Oscillatory thickness dependence of the coercive field in three-dimensional anti-dot arrays from self-assembly

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We present studies on magnetic anti-dot nano-structures with three-dimensional (3D) architectures, fabricated using a self-assembly template method. We find that patterning transverse to the film plane, which is a unique feature of this method, results in novel magnetic behavior. In particular, one of the key parameters for a magnetic material, the coercive field B_c, was found to demonstrate an oscillatory dependence on film thickness.
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The demand for higher magnetic recording densities is stimulating intense research activity into magnetism on submicrometer 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 colloidal particles have been considered for various applications such as photonic materials,[1,2] microchip reactors,[3] and biosensors.[4] In this work we use ordered templates for the electro-deposition of different magnetic materials. This technique offers new opportunities, which are not easily realized by standard lithographic methods, and allows us to create magnetic nano-structures with 3D architectures on a broad range of length scale, 50–1000 nm.

Using slow (3–5 days) evaporation of a colloidal water suspension containing 1 wt % of latex spheres well-ordered templates have been self-assembled on glass substrates with buffer layers of Cr (10 nm) and Au (200 nm) prepared by sputtering. These templates have been further used as molds to prepare nano-porous magnetic structures by electrodeposition methods.[5] After deposition the latex spheres can be removed by dissolving them in toluene. Using this method, well-ordered nano-structured 3D arrays were prepared for various magnetic materials such as cobalt, iron, nickel, and soft-magnetic $Ni_{50}Fe_{50}$ alloy. An SEM image of one of the nano-structured films is given in Fig. 1(a). It shows that the films have excellent hexagonal order. In comparison with conventional lithographical techniques our method has a significant advantage. It also produces structuring in the direction transverse to the film plane. Cross-sectional SEM [Fig. 1(b)] demonstrates that the transverse structuring is also well ordered.

Magnetization measurements, using a vibrating sample magnetometer (VSM), have been used to investigate the effect of the array of uniform spherical voids on the magnetic behavior of the nano-structured films. Characteristic hysteresis loops, for the sample at room temperature and for magnetic fields (***B***) applied parallel to the film plane, are presented in Fig. 2(a). The nano-structuring significantly

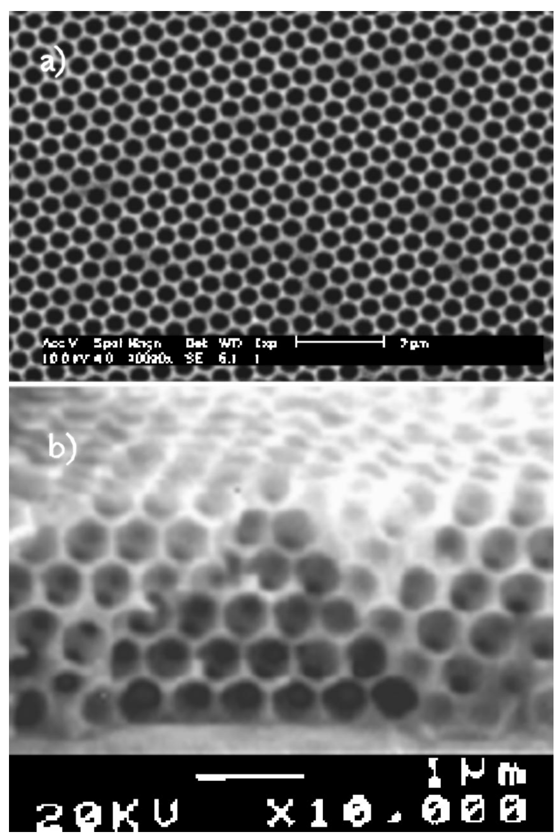

FIG. 1. SEM image of structured Ni film prepared using 0.5 μm polystyrene spheres (a) and cross-sectional view for a thick film after cleaving (b).

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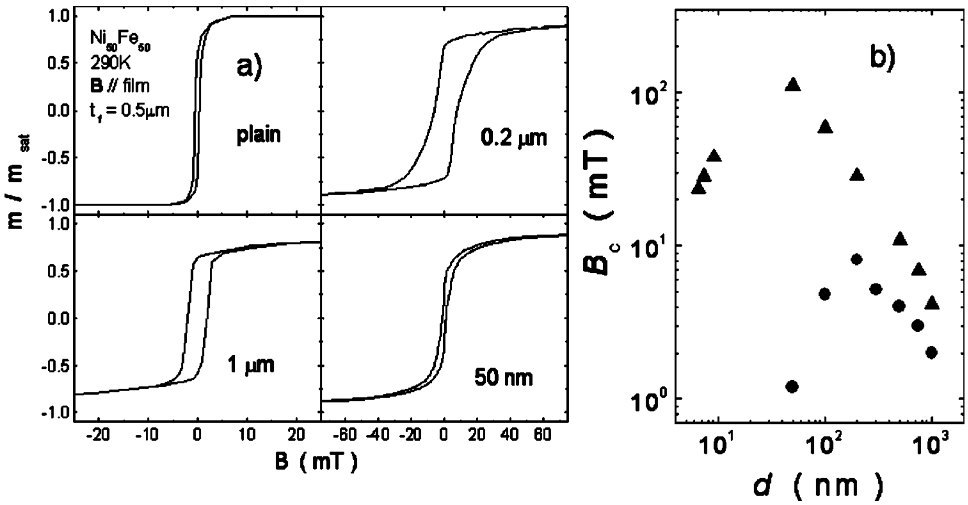

FIG. 2. Magnetization curves obtained by VSM for nano-structured and plain $Ni_{50}Fe_{50}$ films with different pore diameters (a). The dependence of coercive force on pore diameter for $Ni_{50}Fe_{50}$ (circles) and Co (triangles) films (b). The Co films with very small pore sizes ($d<20$ nm) were prepared using soft templates (Ref. 6).

changes the magnetization loops. The loops presented in Fig. 2(a) show that the irreversibility field, which corresponds to the merging point of increasing and decreasing field branches, monotonically increases with a decreasing diameter of the spherical voids. In contrast, the coercive field B_c, shows a nonmonotonic change. This can be seen in Fig. 2(b) for Co and $Ni_{50}Fe_{50}$ structures. First, the coercivity increases by more than an order of magnitude for decreasing pore size ($1000\text{ nm} \geqslant d \geqslant 50$ nm). Note that $B_c=2.8$ mT for Co films and 0.6 mT for $Ni_{50}Fe_{50}$ films deposited by a similar electrochemical route but without template. After reaching a maximum, B_c decreases for further decrease of the anti-dot diameter. We found the value of the diameter corresponding to maximum coercivity, $d_{\max}$, to agree with the condition $\frac{1}{2}d_{\max} \approx w_B$ with w_B the domain wall width for the material. Indeed, the domain wall width is $w_B=14$ nm for Co.[7] For $Ni_{50}Fe_{50}$, taking as a rough estimate an average value between Ni and Fe[7] we get $w_B=70$ nm, again in agreement with the condition $\frac{1}{2}d_{\max} \approx w_B$. This indicates the importance of domain wall pinning for such anti-dot structures.

Measurements of the dependence of B_c on the thickness of the magnetic film revealed a novel effect. Although these films have a homogeneous composition of magnetic material, as evident from the EDSRX studies, we have found that the coercive force changes periodically with film thickness. This is a clear manifestation of the periodical patterning in the direction transverse to the film and the 3D architecture of these structures. Figure 3 demonstrates that B_c shows clear oscillations and reaches a maximum for the case when the top surface of the film is near the center of a layer of close packed spherical voids. For complete spherical layers 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.

Presence of domain boundaries can be seen from the MFM images. They were obtained with a Digital Instruments 3000 scanning probe microscope using lift-mode with a fly height of 100 nm and a standard low-moment ferromagnetic tip. Results are shown in Fig. 4 for a 100 nm thick Co film with $d=700$ nm in the remanent state. These images reveal ordered, rhombic magnetic patterns associated with the hole array. Magnetic structures for two-dimensional (2D) hole arrays were calculated using micromagnetic modeling with the OOMMF software suite.[8] The large number of computational cells ($\sim 10^8$) required for 3D case to explain the $B_c(t_f)$ data appears above present computing powers. Therefore, we

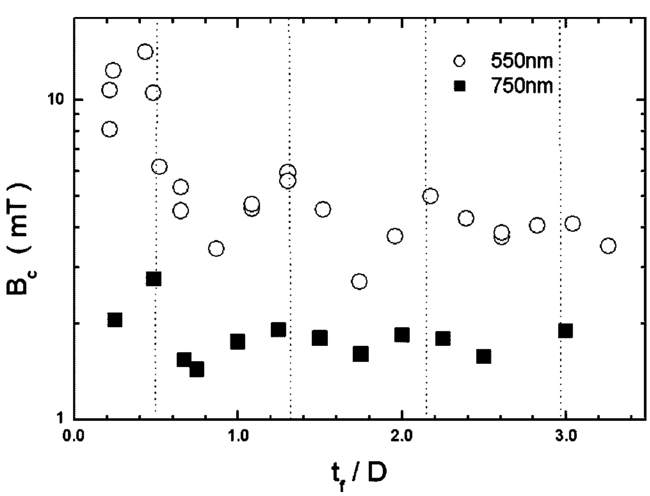

FIG. 3. Coercive field for $Ni_{50}Fe_{50}$ films with different thickness prepared using 550 and 750 nm spheres. For clarity, in the case of 750 nm the B_c values are divided by factor 2. The dashed lines show the positions of sphere centers for each layer in a close packed structure.

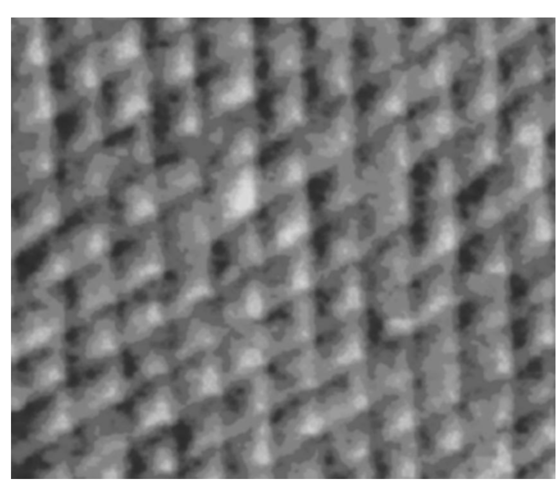

FIG. 4. MFM image for a Co anti-dot film with $d=700$ nm and $t_f=100$ nm.

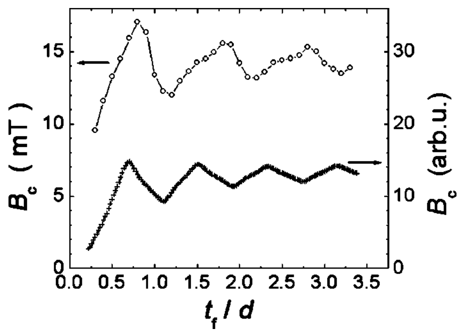

FIG. 5. Model dependencies of the coercive field for $Ni_{50}Fe_{50}$ films with $d=0.5$ μm from the homogeneous-layer micromagnetic model (open circles) and from the domain wall pinning model (with $d/w_B=25$, crosses).

have employed a simplified approach. Using the values of $B_c(D/d)$ found from the 2D numerical simulations we consider our system as a multilayer with properties averaged in the plane direction. By a Monte Carlo method we calculate $B_c(t_f)$ for this stack of exchange coupled layers. As shown in Fig. 5 the results of the numerical simulations reproduce our experimental behavior. Note that for simplicity we have chosen the period in the perpendicular direction to be d in this model; the period of the $B_c(t_f)$ oscillations reflect this.

We can qualitatively understand the results of our experiment assuming that domain wall pinning is the main mechanism for the coercive properties. In this case the narrow constrictions between the spherical voids are dominant pinning sites. The number of effective pinning sites changes periodically with the film thickness t_f. Using this, we can describe the observed behavior using a simple model of domain wall pinning. In this model we assume a flat domain wall and take its energy E_b to be proportional to the volume of the domain wall. From the dependence of E_b on domain wall position we calculate the coercive field B_c.[7] As shown in Fig. 5 model results also reproduce good experimental observation of the $B_c(t_f)$ oscillations.

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