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A new approach to include surface contributions in micromagnetic simulations of nanoparticles.

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Abstract

In this work a micromagnetic model is presented for ferromagnetic nanoparticles where the surface is treated as a single effective layer and not as a separate shell. The model consists of two coupled Partial Differential Equations (PDE), one for the magnetization vector of the bulk volume and the second for the outer nodes. The strength of the coupling depends on the effective width of the layer. Simulations were made by means of the Finite Element Method (FEM). For a comparison FEM for core/shell type and atomistic Monte Carlo simulations were also performed. Our results show that H_s, the field where reversal takes place, varies as $\sim 1/D$, where D is the particle's radius, with the anisotropy strength for any anisotropy direction. Moreover the computational cost of the effective layer model is lower than the core shell one, thus can be easily extended to larger particles where dipolar interactions should also be taken into account.

Keywords: Micromagnetism, Surface effects, Finite Element Method, Monte Carlo

1 Introduction

The low temperature magnetic properties of magnetic nanoparticles as the size decreases are governed by surface effects [1].

Surface effects on magnetic nanoparticles have already been studied by means of atomistic Monte Carlo (MC) [1] and continuum micromagnetic approach using core-shell type morphology [2]. Here a micromagnetic model is presented for ferromagnetic nanoparticles where the surface is treated as an effective artificial layer. The model consists of two coupled magnetization vectors for the

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volume and the boundary respectively. The evolution of each of these vectors is described by a partial differential equation (PDE) under the influence of an effective magnetic field, arising from the exchange interactions, the magnetic anisotropy, the Zeeman and dipolar energy [3]. The two vectors interchange fluxes, based on the ratio a / ℓ_x, where a is the effective width of the surface layer, and ℓ_x is the exchange length.

In atomistic Monte Carlo methods, due to their nature, surface effects can be directly studied by examining the magnetic behavior at each lattice points. In the micromagnetic approach, due to continuity requirements, the change in magnetic parameters must be well defined in terms of geometric parameters. Thus, in order to investigate the surface contribution, the valid approach is to construct a core/shell type particle with different properties and demand continuity of the magnetization vector on the interface. We must notice that this requirement is valid in the case of surface effects, and not along the interface between two different materials, where their magnetic moments may be considerably different, as in the case of a bi-magnetic core/shell nanoparticle.

In the absence of dipolar interactions and at low temperatures, the application of an external field may lead only to coherent reversal processes. Thus, it is expected that the surface layer, in the case of a spherical particle, will not have drastic variations with respect to the radial direction. The purpose of this work is to present a micromagnetic model where the surface shell is simulated by a set of Partial Differential Equations (PDE) only for the outer nodes of the nanoparticle, without adding any shell structure surrounding the bulk volume. In these PDE a parameter a is introduced to describe the effective width of the surface layer. In order to investigate the validity of the model, FEM simulations for core/shell type nanoparticles were also performed. Moreover the computational performance of these two models is traced in order to investigate the advantage of the presented model. For a more compressive study MC simulations were also performed.

2 Modeling

Simulations were made for a spherical particle by varying its size (i.e. it's radius R), and the width of the surface layer (i.e. by varying a). The exchange coupling is assumed be the same in both volume and surface. The anisotropy is different for the core and the surface layer in size and type. The variation of the anisotropy can be attributed to the broken symmetry of the surface region. In sections 2.1, 2.2 and 2.3 the FEM core/shell model, the FEM effective layer model and the Monte Carlo core/surface model are presented respectively.

2.1 Finite Element Method (core/shell model)

In a continuum approach the evolution of the magnetization vector $\boldsymbol{M}$ is described by an equation of the form [3]

$$\tau \frac{d\boldsymbol{M}}{dt} = -\frac{1}{M_s^2}\boldsymbol{M}\times\left(\boldsymbol{M}\times\boldsymbol{H}_{\boldsymbol{eff}}\right),$$

where τ is a relaxation time of the system, $\boldsymbol{H}_{\boldsymbol{eff}}$ is an effective magnetic field and M_s the saturation magnetization. The main contributions to the effective field arise from exchange interactions, anisotropy, dipolar and Zeeman energy. For an isotropic and homogeneous material, the exchange field is written as $\vec{H}_{ex} = \ell_x^2\nabla^2\boldsymbol{M}$, where $\ell_x = \sqrt{2A/\mu_o M_s^2}$ is the exchange length and A is the exchange stiffness. The field for uniaxial anisotropy of constant K is written as $\vec{H}_k = \left(2\mathrm{K}/\mu_o M_s^2\right)\left(\hat{k}\bullet\boldsymbol{M}\right)\hat{k}$. The PDE are solved by means of a finite element method with the mean element size smaller than Bloch wall width $\delta = \sqrt{\ell_x/\kappa}$, where κ is the hardness parameter defined as $\kappa^2 = 2\mathrm{K}/\mu_o M_s^2$. In the core/shell morphology the core of the particle is surrounded by a shell of finite width and different properties with respect to the core ones. As already mentioned, the exchange constant is assumed to be constant in the whole particle, but the surface atoms, are subject to different forces arising from the reduced number of neighbors leading to an implicit reduction of ℓ_x through the reduction of *A* because the latter depends on the number of neighbors. Consequently, by assuming an isotropic and homogenous distribution of atoms on the surface the mean value of the number of neighbors over the whole surface drops to half. Thus for the exchange length on the surface $\ell_{xI} = \ell_x/\sqrt{2}$ is used. The surface anisotropy can be directly introduced by changing its magnitude and direction. Thus the equations to be solved are

$$\tau \frac{d\boldsymbol{M}}{dt} = -\frac{1}{M_s^2}\boldsymbol{M}\times\left(\boldsymbol{M}\times\left(\boldsymbol{H}+\ell_x^2\nabla^2\boldsymbol{M}\right)\right)$$

for the core and

$$\tau \frac{d\boldsymbol{M}_{\boldsymbol{I}}}{dt} = -\frac{1}{M_s^2}\boldsymbol{M}_{\boldsymbol{I}}\times\left(\boldsymbol{M}_{\boldsymbol{I}}\times\left(\boldsymbol{H}_{\boldsymbol{I}}+\ell_{xI}^2\nabla^2\boldsymbol{M}_{\boldsymbol{I}}\right)\right)$$

for the shell.

In the above equations $\boldsymbol{H}$ and $\boldsymbol{H}_{\boldsymbol{I}}$ represent the components of the effective field, in the core and surface respectively, not including the exchange. The continuity of the magnetization vector at the interface requirement leads to the equation

$$\left(\ell_x^2 \nabla \boldsymbol{M_i} - \ell_{xI}^2 \nabla \boldsymbol{M_i}\right) \bullet \hat{n} = 0$$

For the anisotropy terms we define two different anisotropy constants K_V for the volume and K_S for the surface. So the set of PDE needs to be solved with varying coefficients in space. This method becomes very expensive when the volume to surface ratio becomes large. In that case in order to reach an acceptable accuracy a very fine discretization is needed, enlarging computational cost.

2.2 Finite Element Method (effective layer model)

In order to reduce the computational cost and enable the simulation of larger particles, we have developed a new model were we treat the surface as an effective layer. In this approach a new set of PDEs is solved only for the surface nodes, introducing an effective width parameter a. The model assumes that the magnetization does not vary drastically along the direction normal to the particle's surface, which is generally true, when the surface extends in a small number of unit cells. This number depends on the wall width δ. The surface layer width must be smaller than δ in order to block the formation of a domain wall. Under these assumptions the equations to be solved are,

$$\tau \frac{d\boldsymbol{M}}{dt} = -\frac{1}{M_s^2} \boldsymbol{M} \times \left(\boldsymbol{M} \times \left(\boldsymbol{H} + \ell_x^2 \nabla^2 \boldsymbol{M} \right) \right)$$

for the volume and

$$\tau \frac{d\boldsymbol{M}}{dt} = -\frac{1}{M_s^2} \boldsymbol{M} \times \left(\boldsymbol{M} \times \left(\boldsymbol{H_I} + \ell_{xI}^2 \nabla_T^2 \boldsymbol{M} \right) \right) + \frac{\ell_x^2}{a} \frac{\partial \boldsymbol{M}}{\partial n}$$

for the surface nodes. The index I has been omitted to emphasize the fact that in this model there are not two different volumes, but two different equations for different nodes.

∇_T^2 represents the tangent Laplacian on the boundary. The last term arises from the splitting of the complete Laplacian to a tangent and a normal part to the surface and it represents the flux interchange between the core and the surface layer of the particle.

2.3 Monte Carlo (core/surface model)

Atomistic MC simulations with the implementation of the Metropolis algorithm were performed for spherical nanoparticles interacting with Heisenberg exchange interactions as described in ref. [1]. The nanoparticles have total radius up to 40 lattice spacings and surface thickness of 2 lattice spacings, at $T = 0.01J$. The core is assumed to have uniaxial anisotropy along the z-axis, while the surface layer has different type and strength of anisotropy.

3 Results

In all of the simulations the particle is initially at the saturated state, and the hysteresis loop is calculated by reducing the external field until negative saturation is reached. Then the opposite procedure is followed. As mentioned in section 2.2 the validity of the effective layer model is constrained by the width of the surface layer. Thus, is really important to compare the two micromagnetic models for small particles. So the first goal of this work is to investigate the range of the surface layer width where the proposed model leads to reliable results. For this purpose, simulation based on the effective layer model (section 2.2) and the core-shell model (section 2.1), were performed for a spherical particle having uniaxial anisotropy and constant size $R/\ell_x = 1$. The variable of these simulations was the width of the surface layer. Two cases were studied, one with surface anisotropy $K_s = 0.05\mu_0 M_s^2$ and the second with surface anisotropy $K_s = 0.25\mu_0 M_s^2$. In both cases the core anisotropy is $K_V = 0.1\mu_0 M_s^2$. Figure 1 shows calculated hysteresis loops by the two methods .The reversal mode for the selected particle size is coherent rotation for all the values of the width of the surface layer studied. Figure 2 shows the variation of the H_s field with respect to the width of the surface layer for the FEM models measured by the ratio a/ℓ_x. As it can be seen from figures 2a, 2b both models tend to values close to the anisotropy field of the core $2K_V / \mu_0 M_s$ for small values of the width of the surface layer and to values close to the anisotropy field of the surface layer $2K_s / \mu_0 M_s$ for large values of as expected from ref. [1] but with different trends. The convergence of the two methods to the same values (fig. 2) and the same reversal properties (fig. 1) is a strong indication that both methods lead to the same results when the width of the surface layer is smaller than $a < \ell_x/10$ independently of the anisotropy constant of the surface layer.

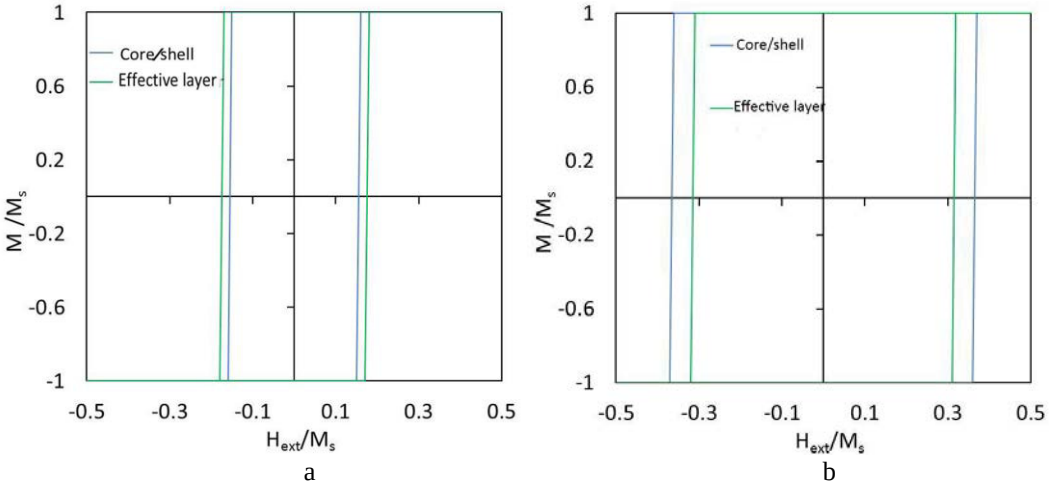

Figure 1. Hysteresis loops for $R/\ell_x = 1$.The external field and anisotropy are assumed to be at the z direction. Both surface (shell) and core have uniaxial anisotropy, with a) $K_V = 0.1\mu_o M_s^2$ and $K_S = 0.05\mu_o M_s^2$ .b) $K_V = 0.1\mu_o M_s^2$ and $K_S = 0.25\mu_o M_s^2$.

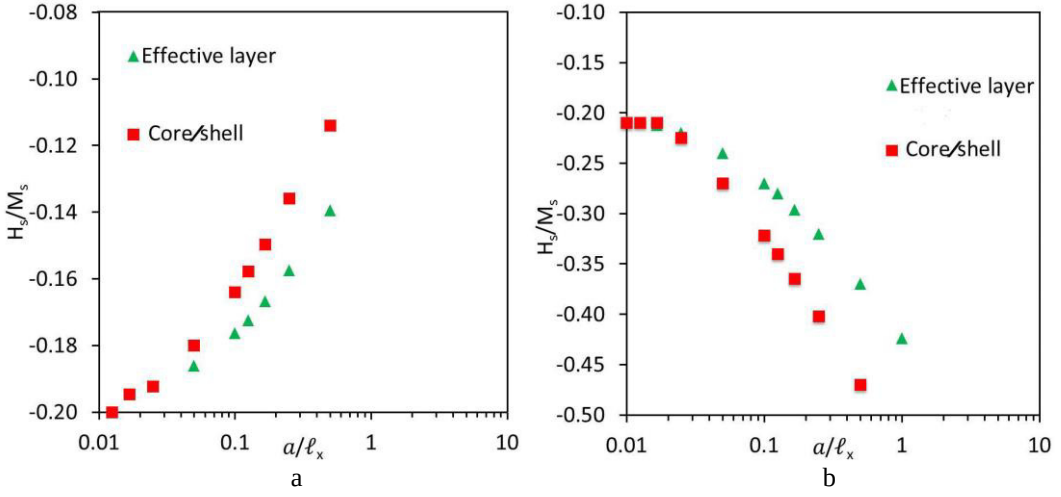

Figure 2 H_s vs a/ℓ_x for $R/\ell_x = 1$. Both surface(shell) and core have uniaxial anisotropy, with a) $K_V = 0.1\mu_o M_s^2$ and $K_S = 0.05\mu_o M_s^2$ .b) $K_S = 0.25\mu_o M_s^2$, and $K_S = 0.25\mu_o M_s^2$.

Choosing the value $a = \ell_x/10$ for the width of the surface layer, simulations were performed by varying the size of the particle in order to investigate the surface contribution to the reversal process. Figure 3 shows the variation of H_s field with respect to R/ℓ_x for the two models for $K_v/K_s < 1$

(Fig. 3a) and $K_v/K_s > 1$ (Fig. 3b). The change of the monotonous behavior close to $3.5\ell_x$ in both cases indicates the change of the reversal mechanism from coherent rotation to incoherent curling [4]. Generally the two methods are in good agreement, for the whole size range we studied.

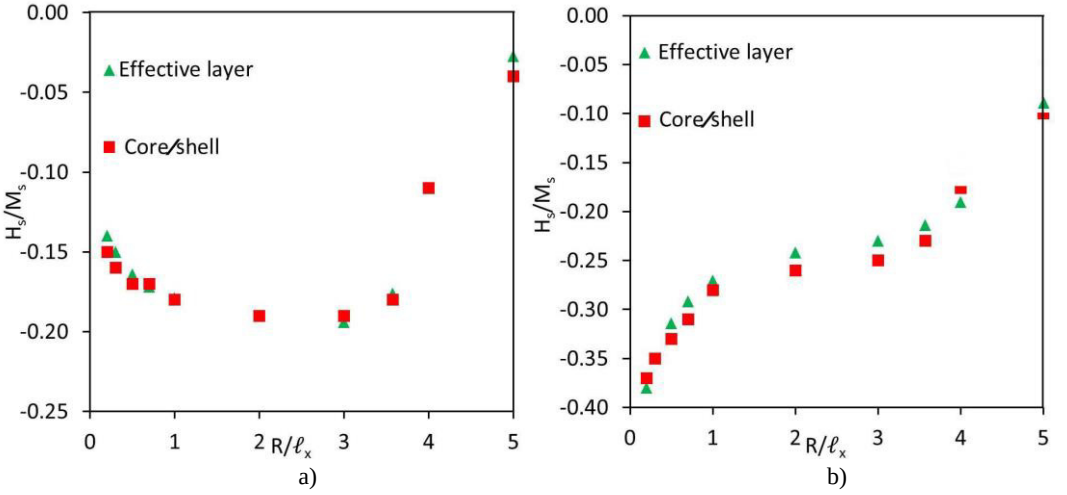

Figure 3. H_s vs R/ℓ_x for $a/\ell_x = 0.1$. Both surface (shell) and core have uniaxial anisotropy, with a $K_V = 0.1\mu_o M_s^2$ $K_S = 0.05\mu_o M_s^2$, b) $K_V = 0.1\mu_o M_s^2$ $K_S = 0.25\mu_o M_s^2$

Finally Monte Carlo simulations were also performed, up to the estimated limit of coherent rotation reversal (Fig. 4). In figure 4c surface anisotropy direction is normal to the particle's surface. Due to the fact that the micromagnetic models are continuum ones whereas the MC is clearly atomistic there is no direct bridging between their associated characteristic lengths in our models. Thus figure 4 is presented with two length scales, one of R/ℓ_x for the continuum models, and D_{MC} representing the diameter of the particle in lattice spacings used in MC methods. All methods indicate dependence analogous to $1/R$, for the H_s field, where R is the radius of the particle [1]. Atomic structure effects are not directly taken into account in continuum approximation, but only as a mean value approach by changing the exchange length, in contrast to atomistic MC where structure effects are directly taken into account. Decreasing the size of the particle uncompensated surface spins arise [1] and clearly affect the reversal process. In the case of higher surface anisotropy convergence with MC is reached for small radii indicating the fact the higher anisotropy constant may wipe out the effect of uncompensated spins.

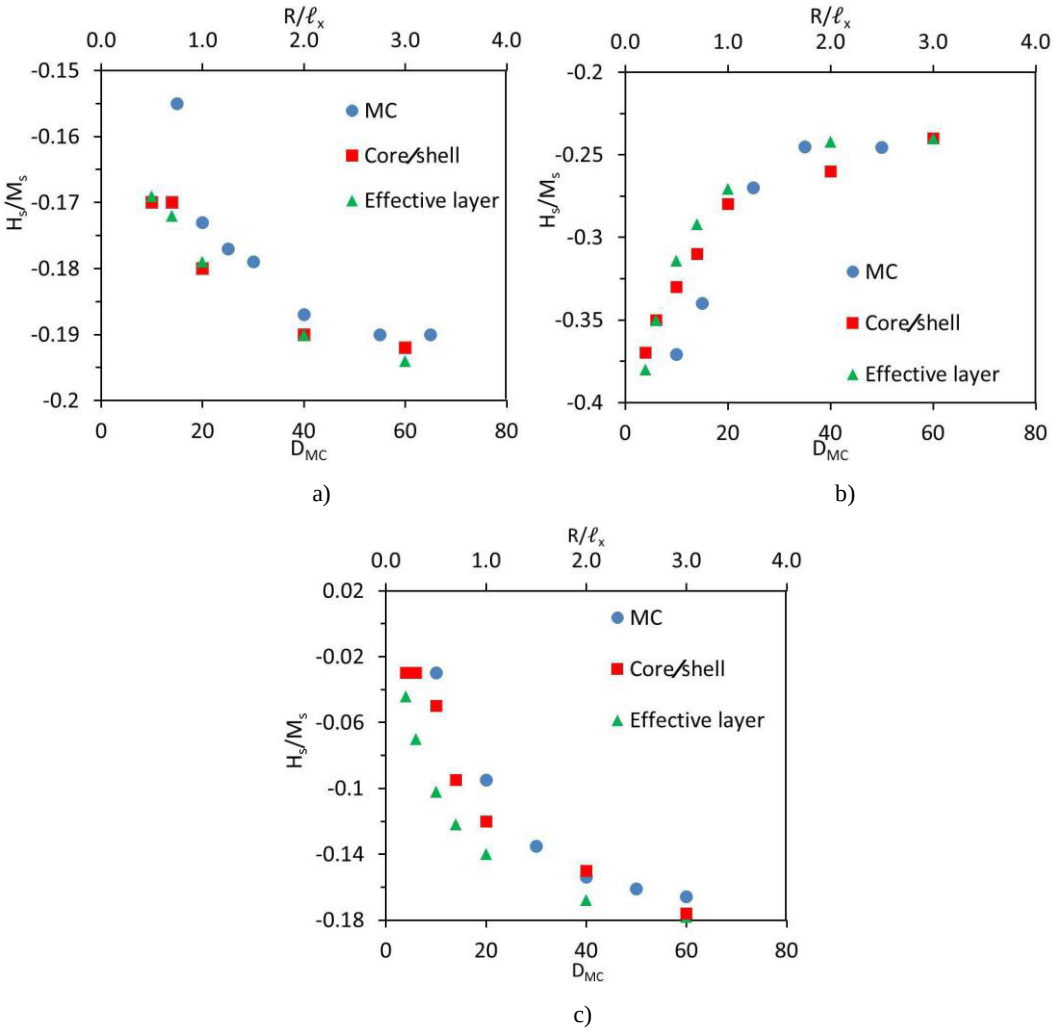

Figure 4. H_s vs R/ℓ_x for $a/\ell_x = 0.1$ a) $K_V = 0.1\mu_o M_s^2$ and $K_S = 0.05\mu_o M_s^2$, b) $K_V = 0.1\mu_o M_s^2$ and $K_S = 0.25\mu_o M_s^2$, surface (shell) and core have uniaxial anisotropy along the z-axis . c) $K_V = 0.1\mu_o M_s^2$ and $K_S = 0.1\mu_o M_s^2$. Core has uniaxial anisotropy along the z-axis and surface (shell) has radial anisotropy direction. D_{MC} represents the diameter of the particle, in lattice spacings, for MC simulations.

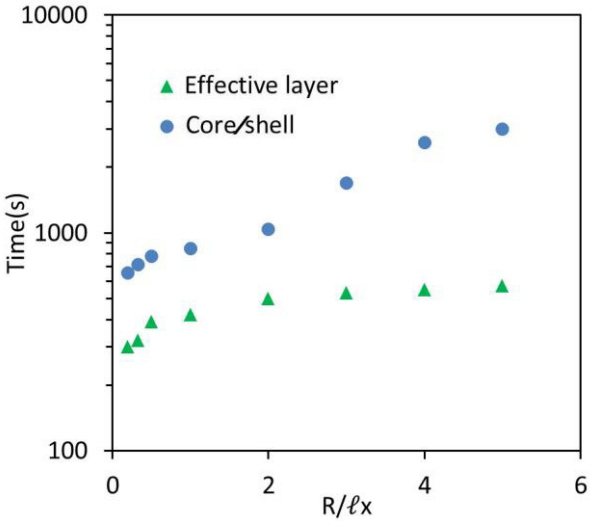

Fig.5 Computational time vs particle' size for the two continuum models.

4 Conclusions

In this work a continuous micromagnetic model for ferromagnetic particles that takes into account surface contributions is presented. Simulations based on this model lead to proper results for the magnetization behavior with respect to the core/shell type morphology and the atomistic MC method, when the surface layer width is smaller than or close to the one tenth of ℓ_x .Treating the surface only by the surface nodes of the core and not as volume nodes of a separate shell leads to a managed increment of computational cost making our method capable for simulations especially at large particles where dipolar interactions have to be taken into account (fig. 5).

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