Dipolar Ordering in Assemblies of Nanomagnets

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In this work, we have set up a computer code to simulate the magnetic equilibrium state of magnetic nanostrutures with different shapes. Dipolar interactions among the magnetic moments have been taken into account and an efficient algorithm to compute the fields during the Monte Carlo simulation has been devised. We have used the code to check the limits of validity of the macrospin approximation for the dipolar fields and we have applied it to study the ground states of assemblies of nanostructures. Finally, we show that simulations with atomistic detail are able to reproduce the known magnetic states in disc-shaped elements and find that the magnetic states of cubic nanoparticles can be tuned by their size in good agreement with experimental observations.

I. INTRODUCTION

During the last decades there has been a renewed interest in the physics of magnetic materials driven by the need to control the magnetism of magnetic structures at submicrometric length scales in order to use them for technological applications. Reducing the size of magnetic structures to the nanoscale has been possible due to the appearance of new fabrication techniques that allow to synthesize nanosized magnetic elements with specific compositions and well controlled sizes and shapes. Thus, Nanomagnetism has emerged as a powerful field of research that has helped to develop a variety of technological and biomedical applications for magnetic systems that range from assemblies of single domain nanoparticles to patterned arrays of nanostructures[1,2].

To describe nanomagnets, two different levels of description are usually employed depending on the properties to be studied and the number of magnetic entities involved. One is micromagnetics, which describes the material in the continuum approximation through the spatial distribution of magnetization in it. This kind of approach is also used to simulate the properties of structures with sizes of the order of tenths of nanometers, when the material is discretized in finite elements that behave as a uniform magnetic moment. The other approach, which will be adopted here when studying individual, is atomistic in the sense that magnetic moments and positions of the magnetic ions as well as the interactions among them are considered. However, when considering assemblies of nanomagnets with long-range interactions, atomistic details cannot be kept since computation time become prohibitive. Then, each magnetic element is replaced by a macrospin with magnetic moment equal to the total magnetization of the element.

The objective of the present work is to build up a computer code that is able to find equilibrium and recreate the dipolar ordering of a given system, taking into account its finite size. With this program we can simulate the ordering in an assembly of nanomagnets, treating each one as a macrospin and ignoring its structure. Adding the exchange interaction to the code we can also simulate the internal structure of a nanomagnet. Using this feature we can explore the possible ground states of certain magnetic systems.

The main interaction responsible for the long-range order in an assembly of magnetic dipoles is the dipolar interaction that is given by the following expression

$$E_{dip} = -\sum_i \vec{\mu}_i \cdot \vec{H}_{dip,i} \quad , \tag{1}$$

where $\vec{H}_{dip,i}$ is the dipolar field generated on the magnetic moment $\vec{\mu}_i$ placed at $\vec{r}_i$ by the rest of magnetic moments $\vec{\mu}_j$:

$$\vec{H}_{dip,i} = -\frac{\mu_0}{4\pi}\sum_j \left[\frac{\vec{\mu}_j}{r_{ij}^3} - 3\frac{\vec{r}_{ij}(\vec{\mu}_j \cdot \vec{r}_{ij})}{r_{ij}^5}\right] \quad , \tag{2}$$

where $\vec{r}_{ij}$ is the vector joining sites i and j.

The dipolar field lines created by a single dipole are shown in Fig.1(a). As can be seen in the figure, if we place a second dipole close to the first, different orderings of the second dipole are favored depending on its position. The preferred orientation is antiferromagnetic (AF) if the angle with respect to the direction of the first dipole is bigger than $\theta^* = 54.74°$, whereas ferromagnetic (FM) alignment is favored for smaller angles. This means that dipolar interactions favor FM alignment if dipoles are placed head to head and AF alignment if their are placed side by side.

With that behavior in mind, one can infer qualitatively the magnetic configuration of an infinite lattice of magnetic dipoles. If the lattice is simple enough, we can induce the ordering of the whole lattice by looking at the position of the nearest neighbor dipoles in the unit cell. This is the case of the triangular and square 2D lattices. Looking at Fig.1(b,c), we see how a triangular arrangement of nearest neighbor dipoles favors FM alignment while a square arrangement favors AF one. Extending this behavior to an infinite lattice of dipoles, we infer a FM order for the triangular lattice and a perfect AF columnar order for the square lattice.

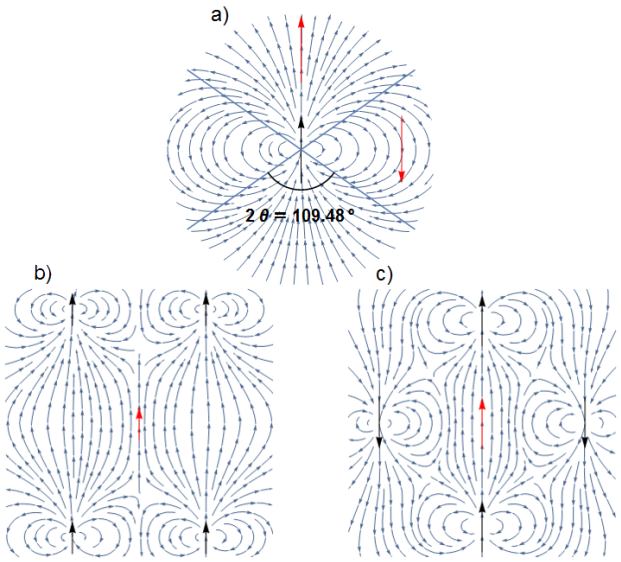

FIG. 1: Representations of dipolar fields. The black arrows show the direction and position of the dipoles that create the fields. The red ones show the direction in which the energy is minimized. In a) is shown the field created by a sole dipole and the regions where the AF or FM orientations are more stable. In b) is depicted the field created by the near neighbors in the triangular lattice. In c) is depicted the field created by the near neighbors in the square lattice.

For the case of 3D lattices, a theoretical analysis by a method first introduced by Luttinger and Tisza [4] shows for example that the ground state of SC dipolar lattices is AF and that for FCC and BCC lattices is FM.

II. CODE DESCRIPTION

The code, developed in FORTRAN 95, computes the ground state of a distribution of magnetic dipolar moments with the same magnitude $\|\vec{\mu}\|$. This dipoles can either symbolize the macrospin of a nanomagnet or the spin of an atom, whether or not the exchange interaction is added. The program is able to create a 3D lattice of dipoles, either SC, BCC, FCC or stacked 2D triangular, filling a cuboid, sphere or disc of given dimensions and computes their ground state. This state is found applying the Monte Carlo method (MC) which also gives us a way to compute it's thermodynamic quantities, aside from it's 3D arrangement.

All the quantities used through the calculations are dimensionless. The distances are expressed as: $\vec{r} = h\hat{a_1} + l\hat{a_2} + k\hat{a_3}$, where h, l and k are integers and a_i are the unit vectors of the lattice. This makes the characteristic length of the lattice, a, as the unit of length. The dipoles are expressed as: $\vec{\mu} = \hat{S}$, where $\hat{S}$ is a unit vector with the same orientation as the dipole, making $\|\vec{\mu}\|$ the unit of magnetic moment. We also take the permeability as: $\mu_o = 4\pi$, making the field, energy and temperature unit as: $\frac{\mu_o\|\vec{\mu}\|}{4\pi a^3}$, $\frac{\mu_o\|\vec{\mu}\|^2}{4\pi a^3}$ and $\frac{\mu_o\|\vec{\mu}\|^2}{4\pi k_b a^3}$ respectively.

A. Dipolar Interaction

One can express the dipolar field (2), in the units of the program, as follows:

$$H^{\alpha}_{dip,i} = -\sum_{j}^{N}\sum_{\beta}^{3} W^{\alpha\beta}_{ij} S^{\beta}_{j}, \tag{3}$$

Where the matrices $W^{\alpha\beta}$, named dipolar matrices, can be written as:

$$W^{\alpha\beta}_{ij} = \frac{\delta_{\alpha\beta}}{r^3_{ij}} - 3\frac{\delta_{\alpha\gamma}\delta_{\beta\nu}r^{\gamma}_{ij}r^{\nu}_{ij}}{r^5_{ij}}, \tag{4}$$

In fact, for symmetry reasons, there are only 6 independent matrices as $W^{\alpha\beta} = W^{\beta\alpha}$. Since the positions of the dipoles are fixed, these 6 matrix are constant and can be computed and stored at the beginning of the simulation if enough memory. This makes them useful to compute the dipolar fields, avoiding the need to run through the array of positions and recalculate the position dependent terms each time a dipole orientation is changed by the MC simulation.

B. Exchange

When we consider the dipoles as atomic magnetic moments we have to add the exchange energy to the energy of the system. This energy, similarly to Eq. 1, can be written in our units as:

$$E_{dip} = -J\sum_{i} \vec{S}_i \cdot \vec{H}_{ex,i} \quad , \tag{5}$$

Where J is the given exchange constant and $\vec{H}_{ex,i}$ is the sum of the dipolar moment of the near neighbors of the dipole at the position i:

$$\vec{H}_{ex,i} = \sum_{j\in nn_i} \vec{S}_j \tag{6}$$

We store the sets of the nearest neighbors of each dipole, nn_i. This way we prevent the necessity to check whether or not a dipole is a neighbor of another one whenever a dipole is reoriented.

C. Monte Carlo Method

The ground state of our system is found thanks to the MC method. This process proposes a random change in the system and calculates the Boltzmann probability of said change: $p = \exp -\frac{\Delta E}{T}$ where ΔE is the increment of energy of the system caused by the change. Then checks

whether or not to keep the new state of the system by drawing a random number $a \in U(0,1)$ and comparing it with the probability p. If $a < p$ the change is accepted and the system is actualized. Repeating this process until, with a large number of tries, the acceptance rate is small enough manages to relax the system to its ground state at the temperature T.

The change in the system proposed by our code is a new random direction of a random dipole. Said new direction is in a cone of a given aperture along the initial direction of the dipole. With the initial and previous orientations we add up the increments in energy:

$$\begin{array}{c} \Delta E = \Delta E_{dip} + \Delta E_{ex} \\ \Delta E_{dip} = -2(\vec{S}_i^f - \vec{S}_i^o) \cdot \vec{H}_{dip,i} \ , \\ \Delta E_{ex} = -2J(\vec{S}_i^f - \vec{S}_i^o) \cdot \vec{H}_{ex,i} \end{array} \tag{7}$$

If the change is accepted the fields must be recomputed:

$$\begin{array}{c} \vec{H}_{dip,i}^f = \vec{H}_{dip,i}^o - W_{ij} \cdot (\vec{S}_j^f - \vec{S}_j^o) \\ \vec{H}_{ex,i}^f = \vec{H}_{ex,i}^o + (\vec{S}_j^f - \vec{S}_j^o) \ \ \forall i \in nn_j \end{array} \ , \tag{8}$$

This shows how important are the algorithms that store the position dependent terms of the fields. For each temperature, more than 10^4 MC steps (number of tries equal to the number of dipoles in the system) are done, yielding a massive number of updates of the fields.

Once the system reaches the ground state at a certain temperature, the algorithm adds up and averages over certain number of MC steps the magnetization, $< m >$, the internal energy, $< e >$, and the specific heat, $< c >= \frac{<e^2> - <e>^2}{T^2}$, per particle.

Looking at the Boltzmann probability it can be seen that it is smaller as the temperature decreases. This causes that to find the lowest energy state of a system at lower temperatures, more tries are needed. To solve this, we use the method known as Simulated Annealing. We find the ground state at a high temperature and use this new found state as the initial condition of a lower temperature. As this new initial condition is closer to the lowest energy state, ΔE is smaller. This increases the Boltzmann probability, also increasing the number of acceptances. Repeating this process makes the system approach the lowest energy state at the wanted temperature, removing gradually the thermal noise and keeping it away of a possible metastable state.

In our program, given the initial and final temperatures, we compute the intermediate temperatures such that they follow: $T_i = T_o \epsilon^i, \ \epsilon < 1$. This is done so that the temperatures are distributed more densely near T_f, due to the higher steps needed in lower temperatures to reach the ground state.

III. RESULTS

Before performing any simulation, we have first made some calculations to check the limits of application of the macrospin approximation and its accuracy as compared with the full atomistic description of a magnetic nanostructure. We have then applied it to find the ground state of 2D assemblies of dipolar interacting nanomagnets, that can be though as small nanoparticles or nanoelements synthesized with a specific shape and size for the purpose of a particular application. Finally, in order to simulate magnetic elements with sizes of the order of tenths of nanometers with atomistic detail, we will argue that previously introduced scaling techniques allow to reduce the sizes so that simulation of magnetic states of disk and cube-shape nanomagnets can be performed using nowadays computer capabilities within a moderate computing time.

A. Macro-Spin Approximation

Using a module of the code that computes dipolar fields independently of the subsequent MC part, we can compute dipolar fields generated by the atomic spins forming a nanomagnet at different points outside the element as a function of the distance to its center. Then, we can compare them with dipolar fields that would be generated by placing a macrospin with magnitude equal to the sum of the atomic spins that form the nanomagnet. This can be done for different element shapes and spin lattices. In this way, we can study under which conditions the macrospin approximation is valid when it comes to describe a nanomagnet.

As an example of such calculations, in Fig.2, we show relative difference between the field created by the nanomagnet, $\vec{H}_{NM}(\vec{r})$, and the macrospin, $\vec{H}_{MS}(\vec{r})$, as a function of the distance to the element center: $\frac{\left|\vec{H}_{MS}(\vec{r}) - \vec{H}_{NM}(\vec{r})\right|}{\left|\vec{H}_{MS}(\vec{r})\right|}$. This is shown along the different directions indicated in the legend of the figure for a cubic shaped nanomagnet having 50 spins per side (125000 spins in total) all oriented along the z axis.

We see that the dipolar field differences decrease with distance and that discrepancies are more important along the 001 direction. For distances of about 120 lattice constants, the relative differences in dipolar fields using both approaches has fallen below 1% . This shows that the macrospin approximation can be faithfully used to describe the magnetic order of an assembly of nanomagnets if they are separated more than two particle sizes. This result is in agreement with similar calculations performed by Politi and Pini[3].

B. Equilibrium configurations

In this subsection, we present the results of simulations for the ground states of assemblies of dipolar nanomagnets in the macrospin approximation and individual nanoelements with atomistic detail and both exchange and dipolar interactions taken into account.

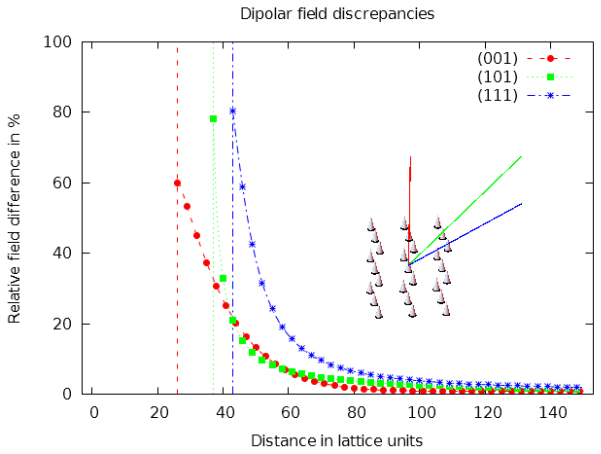

FIG. 2: The relative difference between the dipolar fields created by a cubic nanomagnet and a single macrospin with the same total magnetic moment as the total element but placed at the center of it, is shown as a function of the distance to the center of the nanomagnet along the three directions displayed in the legend. Vertical lines mark the limits of the nanomagnet. The inset shows the three directions along which the differences are calculated.

1. *Only dipolar interaction*

We start by describing the simulations of a lattice of dipoles only interacting through dipolar interaction. This system describes a assembly of identical nanomagnets where their magnetic moment are approximated by macrospins of magnitude $\mu = M \cdot V$ where M is the magnetization of the nanomagnets and V their volume.

The assemblies studied are 2D systems of discs and rectangles in triangular and square lattices. We know that an infinite triangular lattice of dipoles has FM order and that a square one orders in a perfect columnar order. This is no longer true in finite lattices. In this case, it might become favorable to orient the spins in non collinear configurations such dipolar field lines tend to close over themselves, at the expense of an increase in the dipolar energy. With the code we can compute the effects of the finite size of the system and see how it affects the preferred orientation of the lattice.

Looking at Fig.3(a), we see how the FM orientation is favored locally, in accordance with its lattice. Its global orientation, however, is a vortex state. In Fig.3(b) we see how a columnar AF orientation is established in the two symmetry axes of the square, creating loops formed by four dipoles.

We then simulated the states found experimentally by M. Varón *et al.*5. This work provides evidence of the appearance of dipolar domain walls in elongated assemblies of nanomagnets. In Fig.3(c) the FM state is shown by a rectangle of 4x18 dipoles and in Fig.3(d) a longitudinal domain wall is clearly visible for a 6x18 rectangle. We extract the same results, there is an appearance of a domain wall for systems with sufficiently wide thickness.

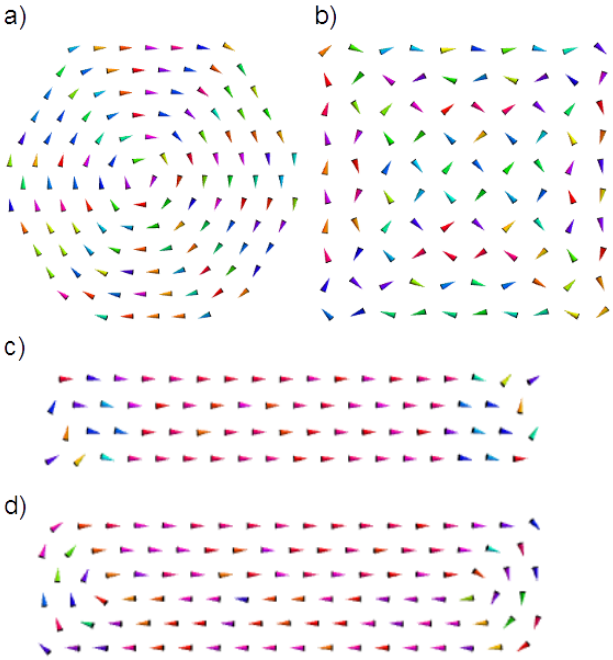

FIG. 3: Figure that shows a 3D representation of the ground state of four different assemblies of nanomagnets. a) depicts a disc with triangular lattice. b) does so for a square with square lattice. In c) and d) is shown the effect of varying width in a rectangle with triangular lattice.

2. *Exchange and dipolar interactions*

Vortex configurations of the magnetization in disc-shaped nanomagnets have been experimentally measured[6]. These vortex configurations consist in a in-plane curled spin configuration with a core in the center where the spins are out of plane. Besides vortex configurations, disc-shaped nanomagnets can exhibit ferromagnetic states. A phase diagram in diameter and thickness between the two states was experimentally determined by R. P. Cowburn *et al.*[6].

The simulation of real nanomagnetic discs is impractical due to computation time. To solve this we adopt the scaling method introduced by J. dAlbuquerque e Castro *et al.* [7]. This was first developed for micromagnetics, but E. A. Velásquez [8] proposed it's application in atomistic models. The method replaces the exchange constant, J, by $J' = xJ$, where x is the scaling factor. This procedure allows to recreate the possible configurations in smaller systems.

Instead of fixing J' and proceed to vary the dimensions of the system, we fix the dimensions of the system and make simulations for a range of J previously estimated thanks to the values found in [7].

Fig.4 shows two states of a disc with SC lattice. In Fig.4(a) we see the vortex state with a core of out of

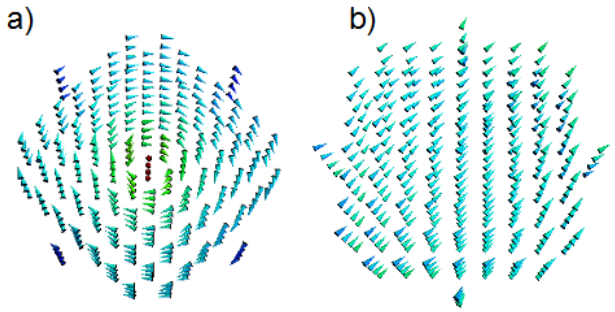

FIG. 4: Figure that shows a 3D representation of the ground state of two discs with SC lattice, 4 nodes of height and 11 nodes of diameter. a) shows the vortex configuration at $J = 5.50$. b) shows ferromagnetic configuration at $J = 8.00$. The colors indicate the angle of the spin towards the z axis.

plane magnetization. And Fig.4(b) a full FM in plane state. We find vortex configurations for the lower values of J and FM configurations for the greater values, as expected.

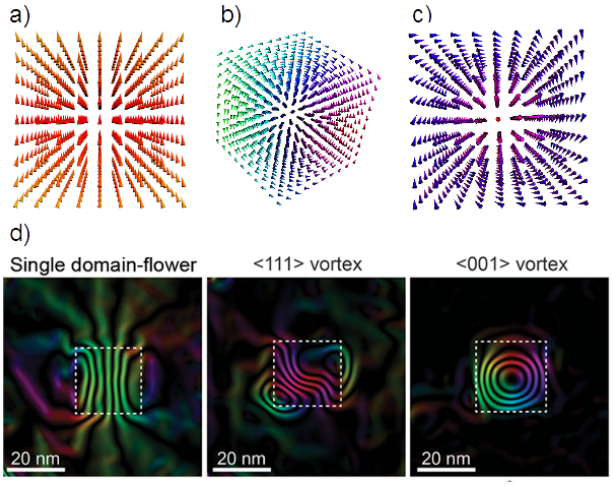

FIG. 5: Figure that shows a 3D representation of the simulations of a cube with SC lattice and the experimental results found by C. Gatel *et al.*[9]. In a) we see a flower configuration at $J = 29.5$ and in c) we see a vortex configuration with $J = 20.0$. b) depicts the (111) vortex configuration found as a metastable state. d) depicts the measured magnetic induction field lines in [9]. The colors indicate the angle of the spin towards the z axis.

We proceed to simulate cube-shaped particles with different scaling parameters. We found that there are two different ground states in this type of systems: a FM state, and a vortex state. Aside from the two ground states, we found a metastable state which presents a vortex along a diagonal of the cube. This metastable state disappears from the simulations when the number of MC steps is increased. However, C. Gatel *et al.*[9] found experimental evidence of the existence of these three states. This indicates that the scaling method could be applied to the study of the phase diagram of nanomagnetic cubes. A simulation of the three states and the experimental results found by C. Gatel *et al.*[9] are shown in Fig.5.

IV. CONCLUSIONS

We have presented the basic structure of a computer code to perform Monte Carlo simulations of assemblies of magnetic nanostructures with different shapes and lattice structures. We have used dipolar matrices to compute the dipolar fields and introduced the appropriate algorithms in order to speed up calculations and making them feasible at the atomistic level. The code has been used to check the limits of validity of the macrospin approximation, that we have been applied to obtain the equilibrium magnetic configurations of assemblies of dipolar interacting structures with different shapes and spatial order. Finally, using the atomistic approach, we have shown that, by applying scaling techniques, the vortex to in-plane transition in a disk-shaped element can be reproduced, and we have been able to reproduce the changes in magnetic order of a cubic FM particle with increasing size observed experimentally.

Acknowledgments

Thanks to Òscar Iglesias for his patience and his guidance through the nanomagnetism and the sea of code. Thanks to my friends for listening me ramble about each new thing I learned. And thanks to my family for enduring my rants every time the code could not compile. We also acknowledge computer time provided by CESCA-CSUC supercomputer facilities.

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