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# Magnetic structures of 2D and 3D nanoparticles

J.-C.S. Levy

*Matériaux et Phenomenes Quantiques, University Paris Diderot, 75013 Paris, France*

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ABSTRACT

The minimization of exchange interactions and dipolar interactions in 2D and 3D nanoparticles is obtained from a powerful variational approach of the local spin Hamiltonian and leads to a different set of equations which correspond to different levels of screening of the long range dipolar interactions. These equations are shown to introduce topological defects which are analyzed on the basis of elementary spin clusters. Four basic topological defects are deduced for 2D nanoparticles, as observed in magnetic samples and simulations and 10 basic topological defects are deduced for 3D nanoparticles. These singularities induce complex variations of magnetization around them and non-linear properties.


## 1. Introduction

Since the very beginning of magnetic domain observations in magnetic materials of various natures, sizes and shapes [1], there has been a strong experimental evidence for intrinsic walls, linear singularities and topological defects in the magnetic structure of materials. More specifically magnetic thin films and ultra-thin films [2] showed labyrinthine and vortex structures as well as other topological defects. More recently experimental results on the magnetic structure of nanofilms were helped by theoretical simulations used to deal with micromagnetism such as OOMMF method [3], Monte-Carlo computations [4] and Langevin dynamics [5] and these approaches showed localized topological defects and their consequences on dynamical properties [6]. 3D magnetic nanoparticles of interest for catalytic applications and for transport properties like printing or biological applications as guided vectors were recently shown to also exhibit a complex 3D domain structure at their external surfaces [7].

There are several difficulties in 3D analyses of magnetic nanoparticles, since mainly external surfaces are observed without data on internal structure. Moreover there is a lack in 3D completely reliable simulations up to now. Recent works explore local magnetic structures in nickel nanocylinders [8] as well as in antiferromagnets [9]. So the natural aim of this paper is to introduce a new theoretical analysis of the magnetic ground state of 2D and 3D nanoparticles by means of a variational approach and so to deduce the 3D observable topological defects.

The obvious origin of complex magnetic structures is the long ranged dipole–dipole interaction competing with local exchange and anisotropy [10]. Real samples are finite, so all dipolar contributions such as local demagnetizing field and dynamical matrix elements are far from being uniformly spread over the sample and the long range nature of dipolar interactions seriously increases this spreading [11]. Non-uniformity breaks the hopes of a magnetic sample translational invariance as assumed from [12] and followers about magnetic structures. As a result, the obvious finiteness of samples induces localization in the spin wave spectra as observed in numerical computations [11] and as a later consequence, it induces observed static topological defects [1] resulting from localized soft modes. So there is a real need for a theoretical approach of nanoparticle magnetic structure and especially for 3D nanoparticles. Here we are looking for a variational treatment from a generalized Landau's local version of the spin Hamiltonian.

The long ranged dipole–dipole interaction is translated into a local interaction by means of Taylor expansion of the spin field introducing spin derivative fields as previously done about 2D samples [10], with lattice sums or more exactly sample sums with symmetry properties. This approach explains the occurrence of non-linear properties [6], which are efficient out of domains, i.e. at walls and defects as observed [6]. The zero order approximation of Taylor expansion is linked with the concept of demagnetizing field while the second order approximation of the spin Hamiltonian gives the main set of equations on magnetic structures deduced from minimization. These equations show a higher level of complexity for 3D samples than for 2D ones. Of course higher order equations correspond to higher levels of screening of the long ranged interaction and can also be deduced as in the two-dimensional case [10].

The search for topological defects in 3D samples requires a local analysis within a spherical frame, i.e. for instance here a basic cluster. For 2D samples, just four basic topological defects when considering neighboring sites only: vortex, two antivortices and a strongly asymmetric one, while for 3D samples, 10 basic

*E-mail address:* jean-claude.levy@univ-paris-diderot.fr

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topological defects occur. For 2D samples as well as for 3D ones complex defects involving several basic defects occur also, so there are many complex topological defects. Harmonic magnetic excitations, i.e. spin waves, are defined in that context of topological defects [10] and can be observed by magnetic resonance [14].

From the local spin Hamiltonian a Landau like variational treatment enables us to obtain the basic equations which are satisfied by topological defects. These topological defects are then deduced from simple geometrical criteria.

## 2. The spin Hamiltonians

Here we consider the exchange interaction between spins located on neighboring states

$$H_{exc} = -\frac{1}{2}\sum J_{ij}\overrightarrow{S_i} * \overrightarrow{S_j} \quad (1)$$

With infinite Taylor expansion of the spin field introducing the spin field derivatives:

$$\overrightarrow{S}_j = \sum_{p,q,r} \frac{x_{ij}^p y_{ij}^q z_{ij}^r}{p!q!r!} \frac{\partial^{p+q+r}}{\partial x^p \partial y^q \partial z^r} \overrightarrow{S}_i \quad (2)$$

It leads to a strictly local interaction in terms of the square of the gradient of the spin field, a standard result. The anisotropy interaction is local and so does not require any translation in terms of local field. The dipolar field induces the well known dipole–dipole interaction:

$$H_d = \sum_{i \neq j} \frac{\overrightarrow{S_i} * \overrightarrow{S_i}}{r_{ij}^3} - 3\sum_{i \neq j} \frac{(\overrightarrow{S_i} * \overrightarrow{r_{ij}})(\overrightarrow{S_i} * \overrightarrow{r_{ij}})}{r_{ij}^5} \quad (3)$$

Again using Taylor expansion of the spin field dipolar interaction reads as a local one in terms of all spin derivative fields when introducing lattice sums, i.e. more exactly sample sums $I_{p,q,r;i}$ and $J_{p,q,r;\alpha,\beta;i}$:

$$H_d = \sum_{p,q,r,i} I_{p,q,r,i}\left(S_i * \frac{\partial^{p+q+r}}{\partial x^p \partial y^q \partial z^r}\overrightarrow{S}_i\right) - 3\sum_{p,q,r,\alpha,\beta,i} L_{p,q,r,\alpha,\beta,i}\left(S_{\alpha,i}\frac{\partial^{p+q+r}}{\partial x^p \partial y^q \partial z^r} S_{\beta i}\right) \quad (4)$$

where isotropic and anisotropic sample sums are respectively

$$I_{p,q,r;i} = \sum_j \frac{x_{ij}^p y_{ij}^q z_{ij}^r}{p!q!r!\left(x_{ij}^2 + y_{ij}^2 + z_{ij}^2\right)^{3/2}}$$

$$L_{p,q,r,\alpha,\beta;i} = \sum_j \frac{r_{\alpha,ij} r_{\beta,ij} x_{ij}^p y_{ij}^q z_{ij}^r}{p!q!r!\left(x_{ij}^2 + y_{ij}^2 + z_{ij}^2\right)^{5/2}} \quad (5)$$

For infinite samples many of these lattice sums would diverge. Here sample sums remain finite and are often assumed to have a weak local variation, for the sake of simplicity. In that case, a continuous calculation by means of 3D integration will be introduced for the estimation of these sums as integrals.

The principle of our variational approach consists of introducing a local arbitrary small deviation of a spin orientation on site $k$ of weak amplitude C:

$$S_i = S_{i,0} + C\delta(i-k)S_{k,0} \wedge \overrightarrow{n} \quad (6)$$

Here $\overrightarrow{n}$ is an arbitrary unit vector, $\delta(i)$ is the Dirac delta function and Eq. (6) ensures the spin amplitude conservation. Since variations for every site are independent, the variation basis is complete.

## 3. Basic results

At level 0 of the Taylor expansion of the dipolar interaction, omitting the spin anisotropy term, the effect of demagnetizing field only occurs as induced by the anisotropic character of dipolar interaction. The set of optimization equations reads, within the assumption of uniform sample sums:

$$(B-C)S_z S_y = 0$$
$$(C-A)S_x S_z = 0$$
$$(A-B)S_y S_x = 0 \quad (7)$$

With

$$A = L_{000,11} \approx \sum\frac{x^2}{r^5}\; B = L_{000,22} \approx \sum\frac{y^2}{r^5}\; C = L_{000,33} \approx \sum\frac{z^2}{r^5} \quad (7')$$

Here only order of magnitude is considered, omitting tedious numerical factors. In other words the classical shape effect of the demagnetizing field is found: for a flat sample in the $xy$ plane, magnetization is in plane within this continuous model. Of course the detailed calculation can be achieved in a general 3D case, at the expanse of complexity according to the sample shape.

At level 2 of the Taylor expansion of the dipolar Hamiltonian, always considering sample parameters as uniform over the sample, a set of three equations over the spin field components and their derivatives is obtained. The first equation of this set reads as a matrix product:

$$\begin{pmatrix} A-3D & B-3G & C-3J & -6E & -6F & \lambda \end{pmatrix} \begin{pmatrix} \frac{\partial^2 S_x}{\partial x^2} \\ \frac{\partial^2 S_x}{\partial y^2} \\ \frac{\partial^2 S_x}{\partial z^2} \\ \frac{\partial^2 S_y}{\partial x \partial y} \\ \frac{\partial^2 S_z}{\partial x \partial z} \\ S_x \end{pmatrix} = 0 \quad (8)$$

Here the coefficients $A$, $B$ and $C$ are issued from isotropic sample sums $I$ while coefficients $D,E,\ldots, L$ come from anisotropic sample sums $L$, and $\lambda$ is a free parameter. Two other equations with similar coefficients are derived according to circular permutations. These equations complete the set of second order equations. It must be noticed that while lattice sums depend on lattice geometry and lattice parameter, sample sums and related coefficients depend also on sample shape as shown in Eq. (7′). Thus Eq. (8) defines a large class of spin fields submitted to second order partial derivative equations.

Introducing a common Fourier transform for the spin field with wavevector$(k_x, k_y, k_z)$, the set of equations leads to the characteristic condition on wavevectors:

This set of properties of 3D magnetic structures is quite more complex than the set obtained for a 2D sample [10] which can be deduced from the more general 3D case from Eqs. (8) and (9). The characteristic Eq. (9) of order 6 means the occurrence of static wavy like deformations in the ground state.

The next point to consider consists of deriving the topological defects which correspond to Eq. (8), within a spherical frame. Since these equations are submitted to very large changes

$$\det\begin{pmatrix} -(A-3D)k_x^2-(B-3G)k_y^2-(C-3J)k_z^2+\lambda & 6Ek_xk_y & 6Fk_xk_z \\ 6Ek_xk_y & -(A-3E)k_x^2-(B-3H)k_y^2-(C-3K)k_z^2+\lambda & 6Ik_zk_y \\ 6Fk_xk_z & 6Ik_zk_y & -(A-3F)k_x^2-(B-3I)k_y^2-(C-3Jl)k_z^2+\lambda \end{pmatrix} = 0 \quad (9)$$