Geometry Dependent Exchange Coupling in Two-Phase Magnetic Nanocomposites

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ABSTRACT

The dependence on geometrical conditions for inter-phase exchange coupling is explored in micromagnetic simulations of two-phase $Nd_2Fe_{14}B$/α-Ferrite nanocomposite thin films. A synopsis of the existence of the phenomenon is extrapolated from energy minimization ab initio. The effect is studied with in-plane magnetization reversal where optimal conditions are determined by nanocrystal shape and array spacing. The effect manifests itself in a multi-step demagnetization process which is substantiated visually by Mumax3. The material selection is intended to be general and to be expanded for numerous nanocomposites with a hard phase matrix and soft phase nanocrystals. Applications toward technology are especially accentuated.

I. Background and Introduction

This REU experience can be summarized by three projects within the scope of micromagnetic simulation. The first being the simulation of a point dipole using Python. The second project describes the out-of-plane magnetization of a thin film computationally and analytically. The third project is the basis for the research; computational analysis on geometrical effects in two-phase magnetic nanocomposites.

The simple case of a point dipole is explored from an elementary simulation. The dipole, however arbitrary, has been chosen to be a classical approximation of the magnetic dipole moment caused by electron spin. A numerical solution is created in Python, and a plot is generated in 3-dimensional real space to express the magnetic field. Two proofs from geometrical derivations are included to validate the calculation.

Out-of-plane magnetization of a thin film is explored for a pseudo-magnetic material. The specimen under consideration is approximated to a uniformly magnetized, 2-dimensional, infinite ellipsoid with the precession of the magnetization derived computationally as well as analytically with numerical approximation. The results are compared by the angle of the magnetization out-of-plane.

The forefront of technological innovation for nearly a century has been based on binary computation and memory storage. As transistors have followed the trend of Mohr's Law, scaling effects e.g. tunneling and parasitic capacitance will be the limiting factor in the performance of future computers. It is imperative that research be focused on new materials and novel phenomena to develop technology for neuromorphic and quantum computing. This paper explores the effect of the geometry of nanocrystals within a magnetic composite matrix. The role of exchange coupling is explored at interfaces between the two-phases. Potential applications are in material design consideration for implementation into devices for magnetic storage and computing.

A series of micromagnetic simulations were run in Mumax3 to observe the precession of the magnetization of a nanocomposite. It has been observed by micromagnetic simulation that exchange coupling occurs between α-ferrite and $Nd_2Fe_{14}B$ in the case of α-ferrite nanowires [1]. It is important to note that in the study by

Wei Li et. al., the exchange coupling was most prominent when the axis of the nanowires was orthogonal to the magnetocrystalline anisotropy of the hard phase. It was observed that the exchange coupling occurred most prominently when the diameter of the nanowires was less than the exchange length of α-Ferrite. A dipolar interaction was observed by the perturbation of the magnetization of finite elements at the end interfaces of the nanowires when the demagnetizing field was aligned with the easy axis of the hard phase. Experimentally, $Nd_2Fe_{14}B$ / α-ferrite nano-composites have also shown clear indication of exchange coupling by remanence enhancing and increase of the energy product in single and multilayer films [2].

This experiment is expected to display enhancement of exchange coupling with dumbbell type nanocrystals. As a foundation, various structures are simulated including quantum dots, nanowires, and cubic grains. Geometrical considerations of nanocrystal arrays are taken into detailed account by Skomski [3]. A void exists within the literature of geometrical possibilities for an enhancement on exchange coupling in nanocomposites. It is the author's prediction that a dumbbell structure will allow for greater coupling to occur due to the large surface area at the end of the nanocrystal that can interact with the hard phase. An important consideration upon selecting nanocrystal shapes is the demagnetizing field. While an ellipsoidal object like a thin film experiences a stray field for out-of-plane magnetization, a uniformly magnetized nanowire experiences a stray field with a factor of 2π for any radial direction of external magnetic field within the x-y plane.

$$\boldsymbol{H}_{D,cyl} = \begin{pmatrix} 2\pi & 0 & 0 \\ 0 & 2\pi & 0 \\ 0 & 0 & 0 \end{pmatrix} \boldsymbol{M}$$

The calculation is simple due to the nanowire being treated as an ellipsoid, however for a dumbbell structure, the calculation is more complex. It can be predicted that the greater surface area at the end interfaces of the dumbbells will allow for a greater and more concentrated build up of magnetic charges. The effect of these charges can be observed as a dipolar interaction which will perturb the magnetization of the hard phase near the end interfaces of the nanocrystals. This study focuses on these phenomena to optimize the performance of nanocomposites by increasing coercivity.

II. Methods

The point dipole experiment was derived numerically from

$$\boldsymbol{B} = \frac{\mu_0}{4\pi}\frac{1}{r^3}[3(\boldsymbol{m} * \hat{r})\hat{r} - \boldsymbol{m}]$$

where $\boldsymbol{m}$ is the magnetic dipole moment, $\hat{r}$ is the radial unit vector, r is the magnitude of the radius of a point from the dipole [4]. The dipole was modelled on the classical model of an electron. The dipole moment caused by the electron spin is expressed as

$$\boldsymbol{m} = -\frac{e}{M}\boldsymbol{S}$$

where e is the electron charge, $\boldsymbol{S}$ is the spin vector, M is the electron mass [5]. A 3-dimensional grid of evenly spaced points was generated in Python. Each of the points was defined in cartesian coordinates and underwent a transformation to spherical coordinates for compatibility. The output was then compared with analytical proofs based on geometrical considerations.

The second experiment studied the precession of the magnetization of a thin film out-of-plane. The experiment was approached by a computational method using Mumax3. Mumax3 is a GPU accelerated

program that is built upon the Landau-Lifshitz-Gilbert equation for the calculation of the precession of the magnetization of finite elements [6]. Simplistic calculations for the stray field were derived from the consideration of a uniformly magnetized ellipsoid. In this case, the ellipsoid takes the form of a thin film with approximately infinite proportions in the x and y directions with respect to the thickness (z-direction). The stray field can be expressed as

$$\boldsymbol{H}_D = 4\pi M_s \sin\theta\, \hat{k}$$

where $4\pi M_s$ is the saturation magnetization in Gauss and θ is the angle between the z component of magnetization to the plane of the film. The initial condition of the thin film has a uniform magnetization in the y-direction. The magnetization is precessed out of the film plane with increasing external field. Multiple experiments were run with varying angles of the external magnetic field to the film plane and different values of saturation magnetization are used.

The experiment was validated by an analytical derivation, approximated numerically. Given the consideration of the Landau-Lifshitz-Gilbert equation

$$\frac{d\boldsymbol{m}}{dt} = \frac{\gamma_{LL}}{1+\alpha^2}\left(\boldsymbol{m} \times \boldsymbol{B}_{eff} + \alpha \boldsymbol{m} \times \left(\boldsymbol{m} \times \boldsymbol{B}_{eff}\right)\right)$$

where γ_{LL} is the gyromagnetic ratio and α is the Gilbert dampening term. For this case, the limit is taken as t→∞, therefore, this lets the time rate of change of the magnetization vector to be zero at each step of increasing external field. This leads to a simple analytical expression to describe the magnetization of the sample. The magnetization vector must be collinear with the effective field

$$\boldsymbol{H}_{eff} = \boldsymbol{H}_{ext} - 4\pi M_s \sin\theta\, \hat{k}.$$

If the external magnetic field is defined by an angle out-of-plane ε, the analytical proof can be expressed as

$$\tan\theta = \frac{H_{ext}\sin\varepsilon - 4\pi M_s \sin\theta}{H_{ext}\cos\varepsilon}.$$

This equation must be solved with numerical approximation. A simple solution is to let $\theta \approx \varepsilon\ for\ H_{ext} \gg 4\pi M_s$.

For the project exploring exchange coupling in nanocomposites, a sample is created with the following parameters: length and width of 160 nm and thickness of 20 nm, soft phase of α-ferrite and hard phase of $Nd_2Fe_{14}B$ with material parameters provided by Wei Li et al. [1]. It should be noted that the $4\pi M_s$ values of these materials are pseudo-realistic to target a large exchange length within the soft phase, see Table 1. The α-ferrite nanocrystals created were targeted to have a radius or thickness less than the exchange length to prevent domain formation [1].

Phase	$4\pi M_s$ (G)	Aex (A/m)	Ku1 (J/m^3)
soft	1700	12.5e-12	4.5e6 $\hat{\boldsymbol{j}}$
hard	10000	25e-12	-0.048e6 $\hat{\boldsymbol{i}}$

Table 1. Material values used in the simulation. Aex and Ku1 sourced from Wei Li et al.

Matrix spacing was varied to determine its effect on the ability for exchange coupling to occur. Patterns of regularly spaced linear and rectangular structures were explored with nanowire, dumbbell, and cubic and spherical quantum dot nanocrystals to determine an optimized array condition.

The appearance of exchange coupling of two-phases of magnetic material must occur within the absence of domains for the nanocrystalline material and when energetically favorable. Therefore, the precession must be favorable within the nanocrystal to break intra-phase exchange interaction in favor of inter-phase exchange. This was observed by unanimous switching during a full magnetization reversal of a sample [1]. The dipolar interaction was caused by un-passivated magnetic charges on the surface of the nanocrystals and presented itself as a two-stage demagnetization process. Mumax3 allows a visual representation to observe the phenomena. The simulation was validated by recreating similar results to those found in literature with nanowires.

III. Results and Discussion

For the simulation of a point dipole, the

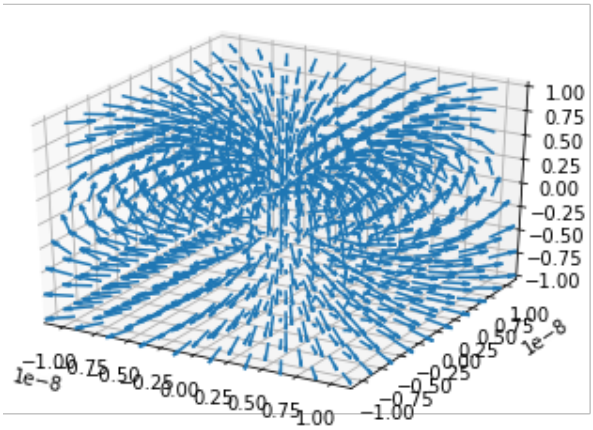

Figure 1. A graphical representation of magnetic field lines caused by an electron dipole moment in real space (meters).

magnetic field followed the pattern of radial planar symmetry with field lines beginning at the positive charge and terminating at the negative magnetic charge, see figure 1.

The validity of the numerical simulation was tested with the following concepts. The first was the geometrical consideration that at a fixed radius r, the magnitude of the generated magnetic field along the z-axis will be twice that of the same radial distance in the x-y plane. The second proof was derived from the concept of a uniformly magnetized

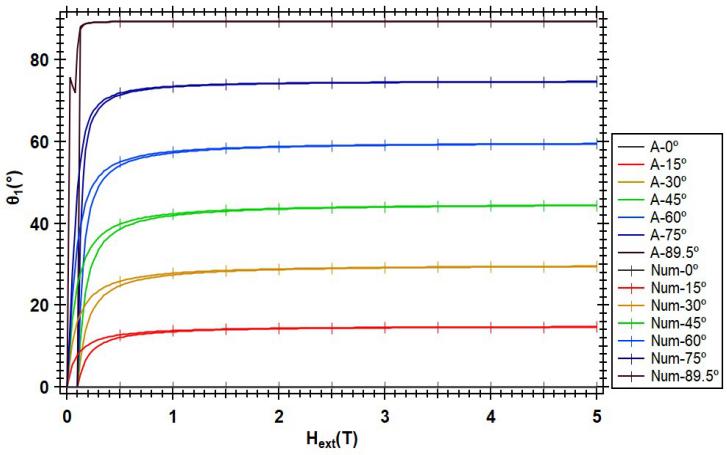

Figure 2. Results of computational simulation and analytical derivation for the out-of-plane magnetization of a thin film. $4\pi M_s$ = 1000G. Crossed lines indicate numerically derived results while straight lines are from Mumax3.

sphere. Along the z-axis, a point on a uniformly magnetized sphere will experience a dipolar magnetic field that is 2/3 the magnitude of the magnetization of a sphere of the same size. In this case, both analytically derived proofs were found to agree with the simulation.

The thin film magnetization experiment resulted in agreement between computational results produced by Mumax3 and the numerically approximated analytical proof. The experiments were varied by 15º intervals, ending at 89.5º to account for calculation error. As predicted, the results agree within the bounds of $H_{ext} \gg 4\pi M_s$, while the analytical derivation deviates significantly outside of that condition. It can be observed in figure 2 that the curves of each method converge as $H_{ext} > 4\pi M_s$, and that the error is significant for $H_{ext} < 4\pi M_s$.

The simulation of two-phase nanocomposites began with an attempt at the validation of the experiment by setting parameters similar to those of Wei Li et al.

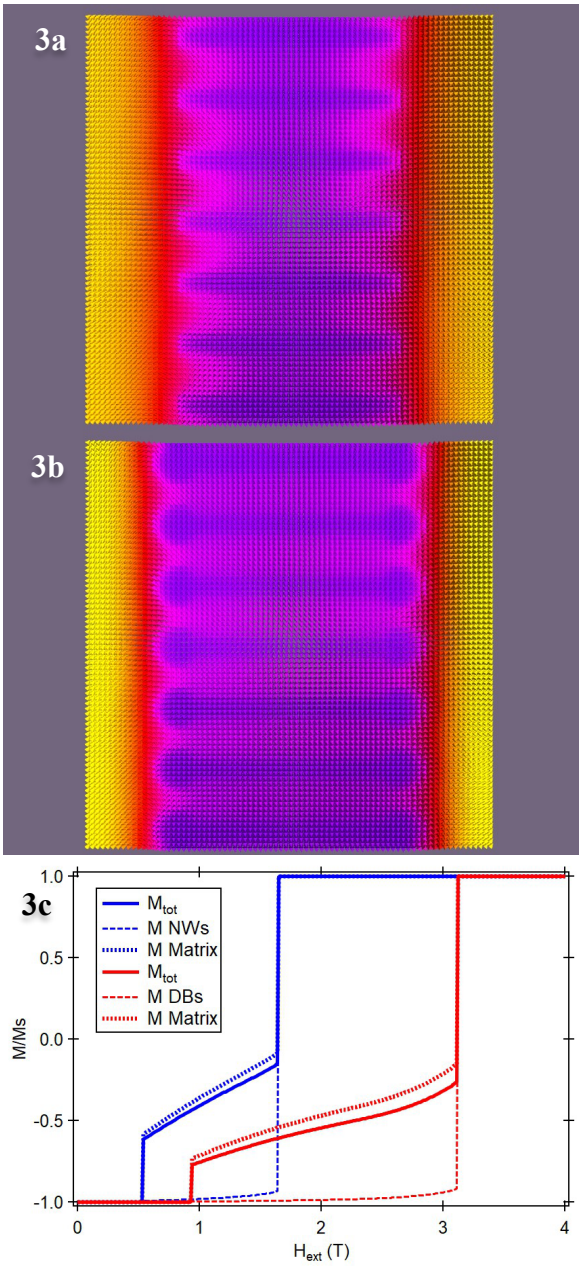

Figure 3 (a-c) a) An array of 90 nm nanowires in a thin film at 1.2 T. b) 90 nm Dumbbells in a thin film at 2.4 T. c) Magnetization reversal curve starting at -1 M/Ms. Components are arranged by contributions of each phase. The blue curve is the nanowire sample, and red is the dumbbell sample.

An array of nanowires of 8 nm in diameter were spaced evenly within a thin film and subjected to a full magnetization reversal as seen in figures 3a-c. A period of the magnetization reversal showing a perturbation of the dipoles at the end interfaces of the nanowires can be observed. This period is dominated by dipolar interaction caused by built up charges on the surfaces of the nanowires. A second stage of the magnetization reversal is abrupt and consists of the switching of the perturbed hard region and the nanocrystals. This unanimous switching is indicative of exchange coupling between the two-phases. Similar effects were observed in this experiment and the literature therefore, the study was then expanded to find optimum geometrical conditions for improving magnetic properties.

The focus shifted towards optimizing the conditions found in the array of nanowires by carefully selecting the geometry of the nanocrystal phase. Dumbbells were selected due to their large surface area at the end interfaces. Various simulations were conducted with dumbbells corresponding to a nanowire length and the coercivity of these

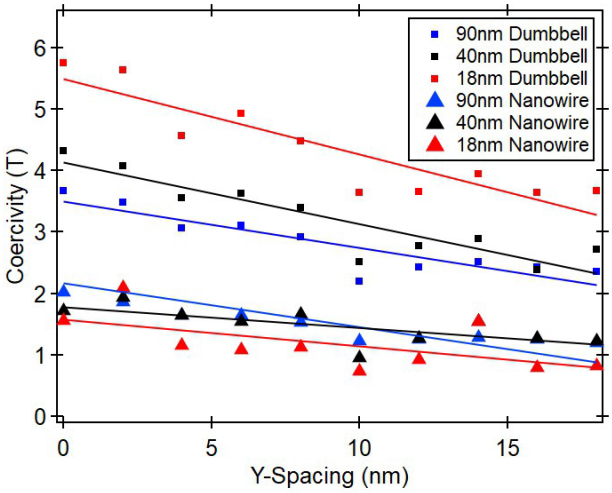

Figure 4. A plot of coercivity varied by spacing of nanowire and dumbbell shaped soft magnetic phase in nanocomposites.

curves are plotted in figure 4. Note the dependence of spacing on the overall coercivity of the sample.

In all cases, dumbbells were shown to improve the coercivity of the nanocomposite. In general, the coercivity is strongest at the tightest spacing of material. This is

interesting due to the balance of multiple effects. The percentage of the sample consisting of the soft phase is maximized with the tightest spacing. Size 40 and 18 nm nanocrystals utilized a 2-dimensional array due to the size of the nanocrystals in relation to the sample size. The exchange coupling between the two-phases is decreased in the case of tighter spacing due to the nearest neighbor being the same phase in two or more locations for every nanocrystal in dumbbells. This would imply that the dipolar interaction dominates for tight spaced nanocrystal array interactions. However, deviations occur due to the space available between the slender portions of the dumbbells. Since this area of the hard phase can interact with the ends of the dumbbells, it can provide exceptions to the dominance of the dipolar interaction.

IV. Conclusions

This paper studied magnetic properties of two-phase nanocomposites by using Mumax3 for micromagnetic simulation. The software was validated by receiving comparable results to literature. The work was expanded upon by taking different geometrical conditions into account for studying the prevalence and dominance of exchange coupling and dipolar interaction, and then studied to find the optimum conditions of coercivity in a nanocomposite sample. Dumbbell nanocrystals were found to optimize exchange coupling in nanocomposites when compared to nanowires of the same size and spacing. Smaller size dumbbells showed a decrease in dipole interaction while maintaining a greater coercivity due to stronger exchange coupling.

It is recommended by the author for further investigation from an experimental basis. It is clear from simulation that geometrical conditions greatly affect the observable magnetic material properties of nanocomposites.

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VI. Footnotes, Endnotes and References

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