Periodic arrays of magnetic nanostructures by depositing Co/Pt multilayers on the barrier layer of ordered anodic alumina templates

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We developed a method to fabricate ordered Co/Pt multilayer nanodot arrays using the barrier layer of anodic alumina templates as two-dimensional curved substrates. Large area patterning of self-assembled alumina nanobumps are formed with hexagonally close-packed order. The subsequent deposition of Co/Pt multilayers on this bumpy surface leads to an ordered array of single-domain nanocaps magnetized perpendicularly to the sample surface. Measurement of reversal field as a function of field angle and images of demagnetized state, composed with monobump-wide stripe domains, confirm the weakness of exchange coupling between bits. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4731640]

Ordered arrays of isolated magnetic nanostructures are of considerable interest to increase the storage density of hard disks beyond the current perpendicular media. In such bit patterned media, each artificially fabricated magnetic nanostructure is capable of storing an individual bit.[1] Fabrication schemes involve either lithographic patterning[2,3] or combination of e-beam lithography methods with other techniques such as electrodeposition[4] and self-assembling block copolymers.[5] However, lithographic techniques are expensive and time-consuming. Therefore, it is of interest to *develop* a non-lithographic patterning method based on self-assembly as an alternative route in order to develop a fast and cheap process capable of producing dense arrays of nanostructures over millimeter wide areas with precise long-range order.

Recently, a simple and inexpensive method of fabricating nanoscale magnetic patterns was developed by using self-assembly of *colloidal nanospheres*.[6] Self-assembled close-packed layer of polystyrene nanospheres of diameter in the range of 50–300 nm were used as curved substrate to deposit Co/Pt and Co/Pd multilayers and to create ordered arrays of single domain and magnetic exchange decoupled nanostructures.[6,7] However, several problems associated with the nanospheres approach were pointed out due to the presence of dislocations and domains, and also the cleanness and hydrophilic properties of the substrate surface are critical in making well-ordered colloidal crystal films.[8] In addition, attractive forces between the spheres prevent the formation of a layer of spheres with good order when considering diameters below 50 nm so that only short-range order can be obtained, or their position need to be templated using lithography to impose long-range order.[9]

In this letter we present another route to fabricate at low cost a densely packed magnetic particle assembly on two-dimensional (2D) curved substrates. The method involves the growth of magnetic nanocaps on the back of nanoporous anodic alumina templates (AAO), which is a typical self-ordered nanopore-array material formed by the electrochemical oxidation of Al in acidic solutions.[10,11] In recent years, there have been a considerable number of studies regarding the use of AAO templates for the fabrication of nanostructured arrays with perpendicular anisotropy by depositing Co/Pt multilayers and CoPt binary alloys. Previous works have reported on the formation of CoPt nanowires embedded in nanoporous AAO templates by electrodeposition. The $L1_0$ CoPt ordered phase was obtained by electrodeposition of either CoPt alloy with near-equiatomic composition[12,13] or Co/Pt multilayered nanowires[14] followed by the appropriate annealing process. On the other hand, Co/Pt perpendicular antidot arrays were obtained by depositing on porous AAO templates.[15] This leads to a percolated perpendicular media where the holes provide a pinning effect. However, until now there has been no report of the use of the bumpy surface formed by the barrier layer of the AAO template to make such ordered perpendicular nanostructured media. In this work, we studied a hexagonally close-packed array of $[Co/Pt]_4$ nanocaps deposited on bumps having about 100 nm lateral modulation period and 50 nm height variation. We show that $[Co/Pt]_4$ magnetic multilayers form an array of magnetic single-domain bits with magnetization pointing perpendicular to the substrate surface. Demagnetized states confirm the weakness of the exchange coupling between bits and allow correlating structural and magnetic disorders.

The voids of the nanoscale-channeled structure of AAO nanotemplates have been widely used in the past to form nanowires and multilayers in filling the pores by electrochemical deposition methods.[16–18] Electrodeposition is usually performed after the removal of the oxide barrier layer that closes the bottom ends of the pores. Therefore this barrier layer is typically considered undesirable. However, herein we emphasize that the surface of this layer possesses an ordered array of bumps that can also be used as a pre-patterned substrate to modulate the thickness of thin Co/Pt

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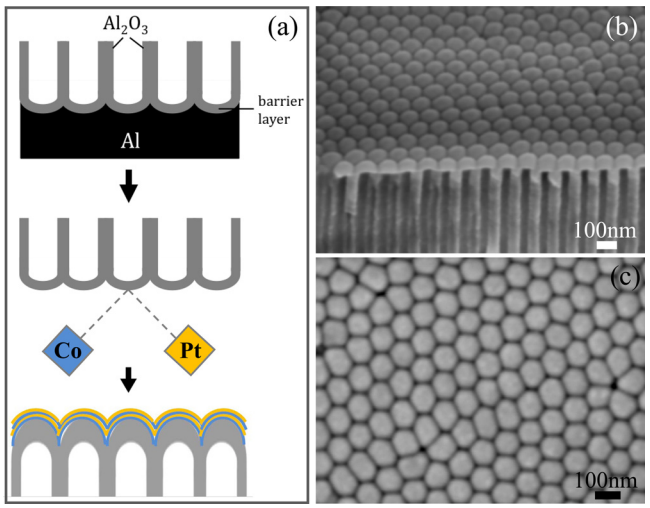

FIG. 1. (a) Schematic representation of the formation of 2D-ordered bumps array from the barrier-layer surface of the AAO template after a two-step anodization process, followed by sputtering deposition. On the right, SEM images of the tilted cross section of the AAO template (b) and its curved surface (c).

multilayers deposited on it, as sketched in Fig. 1(a). The AAO templates were fabricated by a two-step anodization process in order to obtain highly ordered structures.[10,11] The templates with 105 nm average pore spacing consist of long channels ($\sim$50 μm) closed at the bottom end by a round-shaped Al_2O_3 barrier layer [Fig. 1(b)]. The barrier-layer surface is composed of a hexagonal close-packed lattice of nanobumps that possess the same spacing as the porous layer [Fig. 1(c)]. We used the method described in Ref. 19 to analyze the SEM images and characterize the degree of ordering in the barrier-layer surface. According to this triangulation method, the coordinates of the centers of the repetitive bumps are first obtained, serving for the ordering analysis. Then, the triangles defined by coordinates of the centers are identified by Delaunay tessellation.[20] To recognize the most ordered regions, we used a criterion of discrimination among all triangles based on 15% maximum deviation of side lengths relative difference [Fig. 2(a)]. The histogram in Fig. 2(b) represents the triangle side length distribution and can be fitted by a Gaussian distribution. From this result, we calculated a mean inter-bump distance of 105 nm and a full width at half maximum (FWHM) of 15.5 nm.

Co/Pt multilayer film was coated onto the bumpy substrate by magnetron-sputtering deposition. The multilayer consisted in a Ta(5 nm)/Pt(5 nm)/[Co(0.36 nm)/Pt(0.73 nm)]$_4$ and was covered by an additional 3.5 nm thick Pt layer to prevent oxidation. The total thickness of the magnetic stack ($\sim$10 nm) was voluntarily kept lower than the bump height ($\sim$50 nm) and period ($\sim$100 nm). Indeed in that case, the magnetic coating is expected to form an ordered array of magnetic caps with low exchange coupling, with magnetization pointing perpendicular to the substrate surface.[6,7] The same multilayer was deposited on a flat Si/SiO_x substrate (the so-called reference sample) for comparison. The samples were magnetically characterized using a commercial Vibrating Sample Magnetometer (VSM) and a Magnetic Force Microscope (MFM).

Figure 3(a) compares the room temperature magnetization hysteresis curves of the Co/Pt multilayers grown on the bumpy substrate with that of the reference sample. The external magnetic field was applied perpendicular to the substrate, i.e., along the anisotropy axis. The magnetization curve for the Co/Pt multilayer grown on the bumpy sample gets tilted and has larger coercive field as compared with the reference one. These differences in the hysteresis curve mark the exchange decoupling between neighboring Co/Pt nanocaps and the increased influence of the dipolar interactions. The increase of coercivity is explained by the change of magnetization reversal mechanism. In the case of the reference sample, nucleation occurs in a region with lower-than-averaged anisotropy and the full reversal is achieved by domain wall propagation. In the bumpy sample, each nanocap magnetization reverses by a more uniform and more independent process. This change in reversal process reflects in the reversal branch shearing. The reversal field distribution mostly originates from dipolar interaction between caps that tends to maintain anti-parallel neighboring caps,[21,22] as well as the intrinsic cap-to-cap anisotropy distribution. Note that, at the coercive point, when half the dots have switched, in average, each dot has as many up as down magnetized neighbors, and the dipolar interactions are negligible. Figure 3(b) shows that the relative reversal field in the reference sample follows the variation calculated for the Kondorsky-like behavior (i.e., domain wall propagation driven reversal).[23] On the contrary, the relative coercivity ratio for the bumpy samples has a minimum when the field is applied at a 45° angle similar to Stoner–Wohlfarth type reversal.[24] The

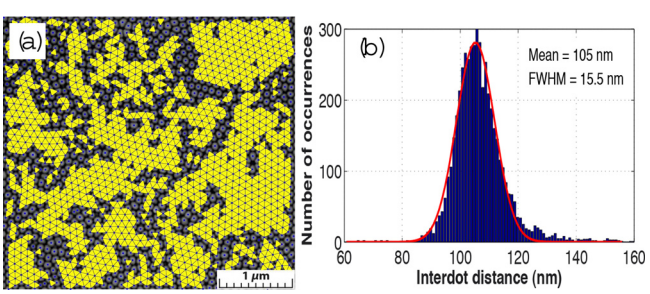

FIG. 2. (a) SEM image of the barrier-layer surface of the AAO template and results of ordering analysis showing ordered regions (in yellow) of equilateral triangles between centers of the bumps (within 15% deviation of side length). (b) Histogram representation of the inter-dot distances.

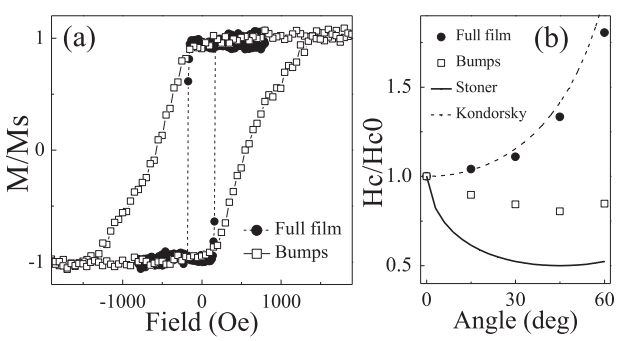

FIG. 3. (a) Hysteresis loops at 300 K, for Co/Pt multilayers with the field applied perpendicular to the bumpy (open squares) and flat (solid circles) substrates; (b) relative coercivity as a function of the angle of the field with respect to the anisotropy axis for the bumpy (open squares) and full film (solid circles) samples. Hc0 corresponds to the coercive field measured with field parallel to the easy axis. The dashed and solid lines in (b) correspond to Kondorsky and Stoner–Wohlfarth models, respectively.

fact that Hc/Hc0 is larger than 0.5 at 45° can be explained by the dispersion of anisotropy axis over the nanocap[6] and by a partially incoherent reversal since the studied nanocaps have diameters of 100 nm.

Besides, the reversal branch tilting in the bumpy sample comes from switching field distribution which originates from the dipolar field interactions between nanocaps and from nanocap intrinsic magnetic properties distribution.[1] In order to estimate the relative influence of both, we performed an AC demagnetization process in a perpendicular field and then imaged the magnetic configuration of the nanocaps using MFM. Note that the AC demagnetization process leads to no stable domain in the film deposited on smooth Si/SiO_x substrate (not shown here). Indeed due to the low number of Co/Pt repeats and the strong lateral magnetic coupling in the full film, the exchange interactions dominate the dipolar ones.[25] This indicates a very low defect density in the reference sample.

Figures 4(a) and 4(b) shows a typical AFM and MFM image of the out-of-plane AC demagnetized bumpy sample. In Fig. 4(b) a bicolor pattern corresponding to magnetic domains having opposite magnetization directions is observed in the MFM image. The bright and dark domains correspond to a magnetization pointing towards the negative and positive field directions, respectively. Contrary to well-known isotropic labyrinth patterns observed for out-of-plane AC demagnetized Co/Pt film,[25] the MFM image in Fig. 4(b) is very asymmetric. The contour plot corresponding to the topology extracted from Fig. 4(a) has been superimposed to the MFM image. The width of the observed magnetic stripes corresponds to the diameter of a bump. To further characterize the demagnetized state we calculated the 2D Fourier transform associated to the magnetic structure on large scale as shown in Fig. 4(c). The peaks positions confirm that domain width is about 100 nm, i.e., the diameter of a bump. Besides, the spatial extension of theses peaks suggest a dispersion of about 30° of the magnetic stripes direction.

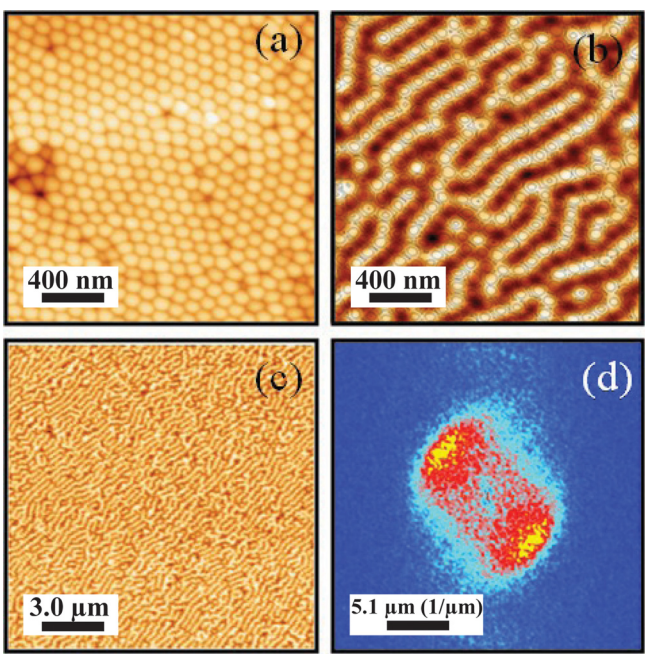

FIG. 4. (a) and (b) correspond to $2 \times 2\ \mu m^2$ AFM and MFM images respectively, obtained after AC demagnetizing the bumpy sample. The contour plot corresponding to the topology in (a) has been superimposed to the MFM image; (c), (d) $10 \times 10\ \mu m^2$ MFM image of the bumpy sample and the associated 2D Fourier transform, respectively.

Let us discuss the origin of the monodots-wide lines AC demagnetized state. The AFM image shows the hexagonal array of dots with randomly spread lattice defects [Fig. 4(c)]. If the exchange-coupling between dots is null, domain pattern is formed to minimize the interdot dipolar field energy, i.e., to maintain neighboring bits with anti-parallel magnetization.[26] In a square lattice, the domain state of minimum energy is a checkboard phase. In a hexagonal array, triangular frustration occurs and lines of aligned magnetization minimize the energy. Finally, the finite length of the magnetic stripes seems to be correlated to the large scale arrangement of the bumps into grains separated by grain boundaries and the dispersion in the magnetic stripes direction to the dot-to-dot arrangement inside the grains.

As a conclusion, we have reported a method to fabricate ordered magnetic nanobumps arrays using the barrier layer of AAO templates. Our self-assembly method has a number of advantages over the lithographic based methods, including low-cost and extremely simple processing. The AAO nanotemplates display unique features, such as easily tunable geometrical parameters, mild preparation conditions, robustness, and resistance to high temperatures. In addition, the extremely small attainable inter-bump period constitutes substantial advantages. Indeed regular nanopore arrays in AAO with ultra small pore diameter (even less than 10 nm) and pore interdistance as small as 15 nm have already been demonstrated.[27,28] We have tested the feasibility of bit pattern media on the nanobumps with regular Co/Pt multilayer. We have shown that the magnetic cap nanostructures are exchange decoupled and therefore can be addressed separately.

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[1]O. Ozatay, P. G. Mather, J. U. Thiele, T. Hauet, and P. M. Braganca, in *Nanofabrication and Devices, Comprehensive Nanoscience and Nanotechnology*, edited by D. Andrews, G. Scholes, and G. Wiederrecht (Elsevier, London, 2010), Vol. 4, pp. 561–614.
[2]S. Y. Chou, Proc. IEEE **85**, 652 (1997).
[3]F. Luo, L. J. Heyderman, H. H. Solak, T. Thomson, and M. E. Best, Appl. Phys. Lett. **92**, 102505 (2008).
[4]C. A. Ross, H. I. Smith, T. A. Savas, M. Schattenberg, M. Farhoud, M. Hwang, M. Walsh, M. C. Abraham, and R. J. Ram, J. Vac. Sci. Technol. B **17**, 3168 (1999).
[5]O. Hellwig, J. K. Bosworth, E. Dobisz, D. Kercher, T. Hauet, G. Zeltzer, J. D. Risner-Jamtgaard, D. Yaney, and R. Ruiz, Appl. Phys. Lett. **96**, 052511 (2010).
[6]M. Albrecht, G. Hu, I. L. Guhr, T. C. Ulbrich, J. Boneberg, P. Leiderer, and G. Schatz, Nat. Mater. **4**, 203 (2005).
[7]T. C. Ulbrich, C. Bran, D. Makarov, O. Hellwig, J. D. Risner-Jamtgaard, D. Yaney, H. Rohrmann, V. Neu, and M. Albrecht, Phys. Rev. B **81**, 054421 (2010).
[8]L. Yan, K. Wang, J. Wu, and L. Ye, Colloids Surf., A **296**, 123 (2007).

[9]P. Kappenberger, F. Luo, L. J. Heyderman, H. H. Solak, C. Padeste, C. Brombacher, D. Makarov, T. V. Ashworth, L. Philippe, H. J. Hug, and M. Albrecht, Appl. Phys. Lett. **95**, 023116 (2009).
[10]H. Masuda and K. Fukuda, Science **268**, 1466 (1995).
[11]O. Rabin, P. R. Hertz, Y.-M. Lin, A. I. Akinwande, S. B. Cronin, and M. S. Dresselhaus, Adv. Funct. Mater. **13**, 631 (2003).
[12]F. M. Rhen, E. Backen, and J. M. D. Coey, J. Appl. Phys. **97**, 113908 (2005).
[13]J. Mallet, K. Yu-Zhang, S. Mátéfi-Tempfli, M. Mátéfi-Tempfli, and L. Piraux, J. Phys. D: Appl. Phys. **38**, 205002 (2005).
[14]L. F. Liu, S. S. Xie, and W. Y. Zhou, J. Phys. D: Appl. Phys. **42**, 205002 (2009).
[15]M. T. Rahman, N. N. Shams, Y. C. Wu, C. H. Lai, and D. Suess, Appl. Phys. Lett. **91**, 132505 (2007).
[16]C. R. Martin, Chem. Mater. **8**, 1739 (1996).
[17]D. J. Sellmyer, M. Zheng, and R. Skomski, J. Phys.: Condens. Matter **13**, R433 (2001).
[18]M. Hernández-Vélez, Thin Solid Films **495**, 51 (2006).
[19]S. Mátéfi-Tempfli, M. Mátéfi-Tempfli, and L. Piraux, Thin Solid Films **516**, 3735 (2008).
[20]B. Delaunay, Izv. Akad. Nauk SSSR, Otdelenie-Matematicheskih i Estestvennyh Nauk **7**, 793 (1934).
[21]B. Pfau, C. M. Gunther, E. Guehrs, T. Hauet, H. Yang, L. Vinh, X. Xu, D. Yaney, R. Rick, S. Eisebitt, and O. Hellwig, Appl. Phys. Lett. **99**, 062502 (2011).
[22]O. Hellwig, A. Berger, T. Thomson, E. Dobisz, Z. Z. Bandic, H. Yang, D. S. Kercher, and E. E. Fullerton, Appl. Phys. Lett. **90**, 162516 (2007).
[23]E. Kondorsky, J. Phys. (Moscow) **2**, 161 (1940).
[24]E. C. Stoner and E. P. Wohlfarth, Philos. Trans. R. Soc. London A **240**, 599 (1948).
[25]O. Hellwig, A. Berger, J. B. Kortright, and E. Fullerton, J. Magn. Magn. Mater. **319**, 13 (2007).
[26]K. Nielsch, R. B. Wehrspohn, J. Barthel, J. Kirschner, S. F. Fischer, H. Kronmuller, T. Schweinbock, D. Weiss, and U. Gosele, J. Magn. Magn. Mater. **249**, 234 (2002).
[27]Y. Matsui, K. Nishio, and H. Masuda, Small **2**, 522 (2006).
[28]G. Q. Ding, R. Yang, J. N. Ding, N. Y. Yuan, and Y. Y. Zhu, Nanoscale Res. Lett. **5**, 1257 (2010).