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Inkjet printing of magnetic materials with aligned anisotropy

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3-D printing processes, which use drop-on-demand inkjet printheads, have great potential in designing and prototyping magnetic materials. Unlike conventional deposition and lithography, magnetic particles in the printing ink can be aligned by an external magnetic field to achieve both high permeability and low hysteresis losses, enabling prototyping and development of novel magnetic composite materials and components, e.g., for inductor and antennae applications. In this work, we report an inkjet printing technique with magnetic alignment capability. Magnetic films with and without particle alignment are printed, and their magnetic properties are compared. In the alignment-induced hard axis direction, an increase in high frequency permeability and a decrease in hysteresis losses are observed. Our results suggest that unique magnetic structures with arbitrary controllable anisotropy, not feasible otherwise, may be fabricated via inkjet printing. © *2014 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4863168]

INTRODUCTION

3-D printing processes are becoming increasingly important as a rapid, versatile, cost-effective production technology. Recent research shows that this technology has great potential in designing and producing magnetic materials.[1–3] "Magnetic ink," which is a dispersion of magnetic nanoparticles[1] or a solution composed of magnetic material precursors,[2] can be ejected by a drop-on-demand inkjet printhead onto almost any kind of substrate.

Unlike conventional thin-film deposition and lithography, inkjet printing allows for the magnetic orientation of the particles to be arbitrarily controlled within the film or component: a magnetic field can be applied during jetting for aligning the particles as desired. The alignment of magnetic particles increases the high frequency permeability and lowers the hysteresis losses.[4] Additionally, the number of printing passes can be varied to rapidly prototype magnetic films in different thicknesses or components in different dimensions for applications such as inductors and antennae.

In this work, we demonstrate inkjet printing techniques for rapidly prototyping films and components comprised of magnetic particles. Magnetic films (square shaped and ring shaped) are printed with and without particle alignment. Magnetic properties are compared by vibrating sample magnetometry (VSM), ferromagnetic resonance (FMR) spectroscopy, and high frequency permeability measurements.

EXPERIMENTAL TECHNIQUE

A printing ink with a high solid content (40% by volume), low viscosity (about 6 cP) aqueous suspension of ferromagnetic Co-based nanoparticles (mean particle size = 40 nm) was used. This ink was jetted using a nozzle with a 60 μm orifice and controlled by an HP Thermal Inkjet Pipette System (TIPS) (Fig. 1) integrated into a custom printing and magnetic alignment setup (Fig. 2). Commercial inkjet paper, which can quickly absorb the ink solvent, was used as substrate. An electromagnet was placed below the inkjet nozzles to apply a magnetic field for aligning the magnetic particles in the printed samples. To avoid clogging from agglomeration of the particles in the inkjet printhead, a magnetic shield made from Mu-metal was mounted on the inkjet nozzles. During printing, the controller and the alignment field direction were fixed, while the inkjet paper was moved by a high-resolution (50 μm) stepper motor XY stage or rotated by a rotation stage.

Two types of samples were made: square and ring shaped. For square samples, during printing, the alignment field was applied in the x direction as shown in Fig. 2. The XY stage moved the substrate in the x and y direction, with the ink being jetted during translation in the x direction. The samples were printed to be 5 mm by 5 mm. For ring shaped samples, the alignment field was fixed, while the substrate was rotated

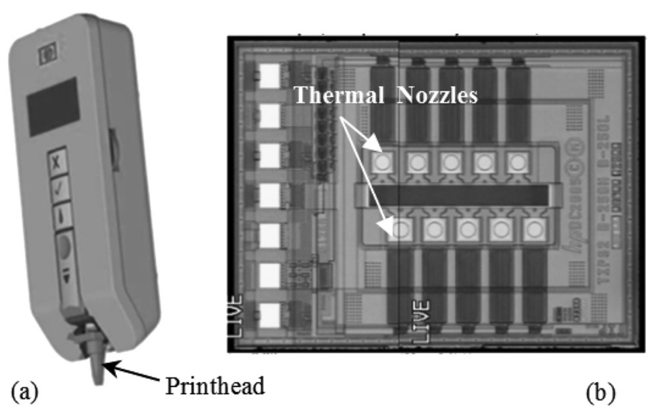

FIG. 1. (a) TIPS controller. (b) Nozzles on the thermal inkjet printhead used with the controller. (Photos courtesy of Hewlett-Packard Company).

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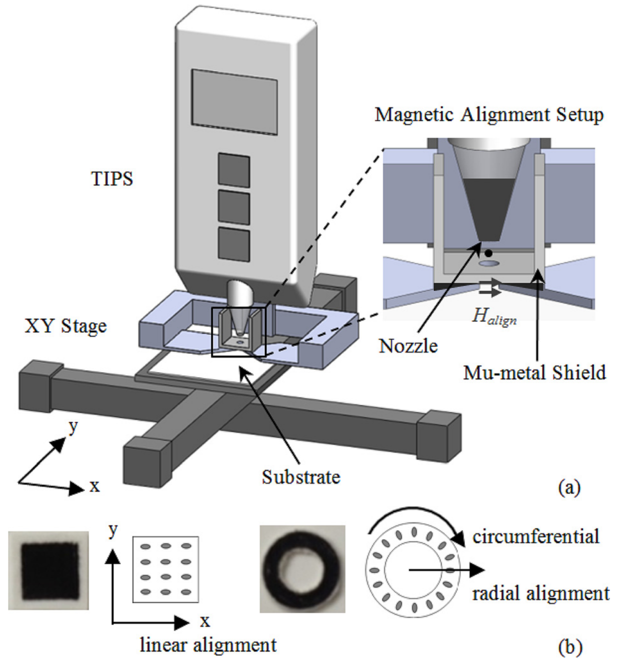

FIG. 2. (a) Schematic of inkjet printing and magnetic alignment setup. (b) Photos and schematics of x direction aligned square sample and radial direction aligned ring shaped sample.

at a speed of 2 rpm during inkjetting resulting in radial alignment (Fig. 2). The ring shaped samples have an inner diameter of 4 mm and an outer diameter of 7 mm. The dot pitch for both printing processes was 100 μm. All samples were printed with one layer and air-dried prior to characterization.

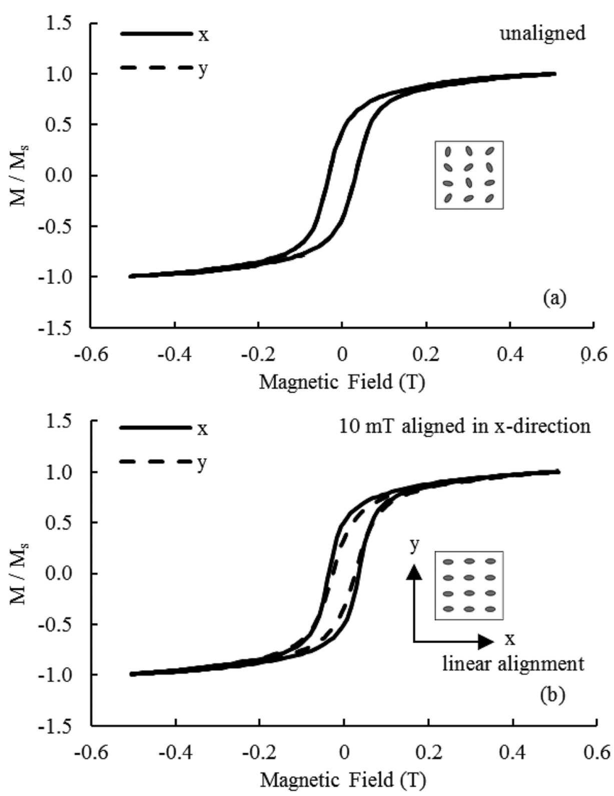

FIG. 3. Normalized hysteresis curves measured in both x and y directions for (a) an unaligned 5 mm square sample and (b) a 10 mT, x direction aligned 5 mm square sample.

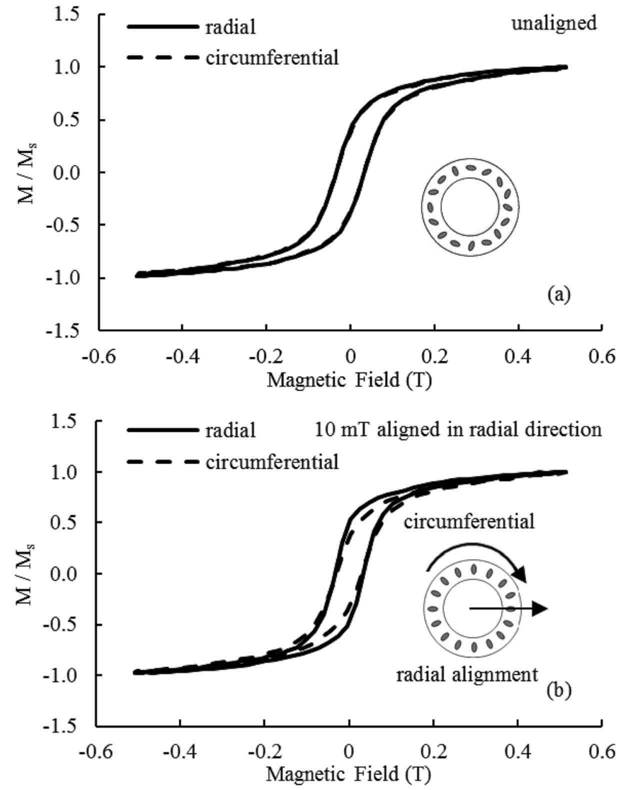

FIG. 4. Normalized hysteresis curves measured in both radial and circumferential directions for (a) an unaligned ring shaped sample and (b) a 10 mT, radial direction aligned ring shaped sample.

Both types of samples were printed with and without a 10 mT magnetic alignment field. The magnetic properties of square samples in x and y direction were compared by VSM and FMR measurements. For ring shaped samples, high frequency permeability measurement was first done by an impedance analyzer (Agilent E4991A RF Impedance/Material Analyzer). Then the samples were cut into slices along radial direction and measured by the VSM and FMR spectrometer in both radial and circumferential directions.

RESULTS AND DISCUSSION

Shown in Figs. 3(a) and 3(b), respectively, are the normalized hysteresis curves of an unaligned and a 10 mT, x

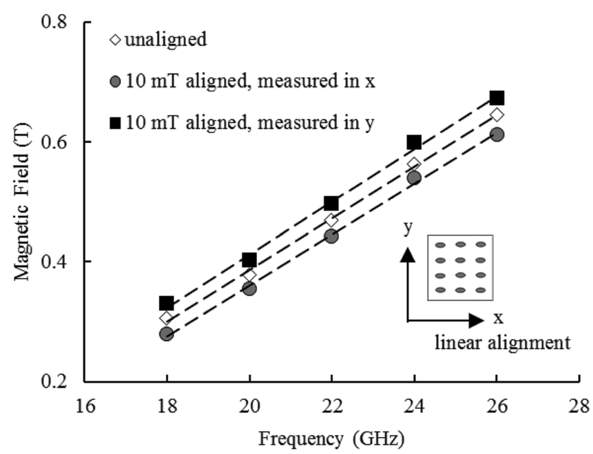

FIG. 5. Frequency dependence of the FMR field in both x and y directions for an unaligned 5 mm square sample and a 10 mT aligned 5 mm square sample.

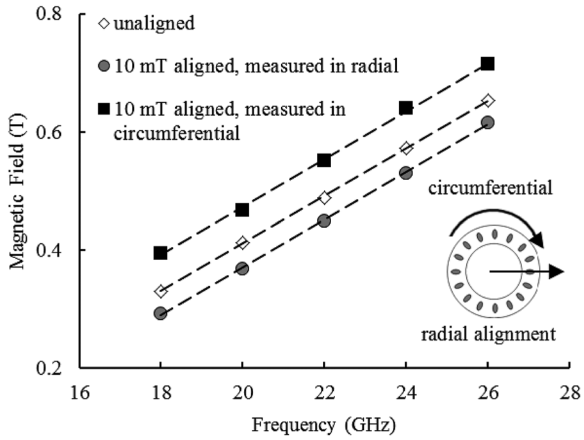

FIG. 6. Frequency dependence of the FMR field in both radial and circumferential directions for an unaligned ring shaped sample and a 10 mT, radial direction aligned ring shaped sample.

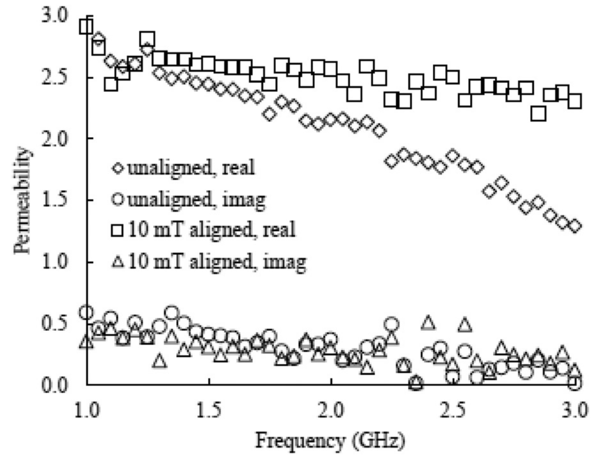

FIG. 7. Complex permeability of an unaligned ring shaped sample and a 10 mT, radial direction aligned ring shaped sample in the frequency range of 1 GHz to 3 GHz.

direction aligned 5 mm square sample. Similarly, Figs. 4(a) and 4(b) show the normalized hysteresis curves of radially cut slices of an unaligned and a 10 mT, radially aligned ring shaped sample, respectively.

For both square and ring-shaped geometries, the unaligned sample shows an isotropic magnetic response: after normalization, the hysteresis curves for the x and y directions overlap with each other. In contrast, the film printed, while applying an alignment field either in x direction (for 5 mm square sample) or in radial direction (for ring shaped sample), exhibits anisotropy. The hysteresis curves in the aligned directions are more square and enclose a larger area compared to the curves of unaligned sample, implying higher hysteresis losses. Measured along the alignment-induced hard axis direction (i.e., y direction or circumferential direction), the hysteresis curves are less square and have lower hysteresis losses compared to the hysteresis curves of unaligned samples. Area enclosed by the hysteresis curve is 17% less in the y direction of aligned square sample and 23% less in the circumferential direction of aligned ring shaped sample. To confirm the repeatability of these results, three 10 mT, radial direction aligned ring shaped samples were measured by the VSM. The difference in any measured value for the normalized hysteresis curves is less than 4% for these three samples.

The hysteresis curve results show the magnetic alignment field has, to some extent, oriented the easy axes of magnetic particles and induced anisotropy in the printed material. This particle alignment effect is further corroborated by ferromagnetic resonance data.

Figs. 5 and 6 show the frequency dependence of the ferromagnetic resonance field for the square and ring shaped sample, respectively. Difference in the resonance behavior between samples is due to anisotropy alignment. In x direction, the FMR frequency shifts by 0.8 GHz from that of unaligned sample, which indicates a 0.028 T increase in sample anisotropy due to the particle alignment. While for radial direction, the FMR frequency shifts by 1.1 GHz. The corresponding increase in anisotropy is 0.040 T. However, in the y direction or circumferential direction, the FMR frequency decreases, which limits the magnetic material working frequency range for high frequency applications.

The effect of magnetic alignment is also seen in the high frequency permeability measurements performed on ring-shaped samples (Fig. 7). Compared to unaligned sample, the permeability of the aligned sample greatly increases as frequency increases (a 77% increase at 3 GHz). Because the inductance is directly proportional to the relative permeability, the alignment also increases the inductance of magnetic composite for inductor applications. For aligned sample, the real part of relative permeability increases, while the imaginary part stays relatively the same compared to that of unaligned sample. Therefore, loss tangent ($\tan\delta = \frac{\mu''}{\mu'}$) is also greatly lowered by the radial alignment.

In sum, magnetic alignment, which orients the easy axes of magnetic particles in the same direction as the applied field, increases high frequency permeability and decreases hysteresis losses. But it also limits the working frequency range by lowering FMR frequency. Magnetic ink composed of magnetic nanoparticles with high shape anisotropy, such as flake shaped magnetic particles,[5] can be used to achieve bigger alignment effects. Moreover, if magnetic nanoparticles can be well dispersed in a UV curable material, the alignment state of magnetic particles can be better preserved by UV curing[1] and would be crucial to developing layer-by-layer 3-D inkjet printing of magnetic films and components.

CONCLUSION

An inkjet printing technique with magnetic alignment capability is demonstrated. The effect of magnetic alignment has been investigated in both square and ring shaped samples. There are significant increases in high frequency permeability and decreases in hysteresis losses in the alignment-induced hard axis direction. Further work on material engineering of magnetic ink is needed to achieve bigger alignment effects.

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