Ferromagnetic Resonance Investigation of Macroscopic Arrays of Magnetic Nanoelements Fabricated Using Polysterene Nanosphere Lithographic Mask Technique

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A dense plane periodical array of cylindrical magnetic nanodots has been fabricated using a lithographic mask formed by self-organization of polystyrene nanospheres. In this paper, we study collective static and dynamic magnetic behavior of this array. We find that this technique produces samples with reasonably small dispersion of magnetic parameters of individual dots. This is evidenced by magnetometry and well-resolved discrete frequencies of standing spin waves measured with cavity and coplanar-waveguide ferromagnetic resonance. The standing spin wave resonances could be reliably observed in a large range of frequencies (4–15 GHz). However the measured linewidth of resonances is about ten times larger than for unpatterned Permalloy. This may be due to some variation in shape and magnetic parameters from dot to dot resulting in inhomogeneous broadening of the resonance lines.

Index Terms—**Ferromagnetic resonance, magnetic nanodot, self-organized structures.**

I. Introduction

PATTERNED metallic magnetic films are important for magnetic memory [1], magnetic logic [2], and microwave signal processing [3] applications. Dense arrays of magnetic elements with small dot-to-dot separation are important for functionality of magnetic logic [2]. Increasing array density also increases density of magnetic memory.

A major issue is low cost fabrication of large-scale periodic structures. Structuring centimeter-sized areas of films into dots or rings with diameters 100–400 nm using traditional lithography tools requires high-precision instruments and is very time consuming. However, there is another way to achieve the same goal in a more cost-efficient way. This is by using "natural lithography" based on self-organization of polystyrene nanospheres on a hydrophilic surface. The self-assembled close-packed monolayer of spheres can be used as a template for lithographic procedures, including subsequent evaporation or electrodeposition of magnetic materials forming structured elements at the places of the nanospheres [4], [5]. These methods are however still limited in the size of the periodic domains (typically $<1 \times 1$ mm^2) which restricts application of characterization techniques requiring larger magnetic volume, if periodic properties are under focus. An additional requirement of growth by electrodeposition also restricts application of these methods for fabrication of epitaxial or high quality sputtered (e.g., multilayered) films. The latter, however, is normally a prerequisite for spin-valve and spin-tunnel structures, utilized in read-head technology, and the new generation of spin-torque transfer devices

Another alternative, which is considered in this article, is the method suggested by Weekes *et al.* [4] in which a periodic single layer pattern of nanospheres is assembled on a liquid-air interface and than transferred directly on a predeposited magnetic layer. This method allows one to significantly improve the periodicity of the structure ($\sim 1 \times 1$cm^2 ordered domains) and can be used for patterning of many metallic or dielectric materials. In this work we fabricated a dense periodic array of Permalloy nanodots of circular shape using this technology and studied static and dynamic magnetic properties of the array.

II. Fabrication

The array sample was fabricated at the University of Exeter. Details of the method can be found in [4], and here we provide a brief outline. Surface-activated polystyrene nanospheres were self-assembled on a water–air interface to produce a 2-D array with hexagonal symmetry. The assembled array was then transferred onto the surface of a Permalloy film predeposited on a highly polished SiO_2 substrate in the form of rectangle with area 25×25 mm^2. Further patterning was achieved by ion milling in argon plasma through the nanosphere template. The dot arrays produced with this method had a well-defined hexagonal symmetry with a long-range periodicity over 10–20 mm. The array period is equal to the initial diameter of the nanosperes and can be adjusted by choosing nanospheres with a required diameter. The diameter of the final dots can be further modified by etching the nanospheres in oxygen plasma.

In this paper, the thickness of the predeposited Permalloy layer was 25 nm. The nanosphere template was created using carboxyl-modified polystyrene spheres (as described in [4]). The nanosphere diameter was 390 nm, which determined the array period.

Scanning electron microscopy (SEM) micrographs taken from the fabricated sample verified that the average diameter of the magnetic dots was 310 ± 5 nm. Thus, in contrast to the sample fabricated in [4], the present array of dots is much

Digital Object Identifier 10.1109/TMAG.2008.2002475

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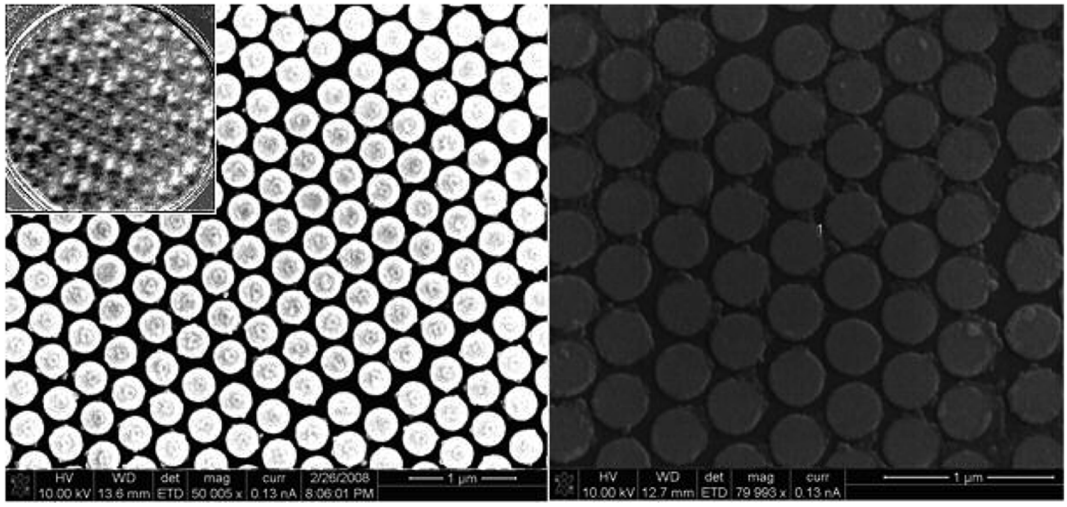

Fig. 1. SEM images of the sample. Left panel: nanodots are covered by polystyrene caps. Right panel: after removing the caps, enlarged. Inset: XPEEM image taken at zero field for a sample (650-nm diameter dots) produced with the same technique and having similar characteristics of a magnetization loop. The image shows magnetic contrast characteristic to a vortex formation.

denser, with average edge-to-edge distances between neighboring dots of 80 nm only. SEM also revealed that the deviation of the lattice translational order in the self-assembled array was less than 5° per 1 cm. The circular shape of the dots was slightly affected by the milling procedure leading to a nonuniform edge with approximately 5-nm variation in diameter.

This technique usually leaves magnetic nanodots covered by caps consisting of the rest of the polysterene nanospheres, as shown in Fig. 1 (left panel) for the sample under study. If necessary, the caps can be removed. An SEM image from a part of the sample where the caps were removed is shown in the right panel of Fig. 1. From these figures, one sees that the array is quite dense. Indeed, starting with the geometry of the dot 2-D lattice, one can find the filling factor of the array plane by Permalloy: 58%.

A 5×10 mm rectangle was cut from the area with high periodicity to be used for ferromagnetic resonance and SQUID magnetometry characterization. The dots on this piece of the sample remain covered by the polystyrene caps.

III. Hysteresis Loop Measurements

A hysteresis loop measured with SQUID at room temperature with the field applied in the array plane is shown in Fig. 2. The loop shows distinctive narrowing at small field values which suggests formation of a vortex state in remanance. Indeed, XPEEM measurements carried out on a different sample from the same series of fabricated self-organized arrays showed formation of a vortex [6] in the remnant state for this range of dot diameters (see inset in Fig. 1). The well defined loop shape indicates uniform response of all dots, suggesting that dispersion of static magnetic properties of individual dots on the array is small.

The filling factor and the total magnetic moment for the array measured in saturation can be used to estimate the saturation magnetization $(4\pi M_{\mathrm{s}})$ for the dots: 3370 G which is surprisingly low. The dashed line in the figure shows a result from OOMMF [7] simulation of the hysteresis curve for this value of $4\pi M_{\mathrm{s}}$. The simulation was performed for a single dot, since the experimental data in Fig. 2 in [8] show negligible influence of

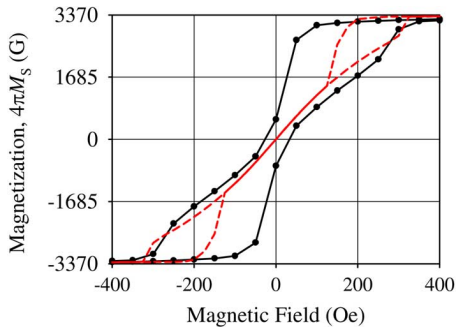

Fig. 2. In-plane hysteresis loop measured with SQUD. Dots: experiment, dashed line: OOMMF simulation. The solid line connecting dots serves as guide for the eye. Material Parameters for OOMMF simulations: $4\pi M_{\mathrm{s}} = 3370$ G, exchange constant: $0.35 \cdot 10^6$ erg/cm.

interdot dipole coupling for our values of the ratio of the interdot distance to the dot thickness. One sees qualitative agreement with the experiment. This simulation shows vortex formation. Both experimental and theoretical loops have the same saturation field about 300 Oe.

The major discrepancy is in the value of field (100 Oe in the experiment, 200 Oe in the theory) at which the vortex starts to form when the applied field is decreased from saturation. From the experimental curve one also sees that the array possesses a net magnetic moment in the zero field. There should be no net in-plane magnetic moment if all dots are in the vortex state in the zero field and the vortex structure of individual dots is perfect. We suppose that there is noticeable distribution of vortex nucleation and formation fields for dots on the array, and that some dots do not form a vortex magnetization configuration at all. Variation of dot shape and sizes from dot to dot may be the reason for this distribution. One cannot also exclude some contribution of dipole coupling of dots to the the non-vanishing magnetic moment of the array at small fields [8]–[10]. MFM imaging of the array is required in order to determine the magnetization configuration of individual dots. This is out of scope of the present paper.

IV. Ferromagnetic Resonance Characterization

Dynamic magnetic behavior of the self-organized array was characterized by cavity and coplanar-waveguide [11] ferromagnetic resonance (FMR). A standard Bruker ESR setup at the Institute of Crystallography, Moscow, operating at 9.55 GHz was used for the cavity FMR study. To characterize the resonance behavior in a vide range of microwave frequencies (0–18 GHz) we used absorption of microwave power by quasi-homogeneous precession of magnetization in a sample placed on top of a coplanar waveguide ("coplanar-waveguide FMR"). These measurements were done at the University of Western Australia. The main details of the method can be found in [11]. The major difference of our setup from that used in [11] is that instead of a microwave vector analyzer we used a microwave generator and a lock-in amplifier. This allows one reliably measure absorbed power but does not have phase resolution. The latter is not important in our case.

We used square-wave modulation of the generator output microwave signal to increase setup resolution (modulation depth: 100%). The modulation frequency was 10 kHz. The modulated microwave signal is split into two channels by using a 3-dB power splitter. The signal R from one channel (the reference arm) is rectified by a microwave detector and sent to an input of the lock-in amplifier which is locked to the modulation frequency. The microwave power from the other channel (signal arm) is applied to the input of the coplanar line. The signal S transmitted through the coplanar line is also rectified by a detector and sent to the second input of the lock-in. Using a variable microwave attenuator in the reference arm one is able to set the $R = S$ for zero applied field prior starting measurements. The power absorption in FMR is proportional to $[R-S(H)]/R$. By sweeping the applied magnetic field H while keeping the microwave frequency constant one can observe ferromagnetic resonance.

Spectra from in-plane and out-of-plane cavity FMR are shown in Fig. 3. One sees three distinct peaks for the out-of-plane configuration. Following [12] these peaks are axially symmetric modes of standing spin waves across the nanodisk diameter. The presence of several well-resolved peaks in the out-of-plane conventional FMR spectrum evidences that the nanosphere lithographic technique is able to produce quality dot samples with small dispersion of geometrical and magnetic parameters. However, the parameter dispersion is not entirely negligible, since the resonance line (235 Oe) is about 10 times larger than one expected for an unstructured Permalloy film. This can probably be partly attributed to a small difference in "magnetic" diameters and shapes of individual elements resulting in a small variation of resonance fields from dot to dot. This conclusion is consistent with the above results from hysteresis loop measurements. Another possible contribution to the large resonance linewidth is the magnetic material itself. The value of $4\pi M_{\mathrm{s}}$ of 3370 Oe shows that the film does not probably have composition and or crystallite structure corresponding to the smallest possible microwave losses for Permalloy.

Using the approximate formulas in [12] one can estimate resonance fields of out-of-plane resonances in circular dots theoretically. This theory does not account for eventual coupling of neighboring dots by static and dynamic dipole fields. As seen in Fig. 1 of [10], the contribution of dot dipole coupling to the total resonance field is well below 100 Oe which gives an error well below 20% in the applied field range used in our experiment.

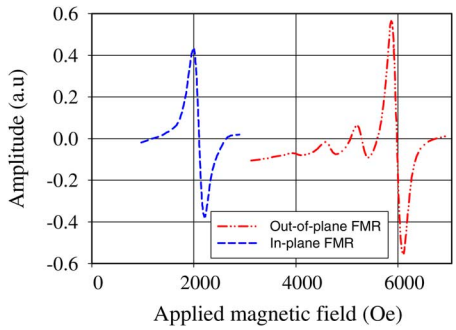

Fig. 3. Cavity FMR spectra taken at 9.55 GHz. Left (dashed) line: applied field in the array plane. Right (dash-dotted) line: applied field is out of plane.

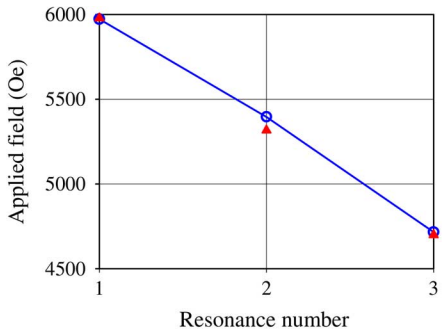

Fig. 4. Out-of-plane resonance fields. Triangles: experimental data extracted from Fig. 3. Open circles: theory from [12]. The solid line connecting the circles serves as guide for the eye. Parameters of calculation: dot thickness: 25 nm, dot diameter: 310 nm, saturation magnetization $4\pi M_{\mathrm{s}}, = 3300$ G, exchange constant: $0.35 \cdot 10^6$ erg/cm.

Fig. 4 shows a comparison of the experimentally measured fields with calculated values. Agreement is very good, but in contrast to [12] the dot size is in our case is in the nanometer range. Therefore one expects considerable contribution of exchange energy to the frequencies of standing spin waves. This contribution is mode number dependent which allows extraction of a value for the exchange constant from the experimental data. The best agreement between the theory and the experiment is obtained setting the exchange constant to 0.35×10^6 erg/cm and $4\pi M_{\mathrm{s}}$ to 3300 G. The $4\pi M_{\mathrm{s}}$ value deviates only by 2% from the measured with SQUID. The exchange constant is three times lower than the standard value for Permalloy.

We used this value of the exchange constant in the OOMMF simulation of the hysteresis loop in Fig. 2. OOMMF simulations for larger values of exchange constant do not show vortex formation and result in hysteresis loops of a different shape. This is additional evidence that the obtained value of exchange constant is consistent with experiment.

Theoretical interpretation of the in-plane spectrum is more complicated because of the strong anisotropy of propagation of spin waves in this geometry [13]. Furthermore, only one (fundamental) mode is seen for this magnetization direction in Fig. 3. We limit ourselves to a comparison of the dot resonance field with the resonance field for an in-plane magnetized continuous film, keeping in mind that the resonance field for the fundamental mode of a dot should be close to this value. By setting $4\pi M_{\mathrm{s}}, = 3300$ G one obtains the resonance field for an in-plane

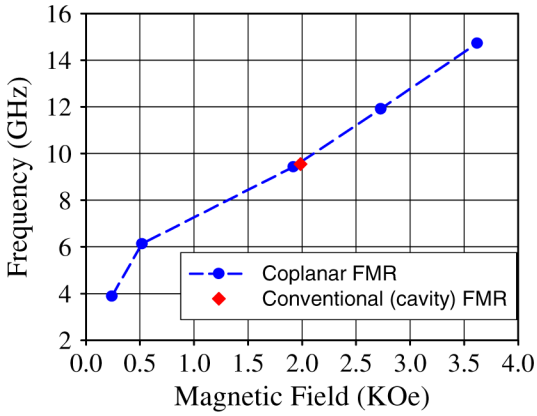

Fig. 5. Resonance frequency of the fundamental in-plane resonance mode as function of applied field. Dots: experimental points. Triangle: point measured with conventional FMR, extracted from Fig. 3. The dashed line connecting the dots serves as guide for the eye.

magnetized continuous film of 2075 Oe. This value is very close to the value in Fig. 2 of 2100 G.

Fig. 5 shows the results of the measurements using the coplanar-waveguide FMR technique. The measurements were done in a range of frequencies and magnetic fields applied in the plane of the array. Sensitivity is an issue, since the amount of magnetic material contained in the sample is very small, and the measurements are done at response levels close to the sensitivity threshold. The experimental point obtained by the conventional FMR technique is also shown in the figure. One sees very good agreement of results for both measurements.

Resonance absorption was reliably registered in the range of frequencies 4–15 GHz. Thus, the obtained coplanar FMR data evidence that the arrays of magnetic nanodots fabricated using the nanosphere lithography shows coherent magnetic dynamics over a wide range of microwave frequencies.

In Fig. 5, one sees softening of the resonance mode in the applied fields below 500 Oe. Comparison of this field range with the saturation field in Fig. 2 suggests that softening is due to vortex nucleation.

V. Conclusion

In this paper, we studied magnetic properties of a dense plane periodical array of magnetic nanodots fabricated using a lithographic mask formed by self-organization of polystyrene nanospheres on a water surface. We were interested in the sum response over the whole dot system. This coherent response has been traced over a wide range of frequencies and magnetic fields. Analyzing the response we find that dispersion of magnetic parameters of individual dots is small. In particular, this is evidenced by well-resolved discrete frequencies of standing spin waves across the dot diameter. We observed these resonances in a large range of frequencies (4-15 GHz) and applied fields (0.2–3.6 kOe). A well-defined hysteresis curve suggests vortex formation in most of the dots in the remnant state.

The linewidth of standing spin wave resonances is about ten times larger than the standard one for unpatterned Permalloy. This can be partly explained by some variation of geometrical and magnetic parameters from dot to dot resulting in inhomogeneous broadening of the resonance line. Another possible contribution to the linewidth is the composition and crystallographic structure of magnetic material of the dots which may be far away from the standard composition of Permalloy. This fact is evidenced by much lower values of saturation magnetization and exchange constant derived from our measurements.

Additional investigations are necessary to understand whether the magnetic film possessed this small value of the saturation magnetization before patterning, or the saturation magnetization value decreased for some reason during "the self-organized" patterning procedure. MFM imaging is necessary to investigate magnetic configuration of individual dots on the array.

Acknowledgment

This work was supported in part by the Australian Research Council under Discovery Grant "Magnetic nanostructures for emerging technologies." The authors would like to thank Yu. K. Fetisov, K. J. Kennewell, and R. Woodward for their assistance and helpful discussions.

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