Equilibrium configurations in magnetic nanotubes

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Abstract: We have studied the magnetic low temperature configurations of nanotubes made of spins with exchange (J) and dipolar interactions (D). For this purpose, we have set up a self-made computer code capable of generating hollow tubes by folding a planar regular lattice at any desired angle and controlling its radius and length. The results of the Monte Carlo (MC) simulations have allowed us to unveil different kinds of magnetic order depending on $\gamma = D/J$. Peculiar helicoidal states are formed in a range of γ that depend on the geometric parameters of the tube. With the addition of magnetic anisotropy, a rich variety of new states with radial magnetization components appear.

I. INTRODUCTION

Shrinking a tube up to a nanometric scale has its importance in how the topology and the size-scaled effects are present [1]. The study of magnetic tubular nanostructures has grown of importance the last years due to its properties in the surface, which can lead on a significant improvement in existing applications in fields such as biomedicine [2], magnetic sensors or storage devices [3].

A better comprehension of the resultant magnetic configurations at low temperatures and the properties of antiferromagnetic materials have lead the interest in this study. Whereas the synthesis of 0D, 1D, and 2D nanomagnetic systems has become a mature technology, e.g., via lithography or thin film deposition techniques [4], in the last years, the technique of making 3D tubular nanostructures has been enhanced with a good control of its geometrical parameters[5].

The single-walled nanotube is constructed from a two-dimensional lattice, choosing the length of basis vectors a_1, a_2 and the angle in degrees between them creating a squared lattice like silicon nanotubes[5] . The direction in which spins will be placed, is important, since magnetical properties are dependant upon the topology of the tube. These spins could be represented by a magnetic momentum of single atoms, or molecular clusters characterized with a macrospin as an effective spin in the lattice.

The main structure worked with is the zig-zag (ZZ) topology (translating the vector 45° from its original position). The program is managed to create any topology, just by changing m and n indexes, curving the surface with adjacent angles and distances between positions.

$$C_h = a_1 m + a_2 n, \tag{1}$$

$$\cos\theta = \frac{a_1 m}{\sqrt{a_1^2 m^2 + a_2^2 n^2}}, r = \frac{\sqrt{a_1^2 m^2 + a_2^2 n^2}}{2\pi} \tag{2}$$

When $m = 0$, it is equivalent to folding the sheet creating a tube of pilled up spins. Otherwise, when both indexes are equaled $m = n$, an adjacent angle of $\pi/4$ is obtained, called zig-zag topology. If $0 < m < n$, it is termed as chiral. Once the geometry is modeled, is time to add peculiar parameters of the material it is about to study, seeking for the equilibrium states resulting from the competence between exchange short range interaction and dipolar long range interaction and finally figuring out how the anisotropy plays a roll in this competition between parameters.

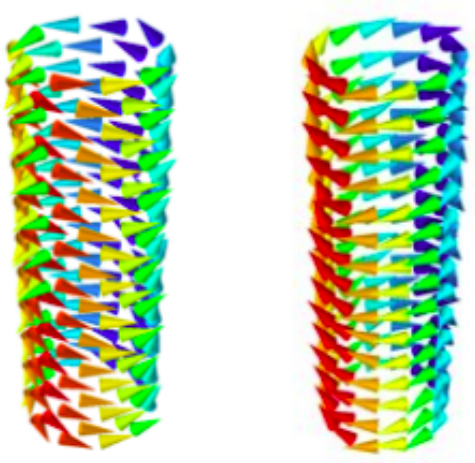

FIG. 1: Snapshots of the two different nanotube lattice arrangements studied in this work: zig-zag (ZZ) on the left and aligned stacked up rings (AA). Both are in a vortex configuration state.

With a proper implementation of standard Metropolis MC simulation one can check which is the equilibrium state with the parameters input by the user. In different circumstances, could be obtained a ferromagnetic phase, where all the magnetic moments are aligned, or may be get vortex states, where the net magnetic value is null. Another states in between could appear, an helical one or we could even attain mix states. But also, the nanotube can reach an antiferromagnetic state as will be present

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in the next sections.

Analyzing the thermodynamics of the system, one can get the expected values of observables, as arithmetic averages. With calculations on response functions like specific heat and susceptibility, as fluctuations of energy and the parameter order respectively, certain conclusions will be drawn therefrom.

II. MODEL AND COMPUTATIONAL DETAILS

Recalling on statistical physics, for determining the energy or the magnetization of a system ruled by a Hamiltonian, we need the calculation of probabilities on a canonical collectivity, where the state of the particle can be defined by a set of variables $\vec{S}_i$.

The idea of MC method is to generate random paths in the phase space. As much steps we take, more precise will be the observables estimated. To generate these random paths, occurring with a proper probability, a Markov chain is used (its probability distribution is unknown). This process is characterized by a set of stochastic events, where the probability depends directly on the preceding one. Before we perform the calculation it is needed to take some steps, what is called relaxation time, until it can be considered that energy gets an equilibrium. After that, we are ready to get the average of the observables we want to have in account.

Our cylinders of length L and radius R are made of a single wall structure which has been curved, where the spins stay on the surface. The magnetic result of extending this plane to $3D$ is the purpose of our study. Every magnetic moment has as components of spin the Heisenberg $\vec{S} = (S_x, S_y, S_z)$ for a cylinder of radius R and length L The Hamiltonian of the system is:

$$\mathcal{H} = E_{ex} + Edip + Eani, \tag{3}$$

E_{ex} is the exchange energy of short-range between first neighbors, where $J_{ij} = J > 0$ is the exchange constant, positive for ferromagnetic interactions. It is relevant up to a length scale of 10 nm. [4]

$$E_{ex} = -\sum_{<i,j>} J_{ij}\vec{S}_i \cdot \vec{S}_j, \tag{4}$$

The dipolar energy is a long-range interaction, which its sum extends to the whole system of spins, where D is the dipolar constant and $\vec{r}_{ij}$ is the relative distance between spins i and j

$$E_{dip} = -D\sum_{i<j} \frac{\vec{S}_i \cdot \vec{S}_j - 3(\vec{S}_i \cdot \hat{r}_{ij})(\vec{S}_j \cdot \hat{r}_{ij})}{\mid \vec{r}_{ij} \mid^3}, \tag{5}$$

The magnetic anisotropy describes how an object's magnetic properties can be different depending on direction

$$E_{ani} = -K\sum_{i=1}^{N}(\vec{S}_i \cdot \vec{n}_i)^2, \tag{6}$$

Implementation of Metropolis algorithm

First of all, the system is initialized in a disordered spin configuration generated at random and its energy and local exchange and dipolar fields are computed. Next, we visit a random lattice site, a new direction for its spin is proposed and the energy variation ΔE between the corresponding configurations is calculated. Then, two cases are possible:

- If $\Delta E \leq 0$ the change is always accepted. Then we update the spin direction and the magnetization. Also the energy with the help of the already stored dipolar and exchange fields, which are finally updated using the change in the spin orientation,

- If $\Delta E > 0$ we compute the Boltzmann probability $p(T) = e^{-\Delta E/k_B T}$. A random number r with an uniform distribution [0,1] is sorted. If $r \leq p(T)$, we accept the new spin direction and other quantities as described above . Otherwise, the initial spin direction is kept.

This process is repeated N times, completing what is called a MC step. After which, the values of the observables of interest are accumulated for subsequent calculation of thermodynamic averages. At every temperature, a number of MC steps are used to perform averages of the quantities of interest.

In order to obtain the configuration in thermodynamic equilibrium we need to lower the temperature as much as possible. The cooling of the system can be done either in a linear way or with a simulated annealing. This second protocol consists on a progressive decreasing from an initial value to zero. We want to get a state with the minimum possible energy, because there are many local minima. For both protocols the initial and final temperature are 3,5 J/k_B and 0,1 J/k_B respectively and 1.2×10^4 Monte Carlo steps, which of them 1.0×10^4 are used for averaging. We step over 85 temperatures with linear protocol, and with the annealing one we do it for 70 temperatures.

III. SIMULATION

The main purpose of the work is getting a well-founded design of the nanotubes with valid results. Before diving in the study of the interactions, the program has been tested comparing the output data with the report of Salinas et al. [6].

The ground state of the system is the equilibrium configuration at the end of the lowest temperature relaxation. The state that can be reached depends on the geometrical properties of the cylinder and its stability is determined by the magnetic interactions as explained in the model.

Firstly, the work is focused in soft magnetic materials where anisotropic energy can be neglected when compared to the exchange and dipolar counterparts[7] It is more convenient to define an interaction parameter $\gamma = D/J$ that treats the competition between both

interactions taken into account. Later on, we are going to add the magnetocrystalline anisotropic energy will be added at the system with the objective of knowing how the introduction modifies the magnetism of the structure.

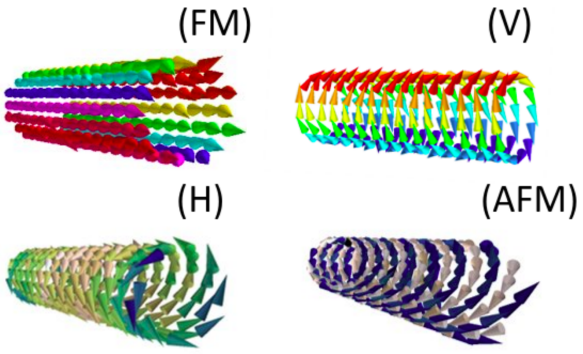

FIG. 2: The four stable states found throughout the study. Ferromagnetic (FM), helical (H), vortex (V) and antiferromagnetic (AFM), in increasing order of γ parameter respectively. .

For low values of the parameter γ, it is clear that the exchange energy of next neighbors beats the dipolar interaction and then a ferromagnetic order is reached. As γ is increasing, we can see how vortexes start being created at the ends of the tube when Monte Carlo sampling procedure is completed, and how those "move along" through the cylinder until getting the vortex state for larger values of γ.

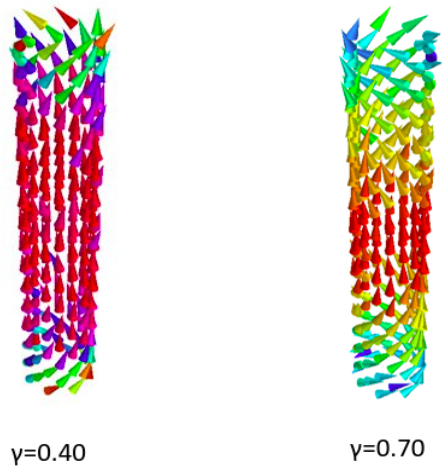

FIG. 3: Mixed states with aligned spins at the center and vortices forming at the ends of the tube. It is a ZZ tube of length 21.2 and radius 1.8 made of 248 spins

Anisotropy

Besides the competence between exchange and dipolar interactions, we aggregate a radial anisotropy (6), causing the formation of out-of-plane configurations, whereas dipolar interaction induces in-plane configurations. We have adjusted the exchange interaction parameter constant equaled to the unity.

When the values of anisotropy are much greater than the dipolar one $K \gg D$, the system has a null magnetization and the spins point perpendicular to Z axis. Something different occurs when both parameters are in the same magnitude order, getting ferromagnetic configurations as well, but in a perpendicular direction.

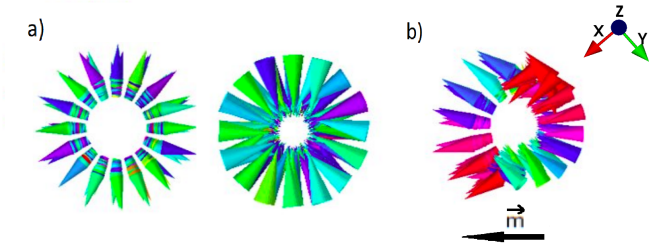

FIG. 4: The tubes are influenced by an anisotropy of value K=3. In a) case, both tubes with γ=0.08, does not appear a net total magnetization. On b) case, with γ=0.10 a ferromagnetic state appears in the direction shown. They are ZZ tubes of length 21.2 and radius 1.8 made of 248 spins

In this section, the inspection of states has been merely optical. A further analysis it is forthcoming, sweeping a range of values of γ.

IV. RESULTS

It is worth noting that length, radius and thermodynamics observables are represented by numbers characterised of having arbitrary units.

	linear & Heisenberg			annealing & cone		
γ	$\lvert m_{z_1}\rvert$	$\lvert m_{z_2}\rvert$	$\lvert m_{z_3}\rvert$	$\lvert m_{z_1}\rvert$	$\lvert m_{z_2}\rvert$	$\lvert m_{z_3}\rvert$
0.05	0.978	0.978	0.979	0.978	0.980	0.979
0.30	0.976	0.962	0.972	0.962	0.965	0.968
0.40	0.921	0.925	0.915	0.917	0.924	0.912
0.50	0.869	0.865	0.881	0.871	0.872	0.869
0.70	0.747	0.795	0.775	0.805	0.752	0.757
0.80	0.329	0.438	0.571	0.470	0.546	0.486
0.85	0.421	0.300	0.146	0.140	0.194	0.003
0.90	0.126	0.170	0.066	0.033	0.075	0.022
0.95	0.056	0.076	0.124	0.075	0.032	0.012
1.00	0.003	0.075	0.101	0.029	0.003	0.003
1.20	0.003	0.001	0.010	0.013	0.008	0.003

TABLE I: Data taken over 3 independent runs. The subscript in the z component of the magnetization means the ith time of the try. It consist of a tube of length 21.2 and radius 1.8 made of 248 spins.

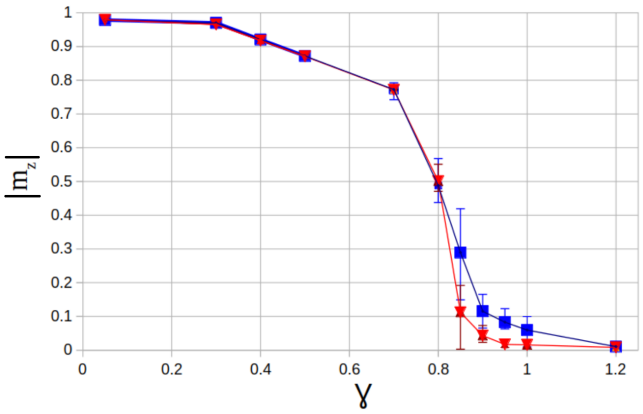

FIG. 5: Dependence of the z component of the magnetization as a function of the interaction parameter. Blue squared points have been obtained using a linear T decrease protocol and random spin trials. The triangular red points are results corresponding to the annealing protocol with a restricted spin trials.

When treating with linear cool-down and random new configuration proposed, as explained in the Metropolis algorithm subsection, it is found a substantial difference between independent runs in those simulations whose equilibrium state is vortex, $\gamma > 0.8$. This nuisance can be interpreted as the system gets stuck in many local minima. To solve this issue, we introduced two main changes in the program. Another configuration is proposed, the new spin will be restricted to lie inside a cone with a maximum overture, instead of a Heisenberg random spin. Therefore, to make more accurate the finding of the lowest energy states consist on a progressive decreasing to the final temperature.

When an anisotropy is introduced, there is a similar framework, but this new influence disorder the spins until it creates a ferromagnetic phase perpendicular to Z axis for a lower dipolar interaction. A persisting descend of the dipolar value leads to non-magnetic configurations where all the spins are oriented inside or outside the tube in a radial way, with same probability.

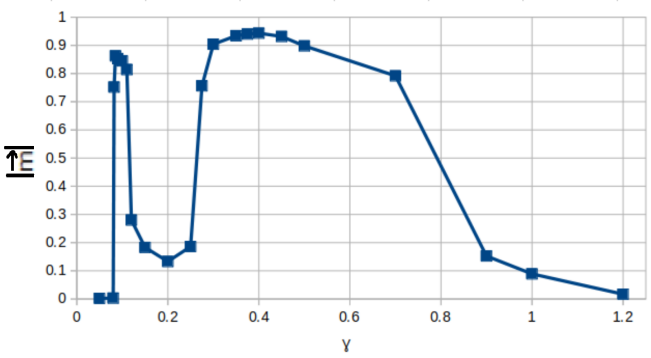

FIG. 6: Dependence of the magnitude of magnetization γ when the magnetocrystalline anisotropy is added for a tube of length 21.2 and radius 1.8 made of 248 spins.

At once, we are analyzing the thermodynamics of the system. In Fig. 7 it is shown a decreasing energy when γ value is increased.

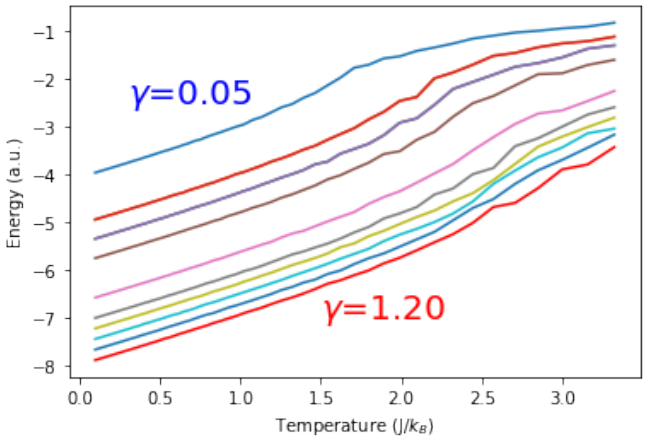

FIG. 7: Thermal dependence of the energy during a linear decrease from a high temperature disordered state to $T = 0.1$ for γ varying between those indicated and tabulated in table I.

We calculate the response functions specific heat and susceptibility as fluctuations of energy and the parameter order respectively

$$c_v = \frac{k_B \beta^2}{N}(\langle E^2 \rangle - \langle E \rangle^2), \tag{7}$$

$$\chi = \beta N(\langle m^2 \rangle - \langle m \rangle^2), \tag{8}$$

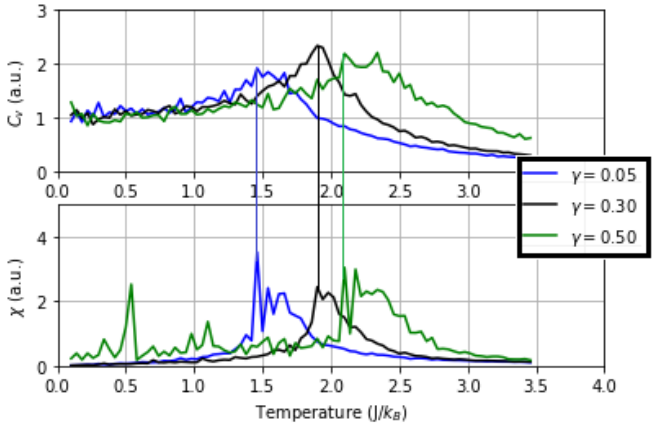

FIG. 8: Thermal dependence of the specific heat and susceptibility. Take notice how the peaks in both pictures are in the same temperature. The units are arbitrary.

The curved peak in the pseudo-critical temperature is between 1.5K-2K and marks the transition from paramagnetic to ferromagnetic system. In a system where

the volume is finite, the concept of singularity and phase transition does not exist and that explains why this peak is rounded instead of tending to infinity. The concept of critical temperature has only a significance in the thermodynamic limit, when the number of spins $N_{spin} \to \infty$ as the Theory of Yang-Lee explains.

For large values of gamma there is no peak as mentioned, since the system passes from a random configuration to a vortex one, neither of them with a net magnetism.

The magnetization as a parameter order is worth within the thermodynamic limit, but for a better study of the configurational transitions could be better a more precise parameter order. A candidate would be the vorticity that depicts the flow circulation of the magnetic nanotube.

$$\vec{\rho} = \vec{\nabla} \times \vec{S_i}, \qquad \chi = \frac{\beta}{N}(\langle \rho_z^2 \rangle - \langle \rho_z \rangle^2), \qquad (9)$$

The parameter order proposed would characterize helicoidal (a metastable state) and antiferromagnetic systems, studying also mechanisms of magnetic inversion.

Whilst hitherto it was considered a ZZ topology, the aim of the next study is comparing how the angle in the construction of the tube affects the final configurations. The length/radius aspect ratio has been maintained. There is no apparent relation between the transition of phases and the angle of the spins around the tube as can be seen in Table II. .

Topology	R	L	L/R	Nspin	γ_{FM}	γ_V
ZZ ($\theta = 45°$)	2.25	14.14	6.28	210	0.15	0.30
AA ($\theta = 0°$)	2.23	14.00	6.28	210	0.10	0.23
Chiral($\theta = 26.57°$)	2.14	13.42	6.28	186	0.10	0.45
Chiral($\theta = 14.04°$)	2.59	16.50	6.28	276	0.08	0.30

TABLE II: There is no apparent relation between the transition of phases and the angle of the spins around the tube.

V. CONCLUSIONS

In spite of not having a demonstrative answer extracted from these last results, it yields some valuable conclusions. For a proper study, it is recommended to have much more runs into account, with higher proportions, going ahead with large radius, large length and keeping or varying the proportions as well.

There are plenty of difficulties to make a better estimation and the obtaining of better results. First of all, having a cylinder in a desired proportion it is already a number theory problem, since for setting up the tube varying the input parameters the user must become aware about the length of the vectors, the angle between them and how many spins can be placed in each chain of spins surrounding the surface of the tube. Besides, it must be taken into account the existence of many local minima, so most of the times the MC process of the simulation takes a path that does not end in the ground state, leading to configurational states that are far to correspond to the typical state with particular known parameters.

The simulation of longer and wider tubes, or a greater quantity of temperatures and MC steps, aiming to get more accurate outcomes, results into a substantial computational time, even for the supercomputer of CSUC facilites, making a better resolution of the results out of the goal of this work, as well as the implementation of the vorticity as parameter order and the vast field that opened this update.

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