

Shape Induced Anisotropy in Anti-Dot Arrays from Self-Assembled Templates

A. A. Zhukov, M. E. Kiziroglou, A. V. Goncharov, R. Boardman, M. A. Ghanem, M. Abdelsalam, V. Novosad, G. Karapetrov, X. Li, H. Fangohr, C. H. de Groot, P. N. Bartlett, P. A. J. de Groot

Abstract—Using self-assembly of polystyrene spheres well ordered templates have been prepared on glass and silicon substrates. Strong guiding of self-assembly is obtained on photolithographically structured silicon substrates. Magnetic anti-dot arrays with 3D architecture have been prepared by electrodeposition in the pores of these templates. The shape anisotropy demonstrates crucial impact on magnetization reversal processes.

Index Terms—Electrodeposition, magnetic hysteresis, nanotechnology, silicon, self-assembly.

I. INTRODUCTION

THE demand for higher magnetic recording densities is stimulating intense research activity into magnetism on sub-micrometer length scales. The main requirement for commercially viable products is an efficient and low-cost preparation process. Methods based on templates formed by the self-assembly of colloidal particles have been considered for various applications such as photonic materials [1], microchip reactors [2] and biosensors [3]. In this work we use ordered templates for the electrodeposition of various magnetic materials. This technique offers new opportunities, which are not easily realized by standard lithographic methods, and which allow us to create magnetic nanostructures with 3D architectures on a broad range of length scale, 20 – 1000 nm [4]. To address limitations of self-assembly (lack of long-range order and control of array directions) we are employing hybrid techniques using well-established Si technology to guide and confine self-assembly templates (e.g., [5]).

Manuscript received February 7, 2005. This work was supported in part by the U.K. Engineering and Physical Sciences Research Council grants GR/S13200/01 and EP/C511786/1.

A. A. Zhukov, A. V. Goncharov and P. A. J. de Groot are with the School of Physics and Astronomy, University of Southampton, SO17 1BJ UK (e-mail: aaz@phys.soton.ac.uk; avg@phys.soton.ac.uk; pajdeg@phys.soton.ac.uk)

M. E. Kiziroglou, X. Li and C. H. de Groot are with the School of Electronics and Computer Science, University of Southampton, SO17 1BJ UK (e-mail: mk03r@ecs.soton.ac.uk; xl04r@ecs.soton.ac.uk; chdg@ecs.soton.ac.uk)

R. Boardman and H. Fangohr are with the School of Engineering Sciences, University of Southampton, SO17 1BJ UK (e-mail: R.P.BOARDMAN@soton.ac.uk; H.FANGOHR@soton.ac.uk)

M. A. Ghanem, M. Abdelsalam and P. N. Bartlett are with the School of Chemistry, University of Southampton, SO17 1BJ UK (e-mail: m.a.ghanem@soton.ac.uk; m.abdelsalam@soton.ac.uk; p.n.bartlett@soton.ac.uk)

V. Novosad and G. Karapetrov are with the Materials Science Division, Argonne National Laboratory, 9700 South Cass Ave., Argonne, IL60439, USA (e-mail: novosad@anl.gov; goran@anl.gov)

II. EXPERIMENTAL PROCEDURE

Using slow evaporation of a colloidal water suspension containing 1wt. % of latex spheres, well-ordered templates have been self-assembled on glass substrates with buffer layers of sputtered Cr (10 nm) and Au (200 nm) or on HF-treated Si. These templates have been used as moulds to prepare nanoporous magnetic (Ni, Co, Fe and Ni₅₀Fe₅₀) structures by electrodeposition methods [6].

Magnetic measurements were performed using a SQUID, a vibrating sample magnetometer and a magneto-optical Kerr effect (MOKE) rig. Most measurements have been performed at room temperature with the magnetic field applied parallel to the plane of the film. Magnetic structures have been imaged using magnetic force microscopy (MFM) by a Digital Instruments 3000 scanning probe microscope in lift-mode with a fly height of 100 nm and a standard low-moment ferromagnetic tip.

III. RESULTS AND DISCUSSION

A. Guided Self-Assembly and Electrodeposition on structured Silicon

To study confinement effects and to guide ordering in defined directions we have used a scaffolding method for guided self-assembly by various lithographically prepared patterns on the surface of a Si substrate. For this purpose a 250 nm thermal SiO₂ layer was grown on the Si wafers and different patterns (stripes, squares, triangles, circles and rings) were defined by photolithography. Subsequently, the wafers were immersed in a 20:1 buffered HF solution to etch the oxide and to expose Si areas of different shapes and sizes ranging from 1.5 μm to 50 μm. Then, the wafers were sawed in pieces, RCA (Radio Corporation of America) process cleaned [7], and dipped in 20:1 buffered HF prior to latex sphere self-assembly. After the self-assembly the wafers with sphere templates were dipped again in 20:1 buffered HF. The spheres adhesion presumably governed van der Waals forces [8] is sufficient to withstand this process. As we can see from the Fig. 1 the template is formed on the structured areas of Si and essentially no spheres can be observed in the region of SiO₂ (dark gray surface). The template structure demonstrates well-pronounced guiding with the side of a triangular unit cell parallel to the stripe direction. Next, Ni films were grown on the conducting Si areas by electrodeposition. To improve the smoothness pulsed electrochemical pretreatment was used. Finally, the spheres were dissolved in tetrahydrofuran.

FIG. 1 HERE

The resulting magnetic anti-dot array is the inverse of the template and also demonstrates well-guided behavior in the case of stripes (Fig. 2). For square Si/SiO₂ microstructures the self-assembly was guided along one of the square sides. No guiding effect was observed for right angle triangles, circles and rings. This probably results from competition between different directions favoured by their shape.

FIG. 2 HERE**B. Magnetization Reversal and Shape Anisotropy**

The template nanostructuring strongly influences the magnetic properties (e.g. [9]). The 3D architecture results in behavior resembling that of multilayers with alternating hard and soft layers: the coercive field B_c has been found to oscillate with the thickness of the film t_f (Fig. 3). B_c reaches a maximum for the case when the top surface of the film is near the void centres. For layers with complete spherical voids the coercive field approaches a minimum. These observations suggest that the points where the spheres touch play an important role in domain wall pinning and hence the coercivity [10]. Our results demonstrate that 3D patterned magnetic materials are prototypes of a new class of *geometrical multilayer structures* in which the layering is due to local shape effects rather than compositional differences.

FIG. 3 HERE

To investigate the local magnetic structures of the templated films, we employed MFM complemented by micromagnetic modeling. Results are shown in Fig. 4a for a relatively thin (100 nm) Co film with sphere diameter $d = 700$ nm in the remanent state. These images reveal ordered, rhombic magnetic patterns associated with the anti-dot array. Magnetic structures for 2D anti-dot arrays were calculated using micromagnetic modeling with the OOMMF software suite [11]. Parameters in the computation were chosen in accordance with the known values for Co [12], cell size was 2nm, and hole diameters of $0.4 d$ correspond to the top of the film. The MFM signal was modeled by computing the second derivative of the dipolar field produced by the film [13] at a distance of the fly height above the film (Fig. 4b). The picture obtained from 2D micromagnetic simulations is similar to the results of MFM.

FIG. 4 HERE

We have measured angular dependences of coercive field using MOKE to reveal the influence of shape anisotropy produced by nanostructuring (Fig.5). The obtained results reveal pronounced six-fold symmetry corresponding to the hexagonal configuration of the nanostructured antidot array. The small distortion along 0° direction is probably caused by

film texturing. The micromagnetic simulations [7] qualitatively reproduce the experimental observations with sharp maxima along the bysectors of unit cell triangles.

FIG. 5 HERE

IV. CONCLUSIONS

Using self-assembly from colloidal suspensions of polystyrene latex spheres, we have prepared well-ordered, nanostructured templates. We present results of electrochemical deposition of magnetic metals and alloys in the voids of these templates, enabling the fabrication of magnetic anti-dot nanostructures with 3D architectures. We find large changes in the coercive field, dominated by the effects of the nanoscale shapes. These 3D anti-dot nanostructures reveal oscillations of the coercive field in the dependence on film thickness. Varying the parameters in the preparation (sphere diameter, film thickness and composition) allows us to produce materials with predetermined magnetic parameters. To address limitations of self-assembly (lack of long-range order and control of array directions) we are employing hybrid techniques using well-established Si technology to guide and confine self-assembly templates. The template growth technique has the potential to lead to a low-cost preparation method for sub-micron patterned magnetic materials.

REFERENCES

- [1] Y. A. Vlasov, X. - Z. Bo, J. C. Sturm, D. J. Norris, "On-chip natural assembly of silicon photonic bandgap crystals," *Nature*, vol.414, pp.289-293, Nov. 2001.
- [2] H. Gau, S. Herminghaus, P. Lenz, R. Lipowsky, "Liquid morphologies on structured surfaces: from microchannels to microchips," *Science*, vol.283, pp.46-49, Jan. 1999.
- [3] O. D. Velev, E. W. Kaler, "In situ assembly of colloidal particles into miniaturized biosensors," *Langmuir*, vol.15, pp.3693-3698, May 1999.
- [4] available sizes of polystyrene spheres
- [5] J.Y. Cheng, C.A.Ross, E.L. Thomas, H.I. Smith, G.J. Vancso, "Templated self-assembly of block copolymers: Effect of substrate topography" *Adv. Mater.* Vol.15, pp.1599-1602, October 2003.
- [6] P. N. Bartlett, M. A. Ghanem, I. S. El Hallag, P. de Groot, A. Zhukov, "Electrochemical deposition of macroporous magnetic networks using colloidal templates," *J.Mater.Chem*, vol.13, pp.2596-2602, Aug. 2003.
- [7] W. Kern and J. Vossen, *Thin Film Processes*, Academic Press: New York, 1978, Ch V-1.
- [8] J.Israelachvili, "Intermolecular Surface Forces" 2nd Edn, Academic Press, London, pp.176 -177, 1992.
- [9] A. A. Zhukov, A. V. Goncharov, P. A. J. de Groot, P. N. Bartlett, M. A. Ghanem, H. Küpfer, R. J. Pugh and G. J. Tomka, "Patterned magnetic media from self-assembly template methods," *IEE Proc.-Sci. Meas. Technol.*, vol.150, pp.257-259, Sept. 2003.
- [10] A. A. Zhukov, M. A. Ghanem, A. V. Goncharov, R. Boardman, V. Novosad, G. Karapetrov, H. Fangohr, P. N. Bartlett and P. A. J. de Groot, "Oscillatory Thickness Dependence of the Coercive Field in Magnetic 3D Anti-Dot Arrays," # *cond-mat/0406091*.
- [11] The "Object Oriented Micromagnetic Framework" developed at NIST, Gaithersburg - <http://math.nist.gov/oommf/>.
- [12] R. Skomski, and J. M. D. Coey, "Permanent magnetism," Institute of Physics Publishing, 1999.
- [13] J. M. Garcia, A. Thiaville, J. Miltat, K. J. Kirk, J. N. Chapman, F. Alouges, "Quantitative interpretation of magnetic force microscopy images from soft patterned elements," *Appl. Phys. Lett.* Vol.79, pp.656-658, July 2001.

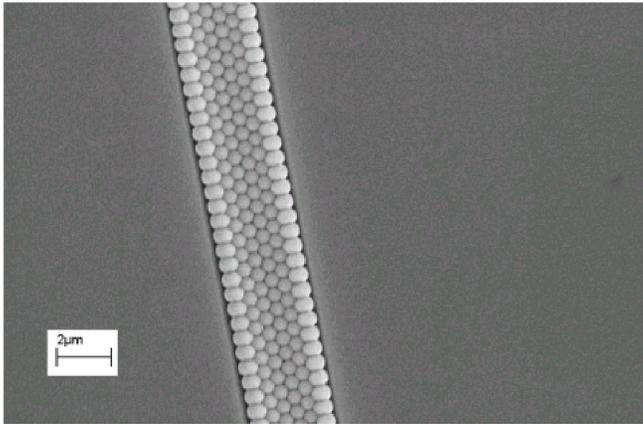


Fig. 1. SEM image of 500nm diameter spheres self-assembled in a stripe groove of a patterned Si/SiO₂ substrate.

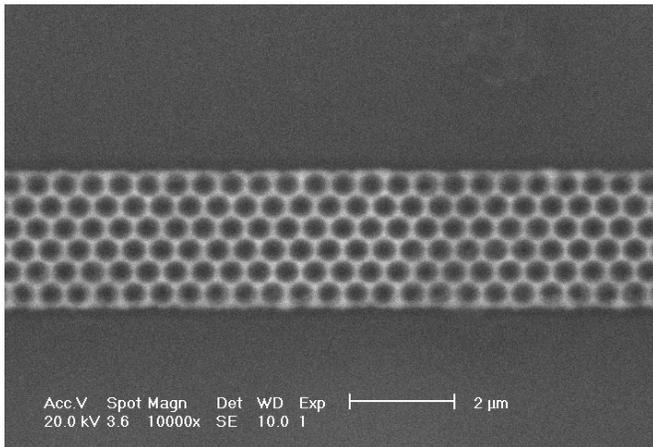


Fig. 2. SEM image of a nanostructured Ni film prepared by guided self-assembly of 500 nm spheres on a patterned Si/SiO₂ substrate

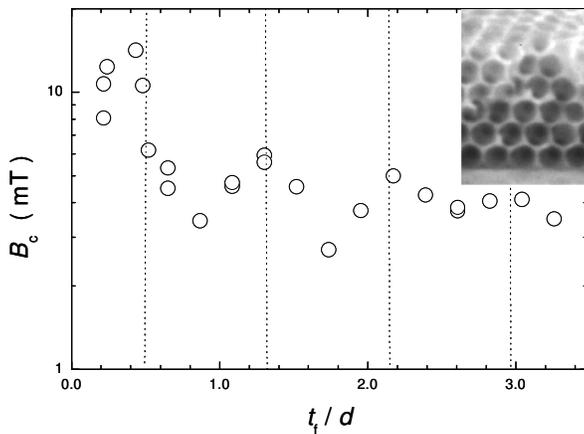


Fig. 3. Coercive field for Ni₅₀Fe₅₀ films with different thickness t_f prepared using 550 nm spheres. The dashed lines show the positions of sphere centers for each layer in a close packed structure. The inset demonstrate layered 3d architecture for a thick film.

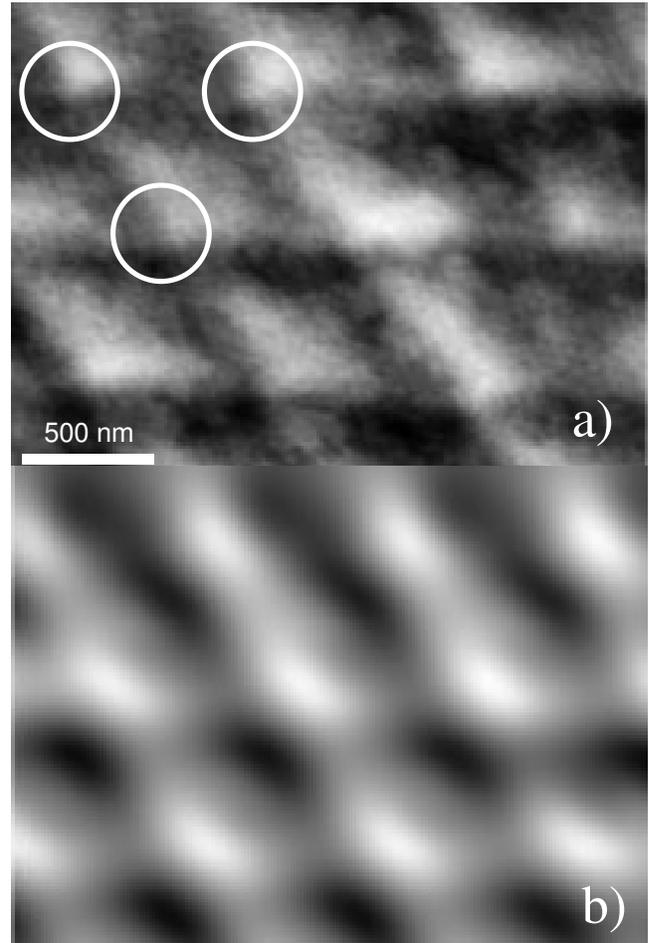


Fig. 4. MFM images – measured for a Co anti-dot film with $d = 700$ nm and $t_f = 100$ nm (a) and from micromagnetic modeling (b). White circles indicate the hole positions.

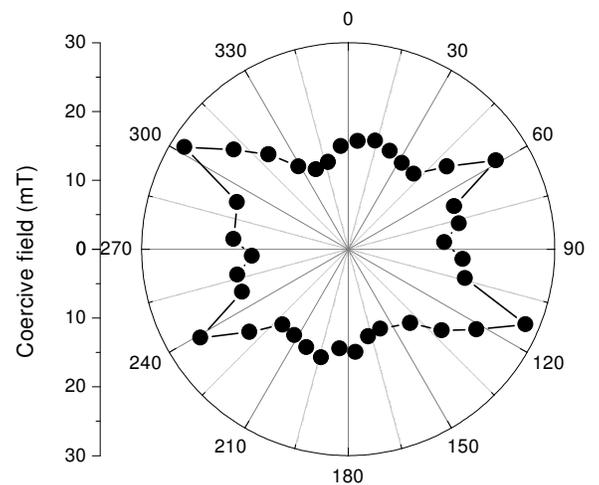


Fig. 5. Anisotropy of coercive field in plain Ni antidot array prepared from 900 nm spheres. Zero angle corresponds to the direction of a unit vector of the array.