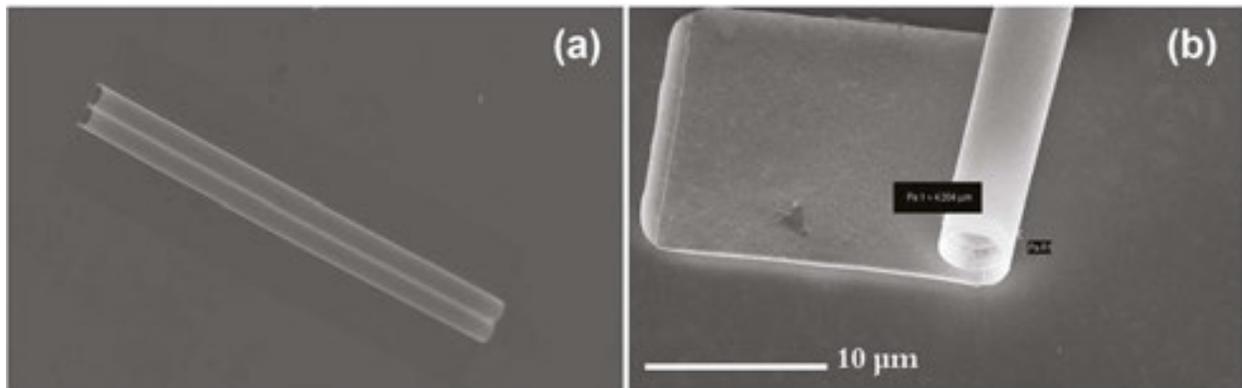
In our experiments, we used anodizing to completely oxidize Ti layer in already formed microtubes. A thin layer of Au was used to protect magnetic films from oxidation. The results of the experiment, displayed in **Figure 5**, show that the stresses from Ti layer, converted into TiO, reduced the diameter of the tubes to about a half of its original value. It was estimated that the strain associated with the oxidation was about 1% [48]. An interesting approach would

be to design processes in which magnetic metals such as Ni or Fe are chemically altered into magnetic compounds such as antiferromagnetic NiO or magnetite ( $\text{Fe}_3\text{O}_4$ ). This would allow engineering stresses in the structures simultaneously with their magnetic properties.

## 2.2. Controlling direction of self-assembly

As demonstrated by the origami, the same flat shape, such as rectangle, can be used to create hundreds of the origami figures. What differentiates these shapes is the sequence and direction in which the original shapes were folded. The fundamental question arises how we can control direction of bending or twisting of the flat patterns in micro- and nanoscale?

First of all, the shape itself may define the direction in which deformation occurs. In our early experiments with rectangular patterns of magnetic films grown on top of PMMA photoresist, we observed that only the longer edges of the AuCo film patterns rolled and formed long double tubes as shown in **Figure 6(a)**. This tendency can be interpreted in terms of the bending moment which for the films with uniform in-plane stresses is proportional to the length of the edge. Thus, when the etching progresses uniformly from all sides, the bending of longer edges prevails. The self-rolling of the same pattern will progress differently if a part of the pattern is attached to the substrate at the short edge, which prohibits rolling of the shorter edge. However, the constraint may not be effective for the rectangles with very large aspect ratio of the sides. Also, rigid parts of the patterns, such as thicker walls of origami figures from **Figure 2**, can act as constraints on bending the patterns. A theory of deformations of thin bilayered films has been developed by Cendula et al. [49]. They predicted that for certain stress gradient level a regular wrinkling is expected rather than bending. Computer modeling can be used for predicting formation of origami figures from more complex film patterns.

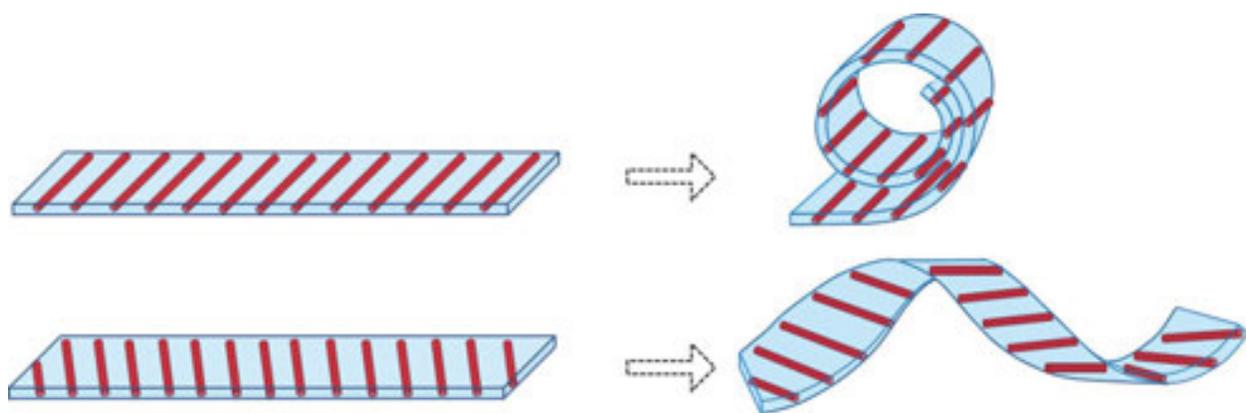


**Figure 6.** (a) Free-standing double tube of Cu/Co bilayer and (b) self-rolling of Ti/Ni bilayer which is attached at one end to the substrate and forms a single microtube.

Control of rolling direction is easier to achieve in heteroepitaxial structures. Prinz et al. [37] took advantage of different etching rates of Si in different crystallographic directions to fabricate microtubes or helical springs from GeSi/Si film patterns, which were deposited at different angles with respect to the crystallographic directions. Elegant experimental results by Li [50] illustrate different stages of the process of formation of an  $\text{In}_{0.3}\text{Ga}_{0.7}\text{As}/\text{GaAs}$  microtube

as the etching of the sacrificial layer progresses. They also demonstrate that anisotropic elastic properties in combination with selective anisotropic etching can be used to fabricate variety of three-dimensional structures by depositing film patterns at different angles with respect to (001) direction of the GaAs substrate.

It is also possible to achieve anisotropic lateral elastic properties of polycrystalline films by engineering their microstructure. This goal can be achieved by depositing films on tilted substrates. The angular deposition results in anisotropic magnetic properties [51]. Another option is to take advantage of nanocomposites. For instance, aligned carbon nanotubes embedded in polymer or magnetic film can control the pitch of the helical structures depending on the angle between the pattern and the direction of the enforcing elements (see **Figure 7**).



**Figure 7.** A concept of using anisotropic properties of composite materials to control rolling direction.

Finally, the external stresses applied to the substrates during the deposition are capable of providing a control over anisotropic film strains. The only inconvenience of this approach is the necessity of varying the stress level or direction inside a vacuum chamber, or preventing film contamination if the stresses are varied outside the chamber.

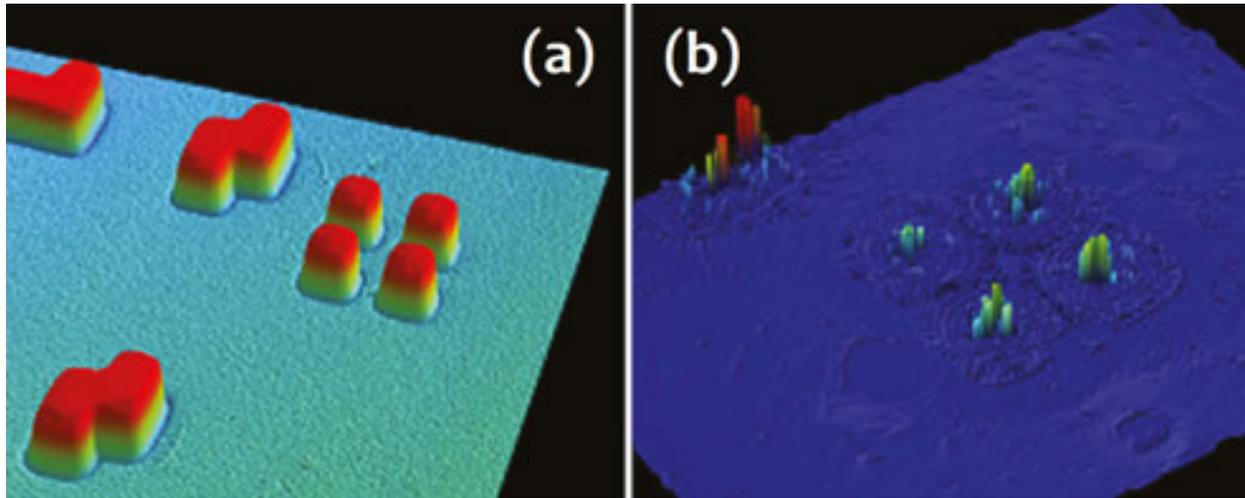
### 2.3. A sacrificial layer

Although it has been demonstrated that it is possible to assemble micro-origami structures on the air/water interface [52], vast majority of techniques uses solid sacrificial underlayers to build patterns on top of them.

In the case of the amorphous and polycrystalline sacrificial micro-origami structures, there are two fundamental requirements:

(a) The sacrificial layer must form a smooth surface, promote proper growth of the film, and enable patterning. It is worth mentioning that the microstructure of the polycrystalline films may strongly depend on the type of a substrate. Metals usually form smooth films when grown on top of a metal or semiconductor surface; however, when deposited on some dielectrics or organic materials they may exhibit enhanced roughness or columnar growth. An example is shown in **Figure 8**. The growth of the films can usually be improved by increasing the

temperature of the substrate with the sacrificial layer, but it can create a problem with some organic sacrificial layers which melt at relatively low temperatures.



**Figure 8.** (a) Al/Pt film pattern deposited on top of photoresist and (b) the same bilayered film deposited on top of glucose film (images obtained by optical profiler).

The sacrificial layer should also be insensitive to the agents used in the patterning process, so that the film can be released only after the pattern is defined. Depending on the patterning technique, the agents used for dry etching (reactive or corrosive gasses) or wet etching (water, acetone, and acids) should not attack the sacrificial layer.

(b) The sacrificial layer must be from the material which has markedly different etching properties than the film pattern. An excellent example is the ratio of HF solution etching rate of GaAs to AlAs exceeding 10,000. This sacrificial layer was used by Prinz et al. [35] and Li [50] to fabricate free standing semiconductor nanostructures. Even much smaller selectivity of 1:80 for Si/SiGe system etched by the 3.7 wt% of  $\text{NH}_4\text{OH}$  allowed fabrication of high-quality micro-origami patterns. Reactive gases, such as xenon difluoride ( $\text{XeF}_2$ ) vapor and  $\text{CHF}_3/\text{O}_2$ , were used to etch Si substrate [43] and to remove Ge layer [36], respectively. Polycrystalline magnetic films can be grown on a photoresist layer which can be dissolved by acetone [52]. Aluminum sacrificial surface layer was used to grow Au/Ti bilayers and was etched with KOH-based solution [47]. Our group used 50 nm thick Cu film on Si as a sacrificial layer to deposit magnetic films [53, 54].

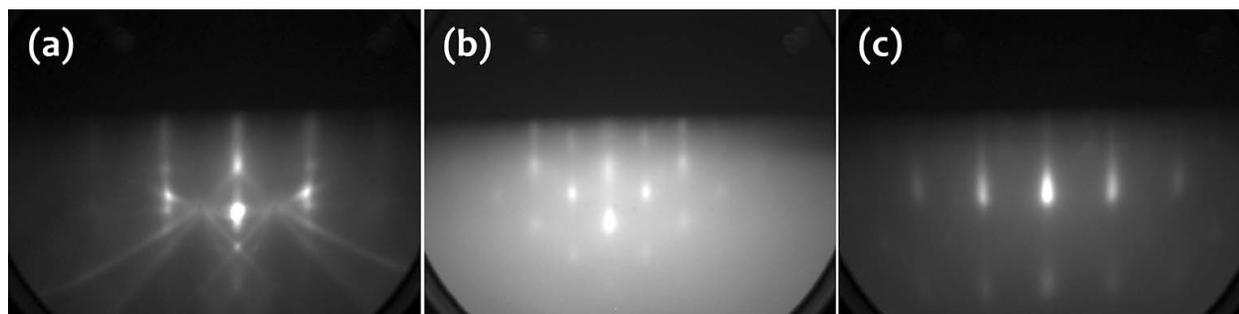
A disadvantage of wet etching method is that the surface tension or capillary forces of liquids used for the etching can interfere with the residual stresses and can prevent deformations of the pattern or even crack the pattern. The test for the strength of the surface tension forces in microscale is the fact that the capillary forces inside the tubes are able to collapse already formed tubes when water trapped inside the tube dries [36]. Potential solution to this problem can be the usage of supercritical conditions by elevating the temperature of water to a boiling point. Another option is to use organic solvents with much lower surface tension. Using gases

instead of liquids removes this problem. However, the gasses used for etching are highly corrosive and toxic.

Finding a proper sacrificial layer for the heteroepitaxial systems is even a greater challenge. In addition to the requirement of high selectivity, the sacrificial layer must also be a part of the heteroepitaxial system. This means that it must match both the structure of the substrate and the films which are grown on top of it. Satisfying all these three requirements simultaneously is a tough task.

This indicates a need for a search for new sacrificial layers for heteroepitaxial structures which promote single crystal growth and yet can be selectively etched. Ideally, such layers would be eco- and biofriendly.

Here, we would like to point at two prospective sacrificial layers. Single crystal NaCl (salt) sacrificial layer is a good candidate to grow magnetic films with cubic crystal structure. NaCl was a popular substrate for deposition of transition metal films in the 1960s [55], but it was not implemented in industrial processes because of its hygroscopic properties. Ironically, the same property predisposes NaCl films for the micro-origami applications. Reflection high-energy electron diffraction (RHEED) images of the MgO substrate, NaCl layer, and Cr film are presented in **Figure 9**.



**Figure 9.** RHEED images of (a) the MgO substrate with 5 nm MgO film, (b) 170 nm NaCl film, and (c) 15 nm Cr film on top of NaCl sacrificial layer.

The following procedure for the electron beam evaporation was used to grow heteroepitaxial structures with NaCl sacrificial layer:

- A thin layer of MgO (typically 5 nm) was deposited on MgO substrate to improve the quality of the surface.
- NaCl was evaporated at the rate of 0.05 nm/s at 350°C.
- Various magnetic and nonmagnetic transition metal films, such as Fe, Cr, V, and Ag, were grown epitaxially on top of NaCl.

Other substrates, such as GaAs and deposition methods can also be used to grow heteroepitaxial structures with NaCl layer. This layer is biofriendly and dissolves in water; therefore, it has a great potential for biomedical applications of micro-origami structures. Glucose films which can be made by spin coating of water solutions on different substrates are also an

interesting option for biomedical applications. However, the growth of some films may lead to unexpected results, as shown in **Figure 8**.

Zn and Mg are interesting materials for the sacrificial layers because they sublime in vacuum at relatively low temperatures. Therefore, they can be removed in vacuum chamber by increasing the substrate temperature. This approach does not require corrosive gasses or liquids which affect the release of the micro-origami structures because of the surface tension. We carried out initial studies on growth and sublimation of Zn films. Zn films have hexagonal structure and a reasonably good match to lattice constants of Ru, Ti, graphene, and, most importantly, magnetic films of Co. Although literature data [56] indicate that the temperature of sublimation of Zn is 250°C, the actual sublimation temperature was found to be substantially lower (below 200°C) in ultra-high vacuum conditions. Our RHEED studies revealed that single crystal or highly textured Zn films can be grown on Ru and Ti films deposited on sapphire substrates. It was also found that the thin film growth is strongly affected by the deposition conditions. Different morphologies of the Zn film due to different growth conditions, such as deposition rate and the temperature of the substrate are shown in **Figure 10**. The best results were achieved for the deposition of Zn on Ti film at deposition rate of 0.02 nm/s and at temperatures above 100°C.



**Figure 10.** Zn film on Ru deposited (a) at room temperature and rate of 0.02 nm/s, (b) room temperature and 0.1 nm/s, and (c) at 100°C at 0.05 nm/s.

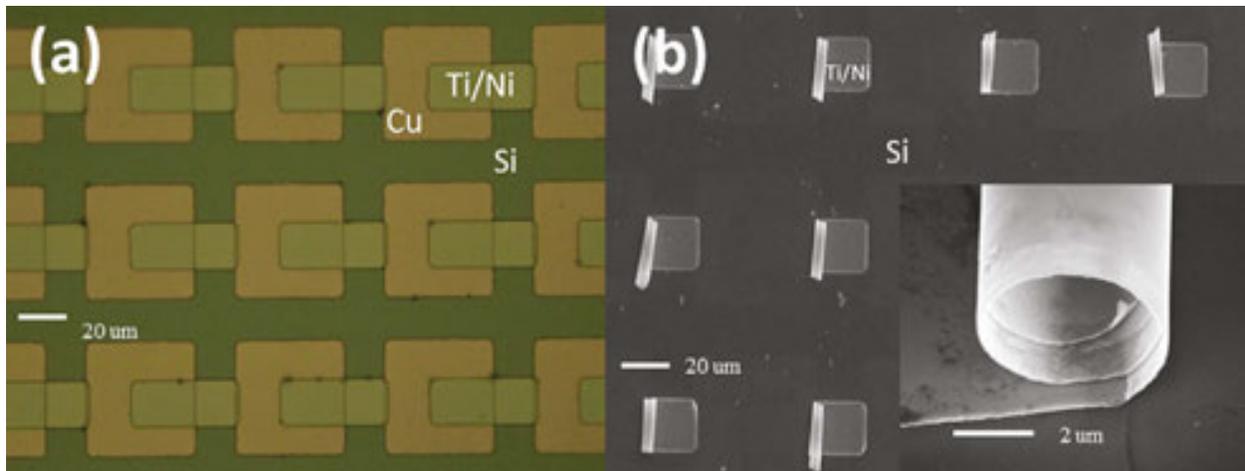
### 3. Link between the shape and magnetic properties

A discussion on static magnetic properties of magnetic scrolls has been initiated in the articles by Müller et al. [51, 57]. Angular deposition of Au/Co/Au films resulted in the in-plane magnetic anisotropy of rectangular thin film patterns. They ascribed it to stress-induced anisotropy in the as-deposited films. They also observed changes in the shape of the hysteresis loop and evolution of magnetic domain structure when the film patterns rolled and formed microtubes.

The films fabricated in our group [48, 53, 54] were isotropic in the film plane. Therefore, the effect of the self-rolling on the static magnetic properties was easier to interpret than for the

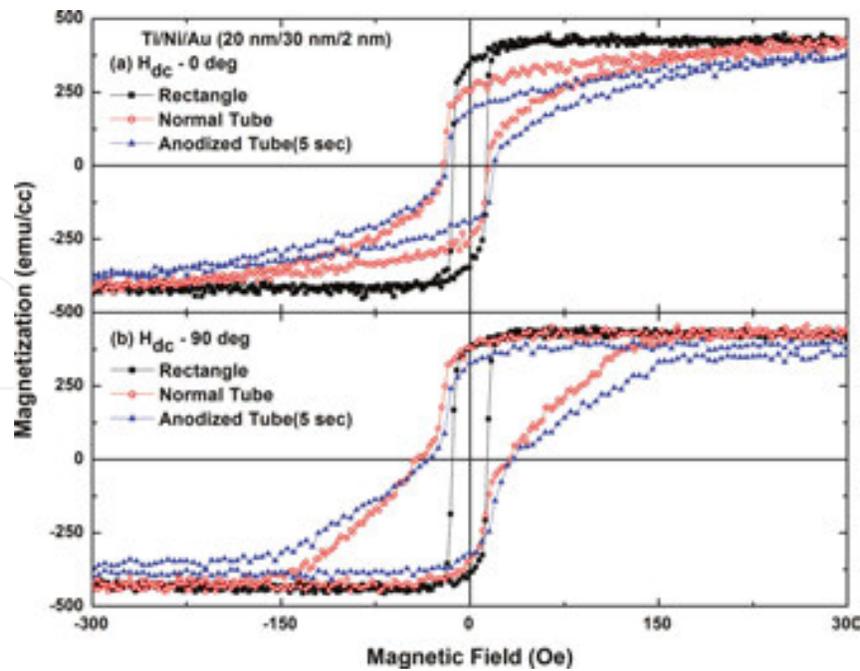
anisotropic films. Two kinds of magnetic microtubes were fabricated using the same technology described below.

First, the  $50\ \mu\text{m} \times 50\ \mu\text{m}$  holes were defined in a PMMA photoresist using exposure of the photoresist to deep UV light through as mask. Thin film of Cu was deposited by sputtering on the Si wafer with (001) orientation. During a lift-off process the Cu film pieces on top of photoresist were removed by ultrasonication of the wafer immersed in acetone. The remaining 50 nm thick Cu squares served as sacrificial layer. Another mask with  $20\ \mu\text{m} \times 50\ \mu\text{m}$  rectangles was used to deposit the trilayered Ti/GaFe/Au and Ti/Ni/Au films. This mask was aligned in such a way that the majority of the area of the rectangular holes in photoresist was on top of the Cu squares and a smaller part on exposed Si. After deposition of the trilayer and the second lift-off process the rectangles remained on top of Cu patches, which were partially attached to Si, as presented in **Figure 11(a)**. During selective etching of the sacrificial Cu underlayer, the films with residual stresses, caused by the grain coalescence, self-rolled, and formed magnetic microtubes with 3–4 turns, diameter of 5–10  $\mu\text{m}$ , and the length of 20  $\mu\text{m}$  (**Figure 11b**).

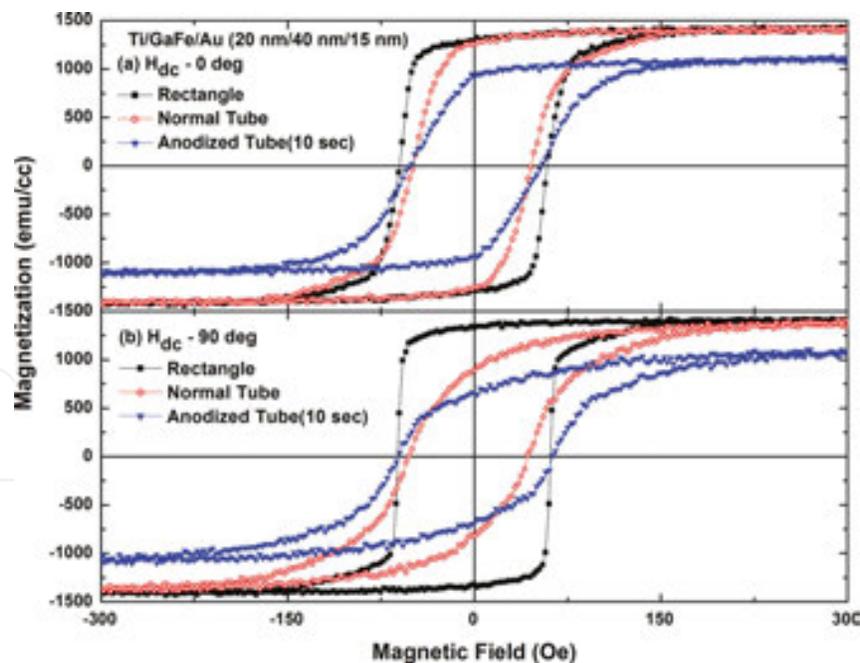


**Figure 11.** (a) Optical microscope image of an array of flat rectangular film patches of Ti (20 nm)/Ni (30 nm)/Au (2 nm) on top of Si wafer and partially overlapping with sacrificial Cu films squares. (b) Scanning microscope image of the array of magnetic scrolls and a magnified image of a single scroll (in the inset) formed after selective etching of Cu.

The same masks were used to deposit Ti/FeGa/Au films. A thin layer of gold was used to protect magnetic layer against oxidation. The static magnetic properties were characterized by means of vibrating sample magnetometry. The hysteresis loops of the microtubes with Ni are presented in **Figure 12**. The magnetic field was applied in two transverse directions in the substrate plane. The direction marked as  $0^\circ$  is the direction of the axis of the tubes formed from the patterns and the  $90^\circ$  is transverse to the tubes. Hysteresis loops (black lines) representing magnetization of flat patterns in these two directions are almost identical. Slight differences between the curves can be attributed to the shape anisotropy of the rectangular shapes of the patterns. Similar statement refers to the hysteresis loops of the flat patterns of the Ti (20 nm)/(GaFe 40 nm)/(Au 15 nm) film. The coercive field of the patterns with Ni was 13.4 Oe, whereas it was 60 Oe for the GaFe films.



**Figure 12.** Magnetization hysteresis loops of rectangular film patterns  $20 \mu\text{m} \times 50 \mu\text{m}$  with Ni layer (black line), microtubes (red line), and anodized microtubes (blue line). Top part refers to the  $0^\circ$  angle and the bottom to  $90^\circ$  angle of the applied magnetic field. (Reproduced with permission from Ref. [48].)



**Figure 13.** Magnetization hysteresis loops of rectangular film patterns  $20 \mu\text{m} \times 50 \mu\text{m}$  with GaFe layer (black line), microtubes (red line), and anodized microtubes (blue line). Top part refers to the  $0^\circ$  angle and the bottom to  $90^\circ$  angle of the applied magnetic field. (Reproduced with permission from Ref. [48].)

Marked differences between the magnetization loops for the two directions of magnetizing field were observed for both types of magnetic materials (red symbols curves in **Figures 12** and **13**). This is the result of the change of the shape and evolution of strains of the patterns undergoing deformation after release from the substrate.

As discussed in Ref. [48], the behavior of the GaFe films could be interpreted in terms of the shape anisotropy of the microtubes for which the technical magnetization saturation is achieved at lower fields for the field applied along the easy magnetization axis (along the tubes), whereas the loops for the hard direction (perpendicular to the tubes) are tilted and approach saturation magnetization at higher fields. However, the behavior of Ni tubes opposes that of GaFe, in spite of similar contribution from the shape anisotropy. In addition, both types of materials exhibit opposite trends in the change of the coercive field. The coercivity of Ni tubes almost triples for the field applied at 90° while it decreases by about 30% for GaFe films as compared to the rectangular patterns measured in the same direction. These facts prove that the change of shape and associated shape anisotropy alone is unable to explain the changes of magnetic properties.

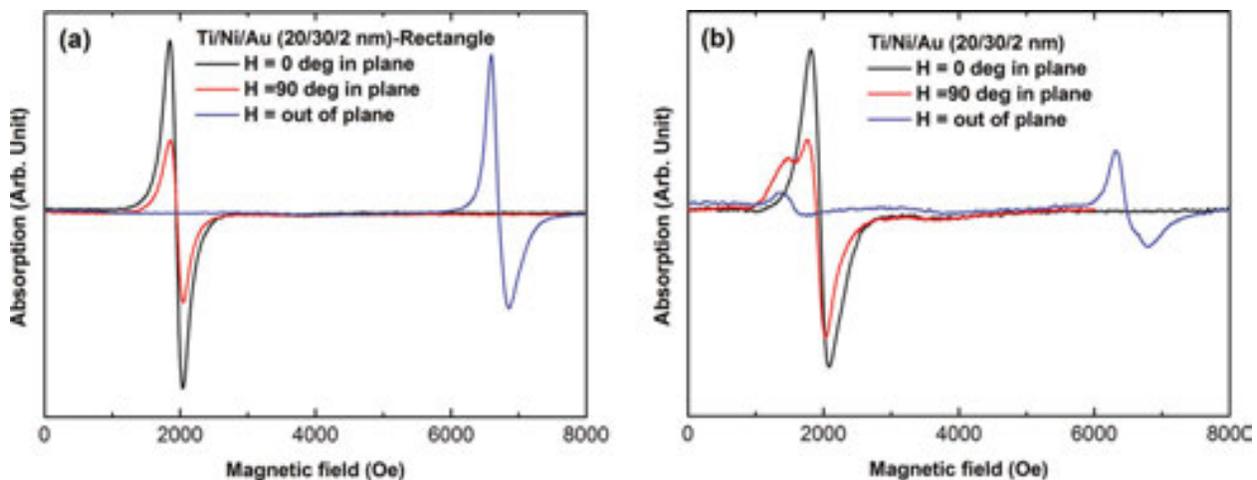
An important distinction between these two magnetic materials is their magnetostriction. Ni has negative linear magnetostriction coefficient of  $-38 \times 10^{-6}$ , whereas GaFe has positive coefficient of magnetostriction of about  $+70 \times 10^{-6}$ . For this reason, the magnetization responds in different ways when strains in the applied magnetic field vary by about 1% during self-rolling process.

Additional evidence of the importance of the magnetoelastic contribution to the magnetization change is that the differences between the materials continue to diversify when Ti layer undergoes anodization. Expanding TiO, formed from Ti layer, exerts additional stress on the magnetic layers and reduces the diameter of the tubes by about 50%, which is also reflected in magnetic properties. The time of the anodizing was adjusted in the range from 10 to 25 s so that the entire layer of Ti could be converted into Ti oxide. The reduced saturation magnetization for the anodized samples (see **Figures 12** and **13**) indicates that the oxidation partially affected GaFe and Ni. The saturation magnetization was unaffected by the fabrication processes (coating with PMMA, using a water solution of a developer, and washing with acetone) because of the presence of a protective layer of gold.

Another magnetic method, which gives even more insight into residual stresses, is based on high-frequency measurements of spin dynamics. Spin precession at Ferromagnetic resonance (FMR) is affected by local magnetic field  $H_{eff}$ , where demagnetizing field  $H_D$  and stress-induced anisotropy field  $H_\sigma$ , associated with stress  $\sigma$ , contribute. The relation derived from Landau-Lifshitz-Gilbert equation for magnetically isotropic material with magnetostriction  $\lambda_s$ , gives the relation between the stress-induced anisotropy field  $H_\sigma = 2\sigma\lambda_s/M_s$  due to stress  $\sigma$ , saturation magnetization  $M_s$  of the material, applied bias field  $H_A$  and a frequency  $f_r$  of microwave absorption at resonant conditions:

$$f_r = \frac{\gamma}{2\pi} \sqrt{(H_A + H_D + H_\sigma)(M_s + H_A + H_D + H_\sigma)} \quad (2)$$

where  $\gamma$  is a gyromagnetic ratio of the magnetic material. The distribution of internal stresses in magnetostrictive materials determines the linewidth of the resonant curve. Examples of FMR spectra for rectangular patterns of Ni films with thickness of 30 nm and for corresponding microtubes are presented in **Figure 14**. The resonant fields are approximately the same for the two orientations of the in-plane applied field. Larger resonant field for the out-of-plane direction of the applied field results from large demagnetization factor for this orientation. The FMR absorption spectra become more complex for rolled-up patterns. The resonance peak along the tube axis remains unaffected, except of some broadening. However, the curves measured with the field applied transverse to the microtubes are markedly different. In particular, the intensity of the absorption measured normal to the substrate decreases and additional peak forms at lower fields. This peak can be attributed to the microtubes. The high field peak may result from the part of the flat patterns which did not roll—they attach the tubes to the substrate. In the Ni tubes, the “unrolled” area was about 40% of the rectangles, but we were able to reduce it to 10% for GaFe films by adjusting technological parameters.



**Figure 14.** Ferromagnetic resonance spectra of the rectangular patterns of Ni films (a) as compared to the spectra of the microtubes (b) measured with the field out of the substrate plane (blue curves) and two directions in the film plane (field at 0° along the tubes and at 90°). (Reproduced with permission from Ref. [54].)

More detailed studies of spin dynamics in magnetic microtubes made by Balhorn et al. [42] involve spin wave properties in multiwall magnetic microtubes prepared by micro-origami techniques. They observed a series of four resonant peaks which corresponded to the constructive interference of Damon-Eshbach-type spin waves traveling along the circumference of the multiwall tubes of 20 nm permalloy film. The resonances could be tuned by the diameter and the number of turns of the tubes.

Magnetic layers can also be a part of multiferroic or magnetoelectric composites. These materials exhibit coupling between polarization and magnetization through the interfacial stresses between piezoelectric and magnetostrictive layer. Changes of external magnetic field produce magnetostrictive strains in the magnetic material which are transferred to the mechanically coupled piezoelectric and result in changes of polarization. On the other hand,

electric field applied to the piezoelectric layer produces piezoelectric strains which are transferred to the magnetic material and change its magnetization [58]. A common problem in the multilayered films is that the magnetoelectric functions are greatly reduced because either piezoelectric or magnetostrictive layer is attached to a massive substrate which prevents straining of the materials adhering to Origami technology. This is called a clamping effect. An advantage of free-standing micro-origami structures is that they do not restrict the exchange of stresses between the piezoelectric and magnetic phases and make the stress-mediated mechanism of electric and magnetic energies highly effective. In addition, magnetic scrolls occupy much smaller area than the cantilevers with the same mass. Piezoelectric/ferromagnetic composite Au (5 nm)/AlN (20 nm)/CoFe (40 nm)/Au (5 nm) form microtubes when released from the substrate [54].

## 4. Functions and applications

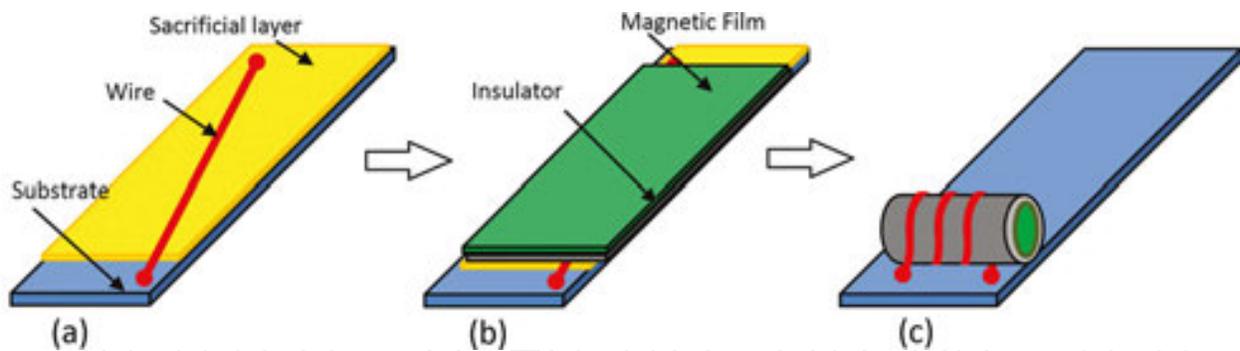
### 4.1. Functionality of the micro-origami structures

Converting flat pattern into micro-origami figures increases their functionality and potential for applications. For example, when magnetostrictive films are attached to the rigid substrate, the only response of the magnetic material to the applied field is a change of its magnetization. For a free-standing micro-origami structure, in addition to the change of magnetization, magnetic force can actuate the structure; it can give rise to a change of shape due to magnetostrictive strains and external strains can vary magnetization, as well.

As already mentioned in the previous section, there is a mutual link between the magnetism and shape of the micro-origami figures. This feature can be used for tuning magnetic properties or resonant microwave absorption of the micro-origami patterns. The shape of the hysteresis loops, magnetic anisotropy, and coercivity can be controlled by fabricating materials with different shapes rather than changing the composition of the magnetic material. Magnetic shape anisotropy can control anisotropic behavior of nonmagnetostrictive materials, whereas residual stresses in the magnetic layers play primary role in magnetostrictive structures. Frequency of ferromagnetic resonance and frequency of spin wave modes can be controlled by the number of turns and the diameter of the microtubes. Because the micro-origami shapes can be fabricated in the form of arrays of identical complex objects, they may exhibit interesting responses to the electromagnetic radiation.

Helicity of magnetic scrolls has been utilized to make meta-materials with negative index of refraction [59]. Similar arrays of free-standing helical structures were envisioned as the way of enhancing magnetic field for magnetic resonance imaging [60].

Micro-origami techniques provide an efficient method of self-assembly of micro- and nano-structures. Just to exemplify this statement, we propose in **Figure 15** the sequence of processes of assembly of a microscopic electromagnet consisting of a coil with a hollow magnetic core.



**Figure 15.** Three stages of fabrication of three-dimensional inductor: (a) Deposition of a wire on top of a sacrificial layer, (b) deposition of insulating and magnetic layers, and (c) self-assembly.

Multiple turns can be fabricated by adding more wires or using diagonal direction of self-rolling to produce helical springs like in Refs. [50] and [61].

It greatly simplifies fabrication of three-dimensional patterns, which usually would require many more masking and deposition processes, angular deposition, and deep etching. In addition to self-assembly of structures attached to the substrate, micro-origami figures disconnected from the substrate can serve as building blocks for meta-materials, where magnetic elements of the figures or surface tension at hydrophilic parts can be used to put these blocks together in two- or three-dimensional networks [26]. Here, tessellation methods of origami patterns can be used.

#### 4.2. Prospective applications

Micro-origami techniques have a great potential for numerous applications, although they have not been implemented in large-scale production, yet.

Magnetic and magnetoelastic properties of the micro-origami patterns can be used in various magneto-electro-mechanical systems (MEMS) such as stress or strain sensors, microscopic actuators, magnetic field sensors, and optical shutters [62]. As described in the previous section, enhanced performance of magnetoelectric functions is expected in the free-standing micro-origami patterns as compared to the planar structures on thick substrates. This gives a promise for increased sensitivity of the multiferroic composites for magnetic field sensors [63], which have potential to reach the sensitivity of SQUID magnetometer. Another potential microelectronic application is in microwave devices based on laminated multiferroic composites such as filters where the frequency of microwave absorption peak at ferromagnetic resonance can be tuned by the means of voltage applied to the piezoelectric layer [64]. Micro-origami design can reduce size and increase tunability of laminated multiferroic composites in such devices.

Because of excellent mechanical performance of microtubes [57], the self-assembly of micro-origami structures can be used to build parts of microfluidic systems. They can serve as ducts and microchambers for chemical reactions. Magnetic and magnetostrictive properties of the tubes can be advantageous for the liquid pressure sensing or measuring flow of liquids or

gases [65]. Micro-origami techniques can also be used to assemble parts of micropumps and valves in microfluidic systems, where either magnetoelastic or magnetostatic energies can be used to actuate micromotor or control shutters.

The most recent publications on micro-origami structures emphasize prospective applications in the biomedical field and biotechnology. First of all, the micro-origami methods are capable of fabricating biomimetic materials. Magnetic materials are envisioned as candidates for targeted drug delivery [23]. Different magnetic cages [21, 23] can be loaded with drugs, dragged with the help of magnetic force to the infected organs, and released in the location where they are the most effective. The targeted delivery of drugs can be achieved with the assistance of microswimmers [16, 66], which are able to move against blood stream. Helical magnetic structures made by micro-origami techniques can function as microdrillers to remove blood clots in the veins when remotely activated by a rotating magnetic field [67]. Microgrippers and magnetic tweezers take advantage of self-folding of micro-origami patterns, as mentioned in the introduction [29].

More advanced devices which combine microtubes with giant magnetoresistive sensors or semiconducting chemical sensors are available [68, 69]. Magnetoresistive sensors prepared by micro-origami method were capable of detecting single magnetic nanoparticle passing through a microtube. This will allow a precise control of functionalized magnetic nanoparticles for drug delivery or hyperthermia.

## 5. Conclusions

Micro-origami techniques are very versatile and can be applied to various materials including magnetic materials. They are envisioned as a very powerful self-assembly method. Unlike traditional origami, where folding of a paper is done by hands, micro- and nano-origami methods take advantage of forces in the microscale, such as surface tension or residual stresses, to shape materials by bending, twisting, and folding thin film patterns. The curvature of the structures and direction of deformation can be controlled in wide range by the magnitude of the residual or interfacial stress, elastic properties of the patterns, the shape, and thickness of the film patterns. This makes possible fabrication of complex three-dimensional architectures from flat patterns, which are released from the substrate by selective wet etching, reactive gas etching, dissolving, or sublimation of the sacrificial layer.

Magnetic micro-origami has a special place among various micro-origami designs. Magneto-static interactions between magnetic parts of the structures and/or external magnetic field can be used to assemble building blocks into meta-materials or actuate parts of microrobots. The external field gradient can be used to direct various micromagnetic cages loaded with drugs to infected organs, where the drugs will be released. Microswimmers take advantage of alternating fields to move in liquids in controlled way and rotating magnetic field can be used to rotate microdrills to open blood clots in veins.

The magnetic properties of the micro-origami structures can be affected by their shape in three ways:

- Shape magnetic anisotropy varies when the flat patterns form three-dimensional structures. An example was provided where flat patterns possessed isotropic magnetic properties when measured with the field along different directions in the film plane, but magnetic hysteresis loops and ferromagnetic resonance curves showed marked differences for the transverse configuration of the applied fields when magnetic patterns scrolled and formed microtubes.
- Magnetoelastic properties affect magnetization of magnetostrictive films when the stress level and/or direction in the films change. This refers to the relaxation of residual stresses in the magnetic material, as well as external stresses from the adhering nonmagnetic layers exerted on magnetic layer when the multilayered film patterns bend after release from substrate. The stress-induced anisotropy affects domain structure and magnetization processes. For example, self-rolling of magnetic films with positive (FeGa) and negative magnetostriction (Ni) lead to different trends in magnetization changes as evidenced by **Figures 12 and 13**.
- The change of chemical composition of magnetic films can alter magnetic properties. For example, the ferromagnetic Ni subjected to oxidation changes into antiferromagnetic NiO, and soft magnetic Fe may evolve into magnetite ( $\text{Fe}_3\text{O}_4$ ) with larger magnetocrystalline anisotropy, coercivity, and saturation field, but reduced saturation magnetization. The changes of magnetic properties of chemically altered films can be associated with enhancement of residual stresses which are responsible for the formation of the micro-origami patterns. Thus, in certain cases the changes of magnetic properties can promote change of the shape if the chemical changes are followed by the phase or composition change which produces deformation.

The changes of magnetic shape anisotropy and stress-induced anisotropy, associated with the deformation of the film patterns, provide a convenient method of tuning magnetic properties by shape design.

The coupling between elastic and magnetic properties is mutual, i.e., the magnetization changes produce magnetostrictive strains but external stresses can change magnetization. Free-standing micro-origami patterns have increased functionality. Bending of magnetostrictive layers can be used for stress, strain, or pressure sensing. Straining magnetostrictive layers in magnetoelectric composites is useful for tuning ferromagnetic resonance frequency in microwave filters by voltage applied to the piezoelectric layer. On the other hand, magnetostrictive strains generated in multiferroic composites by external fields can be converted into voltage and serve for the magnetic field sensing. Magnetoelectric performance of micro-origami structures is expected to be superior to that of flat patterns on substrates because of lack of the clamping effect. Giant magnetoresistive structures integrated with the microfluidic ducts in a single micro-origami assembly process are capable of detecting a single nanoparticle flowing through a duct.

The micro- and especially nano-origami architectures have great potential for biomedical applications. Majority of publications on magnetic origami concern objects with the radius of a few micrometers. Future applications of the micro-origami in biotechnology may require reduction of the size of magnetic structures to sub-micrometers or nanometers and combining them with organic materials. Also, the preference for these applications is for biocompatible

magnetic materials such as magnetite and biofriendly sacrificial layers. Initial results presented here look promising. More information about nano-origami techniques is available in the recent publication by Cavallo and Lagally [70].

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