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Abstract

The geometry of a magnetic nanoobject, namely its shape and dimension determines the complex electromagnetic responses. Here, we address the geometry-induced changes of the magnetic properties of thin ferromagnetic Co/Pd multilayers with out-of-plane magnetic anisotropy deposited on 3-dimensionally curved templates. For this pourpose, arrays of self-assembled cone-shaped nanoobjects with a chracteristic size of either 30 or 70 nm were created in GaSb(001) by the ion erosion technique. The templates are designed in the way that the shape of the cone remains the same for all the samples; namely, we keep the opening angle at about 55º by adjusting the ratio between the cone height and its base diameter to be about 1. In this case, we are able to address the impact of the linear dimensions of the object on the magnetic properties and exclude

the impact of the shape from the consideration. Deposition of 15-nm-thick Co/Pd multilayers on top of the cone templates results in the formation of a close-packed array of 2-dimensional magnetic cone-shaped shells. Integral angle-dependent magnetometry measurements demonstrate that local curvature results in the spread of the easy axes of magnetization following the shape of nanocones independent of the linear dimensions of the cone. At the same time different local magnetic domain patterns are observed for samples prepared on 30 and 70 nm large cones. When the thickness of the magnetic shell is only half of the linear dimension of a cone, a clear multidomain state is observed. Remarkably, we find that the neighboring magnetic cone-shaped shells are exchange decoupled when the linear dimension of a cone is 4 times larger compared to the thickness of the magnetic shell. These findings are relevant for the further development of tilted bit patterned magnetic recording media as well as for the emergent field of magnetism in curved geometries.

Introduction

In order to make progress in future advanced technologies based on magnetic materials – including magnetic data storage, logics and sensorics – the development of novel growth and fabrication techniques to obtain large area arrays of nanosized elements with controlled and reproducible magnetic, structural as well as topological properties is strongly required. To this pourpose, a great effort has been recently made to develop new cost- and time-efficient patterning methods, alternative to the conventional lithographic processes, to obtain ordered arrays of magnetic nanostructures on a large area[1][2]. In particular, significant progress has been achieved by self-assembling of nanoparticles by using nanoporous templates [3][4][5] or by depositing magnetic films on pre-structured templates. The latter can be made either of self-assembled densely packed nanostructures, e.g. obtained by using block copolymers [6][7][8], cylinders [9], spheres [10][11] [12][13][14][15] or regular patterns of nanostructures of ripples, nanocones or nanodots prepared by low energy ion beam sputtering of semiconductor and oxide surfaces[16][17][18][19]. Keeping in mind that the magnetic properties of thin films fabricated on patterned templates are strongly affected by the objects geometry, namely, shape, local curvature and dimension, the analysis of the geometry-induced modification of the magnetic responses gains in it importance in the modern magnetism research. Indeed, the deposition of magnetic thin films onto curved templates results in the formation of, e.g. quasi 1D nanowires or 2D curved nanoshells. Those objects attracted much attention in both theoretical and experimental research due to striking properties [20][21][22][23][24] originating from curvature-induced contributions to the magnetic exchange energy [25][26][27][28]. The progress in these directions strongly relies on the novel fabrication methods to realize 3D curved magnetic thin films as well as on the understanding of the specific electromagnetic responses of those objects. While there are already extensive experimental studies of magnetic properties of thin films prepared on nonmagnetic cylinder-shaped [9][16][18] or spherical particles [13] addressing static, dynamic [29] and topological properties [30][31] of the deposits, different shapes are not explored in the literature.

Here, we present a study on the geometry-induced changes in magnetic properties of out-of-plane magnetized Co/Pd multilayers deposited on top of large-area arrays of cone-shaped nanoobjects prepared by Ar^+ ion beam errosion in GaSb substrates. Ion beam irradiation was adjusted to fabricate two different pre-patterned GaSb substrates with the same shape of the objects (i.e. the same opening angle of a cone of about 55º, which is achieved by adjusting the ratio between the cone height and its base diameter to be about 1) but with different length scales as illustrated in Fig. 1. Both the composition and thickness of the Co/Pd stack were kept fixed in order to specifically adress the effect of the linear dimensions on the magnetization behaviour in cone-shaped magnetic nanoshells. The Co/Pd multilayers are a convenient model system as they are proven to be a robust system to achieve out-of-plane easy axis of magnetization virtually on any substrate [32][33][34] including 3-dimensional architectures [10][13][35][36]. In the present work, by fixing the shape of the nanoobject, we address the impact of the cone dimensions in the relationship with the film thickness on the static magnetic properties of the out-of-plane magnetized cone-shaped nanoshells.

Experimental

Densely packed cone-shaped nanoobjects were produced by normal incidence ion erosion of crystalline GaSb (001) surfaces using Ar^+ ions [37]. A Kaufman type ion gun with a single graphite extraction grid (5 cm diameter) was used. The irradiation was done at room temperature. The average distance among the cones corresponding to the base diameter of the cone, here defined as l_c (Fig. 1, c), is controlled by the kinetic energy of the incident Ar^+ ions [38]. Two different ion energies of 400 eV and 1200 eV were used to produce nanocone structures with l_c = 29(4) nm (referred to as *S* cones) and l_c = 75(7) nm (referred to as *L* cones), respectively (Fig. 1 a, b).

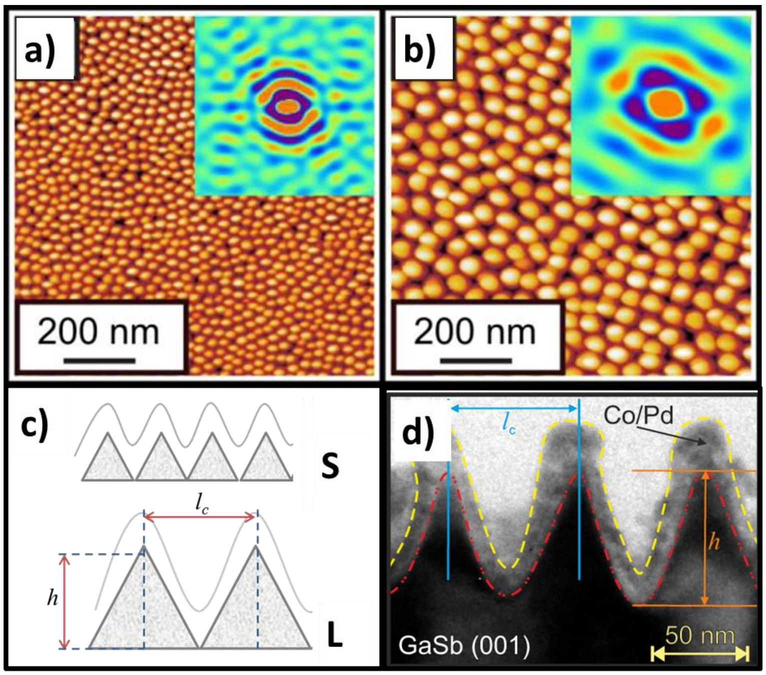

Figure 1. AFM images of uncoated GaSb nanocones for ion energies E of (a) 200 eV, (b) 1200 eV (inset: 2D autocorrelation function). (c) Sketch of pre-structured GaSb templates with different cone size (small – S – and large – L –) coated with a magnetic film represented by the curved line. (d) Cross-sectional transmission electron microscopy image of a Co/Pd multilayer deposited on top of L cones. The dashed lines depict interfaces for clarity. Red dashed line highlights the morphology of the template in a GaSb substrate. Yellow dashed line indicates the top surface of the deposited magnetic film.

The cone height *h* increases with the enhancement of the fluence [39]. Hence, to prepare cone-shaped nanoobjects of different dimentions but with the same opening at the apex, the ion fluence was adjusted. Therefore, the *S* cones with an averaged height of 30(3) nm were prepared with a fluence of $1.0\text{x}10^{18}$ Ar^+/cm^2 while *L* cones with *h* = 67(8) nm were prepared with a fluence of $1.5 \cdot 10^{17}$ Ar^+/cm^2. As evidenced in table 1, the h/l_c aspect ratio is almost the same, being 1.0 and 0.9 for S and L cones, respectively.

Sample ID	**E (eV)**	**Fluence (Ar^+cm^{-2})**	**l_c (nm)**	**h (nm)**	**h/l_c**
S	400	$1.0 \cdot 10^{18}$	29(4)	30(3)	1
L	1200	$1.5 \cdot 10^{17}$	75(7)	67(8)	0.9

Table 1: Ion irradiation parameters and resulting dimensions of the cone-shaped nanoobjects.

Onto these patterned substrates, Co/Pd multilayer films were sputter deposited at room temperature. The magnetic stack is Pd(3nm)/[Co(0.3 nm)/Pd (0.9nm)]$_8$/Pd(2 nm)/Subst., with a total nominal thickness of about 15 nm. The deposition rate was 0.04 nm/s for Co and 0.02 nm/s for Pd under a partial Ar pressure of $3.5 \cdot 10^{-3}$ mbar. The cross-sectional transmission electron microscopy image (TEM) of the film deposited on the *L* cones is shown in Fig. 1d. The flux during the film depositon is perpendicular to the substrate, which results in the nominal thickness of the film on a cone's apex, which is reduced about twice for the deposit on the sidewalls. [40].

The magnetic properties were studied at room temperature using a Vector Vibrating Sample Magnetometer (VSM model 10 – MicroSense), equipped with a rotating electromagnet. The direction of magnetization with respect to the applied magnetic field was determined by measuring a series of hysteresis loops with the external magnetic field applied along different polar angles θ_H ranging from -90° to +90°, where θ_H = 0° defines the film's normal. In-plane angle-dependent measurements were also performed but are not shown, as they exhibit isotropic behavior in agreement with the structural in-plane isotropy of the cone template.

The magnetic force microscopy (MFM) measurements were carried out using a Nanoscope Multimode Scanning Probe Microscope (Digital Instruments) with MFM tips from Team Nanotec.

Results and discussion

In Fig. 3, the in-plane and out-of-plane hysteresis loops are presented for Co/Pd films deposited on *S* and *L* cones and compared with those of a continuous reference film deposited on a flat substrate. A squared loop is observed for the reference sample when the field is applied perpendicular to the surface ($\theta_H = 0°$), as typically expected for Co/Pd multilayers. A dominant perpendicular magnetic anisotropy is mainted when a pre-structured substrate is used. Still, the coercivity extracted from the loops measured at $\theta_H = 0°$ is reduced from 0.70 kOe (planar) to 0.41 kOe (*S* cones) down to 0.28 kOe (*L* cones).

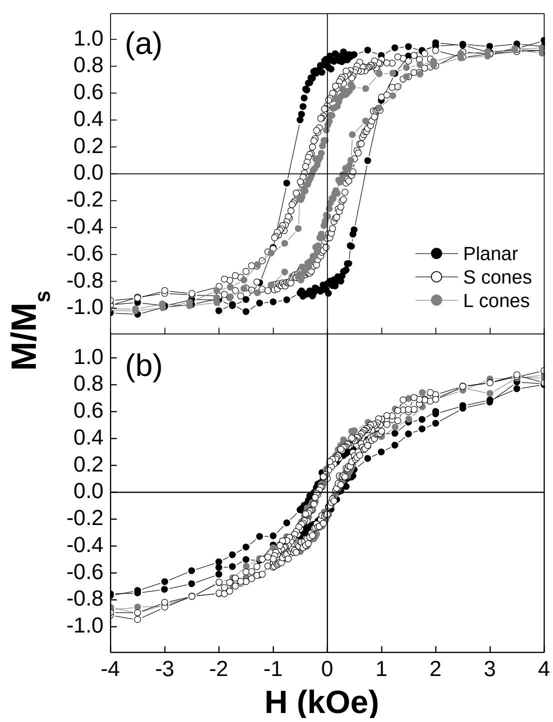

Figure 3. Normalized hysteresis loops measured at room temperature for Co/Pd on *S* and *L* cones and on a planar substrate (reference sample). (a) $\theta_H = 0°$ (out-of-plane) and (b) $\theta_H = 90°$ (in-plane).

Such a decrease is consistent with the tilt of the easy axes of magnetization following the cones's shape. Such a behaviour is confirmed by polar angle-dependent measurements of the coercivity H_c (Fig. 4a) and normalized magnetization remanence M_r/M_s (Fig. 4b). Present findings taken of cone-shaped magnetic shells are consistent with the behaviour observed in magnetically capped spherical particles [40]. The polar curves show a maximum at θ_H = 0° for all three samples, indicating the preferential perpendicular magnetic anisotropy. Overall, both M_r/M_s and H_c decrease when moving from a flat to the patterned substrates, due to the tilting of the easy axis induced by the cone's shape. Moreover, the M_r/M_s angular dependence broadens with increasing cone size suggesting that such a local spread of the easy axis of magnetization enhances with increasing the cones' size.

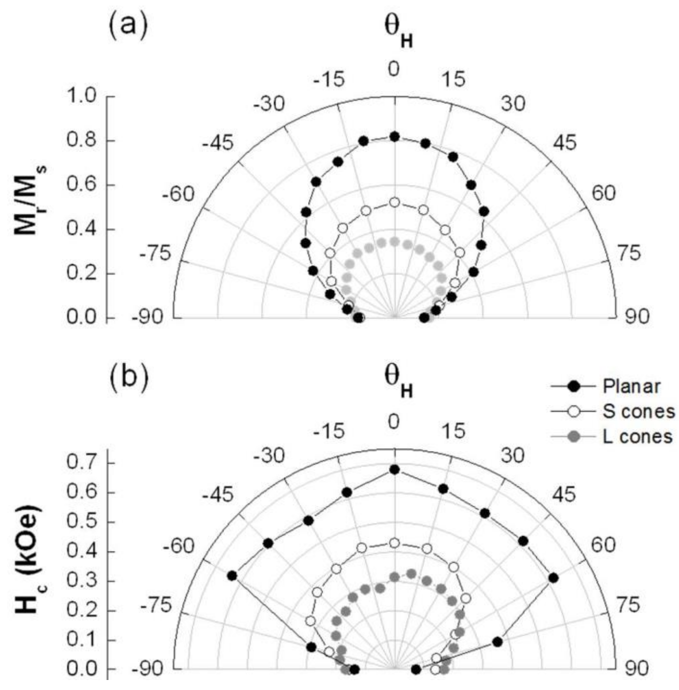

Figure 4. Room temperature angular dependence of coercivity H_c (a) and normalized remanence (M_r/M_s) of Co/Pd samples with three different morphologies.

In order to elucidate the effect of the template size on the M_r/M_s values, the remanent state was investigated using MFM at room temperature. MFM images taken after ac-demagnetization of the

Co/Pd multilayers on the reference sample, *S* and *L* cones are presented in Fig. 5. The reference sample on planar GaSb substrate was found to be in a multidomain state with a domain size of about 200 nm (Figs. 5(a, d)), which is typical for Co/Pd multilayer samples [41]. The sample with smaller cones (Figs. 5(b, e)) is in a multidomain magnetic state, where the domain size is in the range of 115(15) nm, the position of the magnetic domain walls being correlated with the locations of the cones. For the film on *L* cones (Figs. 5(c, d)), a clear dark and bright MFM contrast is observed on the cone locations, indicating that individual magnetic structures are in a single-domain state. Furthermore, MFM image suggests that neighboring cap structures are more magnetically exchange decoupled. The decoupling mechanism is expected to be similar to the one observed for magnetic caps on arrays of spherical particles [40] where the Co/Pd thickness varies across the particle surface resulting in a transition from ferromagnetic (at the top) to paramagnetic closer to the base of the cap/cone [42].

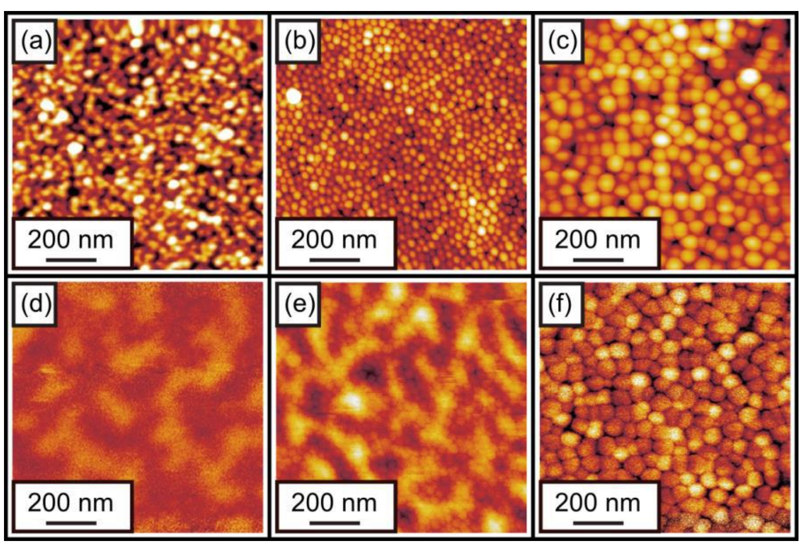

Figure 5. AFM and MFM images of the Co/Pd multilayers on a planar substrate (a, d), *S* cones (b, e) and *L* cones (c, f)

According to the results obtained by the MFM analysis, the difference in the remament state between the two systems would suggest a significant difference in the domain configuration and, consequently, in the magnetization reversal process, which has been demonstrated to be strongly altered by the substrate curvature, following unusual mechanisms that cannot be univocally described with the cassical theories (Stoner-Wolhfart or Kondorsky-like for single domain uniaxial nanostructures or multi-domain films, respectively) [40]. Actually, although both samples are characterized by the same cones shape (i.e. the same tilting of the lateral surface), when the Co/Pd multilayer is deposited on S cones, its reduced thickness, whose value is only half of the cone's height (see figure 1), does not allow a complete decoupling between the Co/Pd caps, leading to a continuous film whose magnetic configuration resembles that of the reference film (deposited on a flat substrate) but with a significant reduction of the out of plane coercivity. On the other hand, L cones (where h >> film thickness) induce a significant decoupling between Co/Pd islands deposited on the cone's apex, according to what has already been observed in Co/Pd multilayers with variable bilayer number n deposited onto polystyrene nanoparticles, where the decoupling takes place for small n, i.e. when the total film thickness is smaller than the particle diameter [42].

Conclusions

In this work the changes of the magnetic properties of Co/Pd multilayers with perpendicular magnetic anisotropy on templates of ion-eroded self-assembled GaSb nanocones with $h/l_c \approx 1$ were investigated. The integral magnetic properties characterized by vector VSM magnetometry indicate that all samples possess the overall magnetic easy axis perpendicular to the substrate's surface. The remanent magentization as well as the coercive field decrease with increasing the lateral size of the cone due to a larger extension of the cone's sides, which accordingly induces a wider tilt of the easy axis of magnetization. However, only if the pattern modulation (i.e. the heigh of the cones) is much higher than the film thickness, the template is able to signficantly affect the film domain structure, leading to a patterned system where the magnetic elements are exchange decoupled, as in the case of *L* cones. Indeed, in the Co/Pd system deposited on *S* cones, being the thickness of the magnetic layer comparable with the heigh of the pattern elements, the continuous magnetic film does not break into isolated uncoupled elements but retains the perpendicular anisotropy although the coercivity is reduced by the tilting of the easy axis of magnetization induced by the underlying template. In other words, the fixed Co/Pd thickness and its relashionship with the cones' heigh is expected to bring to different coverage morphology and, ultimately, to a different magnetic behaviour. In particular, a significant modulation of the exchange interactions was observed, with a net evolution from a multidomain system to uncoupled magnetic regions located at the cone's apex. The present study demonstrated the feasibility of using GaSb templates as a new time-efficient way to get patterned magnetic systems on large areas, by a proper choice of the geometrical relationship between film thickness and cone's height. The possibility to simultaneusly modulate the easy axis tilting [43][44] and the nanostructures exchange coupling by properly choose the template geometry has a great impact in the field of the bit patterned media (BPM) technology [45]. Indeed, it has been recently demonstrated an improvement in the recording density of bit patterned media with inclined anisotropy axis and exchange coupling among dots, both the effects concurring in reducing the magnetostatic interactions among bits [46][47][48]. Here, the use of a GaSb nanocones as a templated substrates for the Co/Pd growth allowed to induce a tilting of the easy axis, which can be suitably varied according to the template curvature, as succesfully demonstrated in the case of depositions on nanosphere substrates [10]. The possibility to further control the magnetic behaviour by affecting the magnetic domain isolation paves the way for a new class of magnetic systems where the magnetic properties are suitably tailored to match technological requirements. Additional efforts on the production of self-organized nanocones with a higher spatial order are expected to fully unfold their potential for technological applications especially in the field of magnetic data storage. The presented results are also relevant beyond the field of magnetic data storage. We envision that the deposition of magnetic thin films on templates prepared using ion beam erosion might open new pathways for the experimental realization of relevant samples for the emergent research field of magnetism in curved geometries [24][25][36], where the shape as well as the curvature of magnetic thin films play a crucial role in the determining their electromagnetic [49][50] and topological properties[51] [28].

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