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Magnetization in uniaxial spherical nanoparticles: consequence on the interparticle interaction

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Abstract

We investigate the interaction between spherical magnetic nanoparticles which present either a single domain or a vortex structure. First the magnetic structure of a uniaxial soft sphere is revisited, and then the interaction energy is calculated from a micromagnetic simulation. In the vortex regime the orientation of the vortex relative to the easy axis depends on both the particle size and the anisotropy constant. We show that the leading term of the interaction is the dipolar interaction energy between the magnetic moments. For particles presenting a vortex structure, we show that the polarization due to the dipolar field must be included. The parameters entering in the dipolar interaction are deduced from the magnetic behavior of the isolated particle.

Keywords : magnetic nanoparticles; micronetic simulations; dipolar interaction

PACS: 75.75.+a

1 Introduction

The magnetic behavior of nanometric particles either isolated or in nanostructured bulk materials is now quite well understood both from experiments or numerical calculations [1, 2]. With the growing diversity of systems made of such nano-objects as building blocks either as 2D or 3D systems in non magnetic environment or in colloidal suspensions as ferrofluids, a precise knowledge of interparticles interaction is needed. This has already been done at least partially in a variety of systems, such as nanograins [3, 4] nanorings [5] or flat nanodots [6, 7], with a predominant attention paid on short range effects. A lot of work remains to be done in this field and especially for spherical particles. In particular it seems important to develop models for the long ranged and anisotropic dipolar interaction. A further interpretation of experimental results as those of [8] necessitates such models.

2 Magnetization structure and hysteresis

We consider a spherical particle characterized by a radius R ranging from $10nm$ to $45nm$, an exchange constant, $A_x = 1.10^{-11}J/m$ and a saturation magnetization $J_s = 1T$ corresponding roughly to Permalloy. The anisotropy constant of the uniaxial magneto-crystalline energy is varied between $K_1 = 0$ and $7.10^4 J/m^3$. The numerical calculations are performed with the framework of micromagnetism with the code MAGPAR [9] based on a FEM method. The particle volume is v_s and hatted letters denote unit vectors. Small particles, up to roughly $R = 20nm$ for our parameters, are uniformly magnetized as single magnetic domains with square hysteresis curve. When the particle radius increases beyond roughly $R = 20nm$, a vortex structure is obtained, and the local magnetization profile, $\vec{m}(\vec{r})$, is decomposed in its cylindrical components using the vortex axis, say \hat{v} , as the cylindrical axis. At zero or small values of the external field, the direction of the vortex relative to the easy axis, \hat{a} , depends on both the value of R and of K_1 , a behavior already obtained in the case of the cubic anisotropy [10]. We find, as expected, $\hat{v} \perp \hat{a}$ either for $R > R_{th}$ or $K_1 > K_{1,th}$ at constant K_1 or R respectively, R_{th} and $K_{1,th}$ being some threshold values. ($R_{th} = 26nm$ for $K_1 = 3.10^4 J/m^3$ and $K_{1,th} = 2.10^3 J/m^3$ for $R = 45nm$). The orientation of \hat{v} relative to \hat{a} affects strongly the $M(H_{ex})$ curve. With an external field along \hat{a} , $\hat{v} \perp \hat{a}$ leads to a magnetization curve qualitatively similar to that of a flat nanodot with an in plane external field [11, 2]: there is no remanence and $M(H_{ex})$ presents two lobes. For spherical particles these lobes are associated to the rotation of the vortex core in the direction of the field prior to its annihilation. The magnetization normal

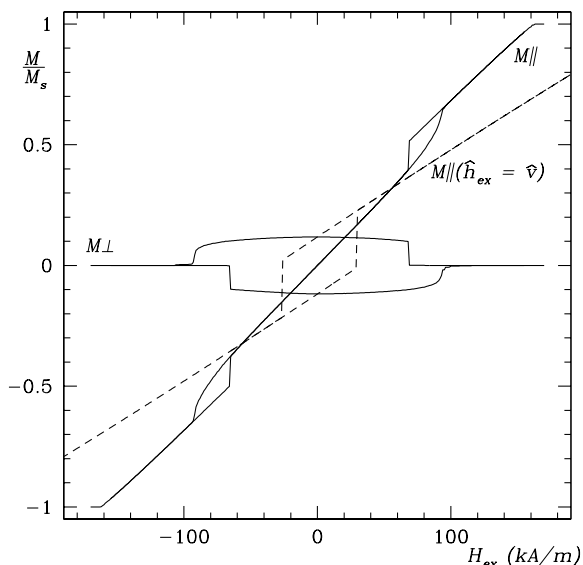


Figure 1: Magnetization parallel and normal to the field. $\hat{h}_{ex} = \hat{a}$ (solid line) or \hat{v} (dashed line). $R = 45nm$, $K_1 = 3.10^4 J/m$.

to the field takes a non zero constant value corresponding to the vortex core magnetization, when the variation of $M \parallel$ results from a shift of the vortex normal to the field. this value coincides with the remanence obtained with $\hat{h}_{ex} = \hat{v}$. This behavior is displayed on figure 1.

As is generally obtained in nanodots or spherical particles [11, 12, 13] the magnetization M in the direction of the external field depends nearly linearly on the field in the vicinity of $H_{ex} = 0$ and away from switching points where the vortex reverses as a whole. Such a linear behavior is observed both when $\hat{h}_{ex} = \hat{v}$ or $\hat{h}_{ex} \perp \hat{v}$. This means that the susceptibility χ defined as $\frac{\partial M}{\partial H_{ex}} = \chi$ does not depend on the field to a very good approximation. This can be exploited to obtain the variation of the total energy with respect to the external field. We analyse the variation of the magnetization, ΔM as the polarization of the sphere induced by the field. Starting from $\Delta M(H_{ex}) = \chi H_{ex}$, we deduce $\Delta M(H_{ex})$ from the energy, $E(H_{ex})$ by writting an equilibrium equation $\frac{\partial E(\Delta M)}{\partial \Delta M} = 0$. The total energy depends explicitly on H_{ex} through the Zeeman term, $E_Z = -\mu_0 H_{ex} (m(0)\hat{v} \cdot \hat{h}_{ex} + \Delta M)$, where we have expressed the permanent magnetization in the absence of the field as $\vec{M}(H_{ex} = 0) = m(0)\hat{v}$ and $m(0)$ denotes the magnitude of the vortex core magnetization in the absence of the field. Then we get

$$\frac{\partial}{\partial \Delta M} (E_{dm} + E_x + E_a) = \mu_0 H_{ex} \quad (1)$$

Therefore the variation of the total energy is

$$E(H_{ex}) - E(0) = \frac{\mu_0}{2\chi} \Delta M^2 + E_Z(H_{ex}) \quad (2)$$

where we have used $\Delta M(H_{ex}) = \chi H_{ex}$. μ_0 is the vacuum permeability and E_Z the Zeeman energy. Notice that (1) is exact while (2) holds only in the case of a linear dependence of $\Delta M(H_{ex})$ with respect to H_{ex} . The first term of the r.h.s. of (2) is the polarization energy of the sphere [14] and corresponds to the energy cost for the reorientation of the magnetization inside the sphere. It can be written equivalently as $(\mu_0 \Delta M H_{ex})/2$

3 Interaction between magnetic nanospheres

The interaction energy between two magnetic nanoparticles in terms of the interparticle distance, r_{12} , is defined from the total energy of the two spheres system

$$E_{int}(1, 2) = E_{tot}(1, 2) - E_{tot}(r_{12} \rightarrow \infty) \quad (3)$$

where $(1, 2)$ is a short notation for the orientation and location variables of the particles. We expect to get a form dictated by the dipolar interaction between the magnetic moments of the approaching spheres which reads

$$E_{dip} = \frac{\mu_0 m_1 m_2}{4\pi r_{12}^3} d_{112}(\hat{m}_1, \hat{m}_2, \hat{r}_{12}) \quad (4)$$

$$d_{112}(\hat{m}_1, \hat{m}_2, \hat{r}_{12}) = \hat{m}_1 \cdot \hat{m}_2 - 3(\hat{m}_1 \cdot \hat{r}_{12})(\hat{m}_2 \cdot \hat{r}_{12}) \quad (5)$$

where m_i are the magnitude of the magnetic moments. For single domain particles, $m_i = M_s v_s$ where M_s is the saturation magnetization of the particles, and the orientations \hat{m}_i result from the minimum of d_{112} given in (5). For particles without magnetocrystalline anisotropy, this gives: $\hat{m}_1 = \hat{m}_2 = \hat{r}_{12}$ and $d_{112} = -2$. For non zero magnetocrystalline energy on both particles with easy axes \hat{a}_i , the orientations \hat{m}_i result of the interplay between the anisotropy energy tending to align \hat{m}_i on \hat{a}_i and the energy (4) tending to minimize the angular function (5). If K_1 takes a non vanishing value only in one particle say $i = 1$ and is large enough to impose $\hat{m}_1 = \hat{a}_1$, \hat{m}_2 must orient in the dipolar field due to particle 1 *i.e.* in such a way that $d_{112} = \hat{m}_2 \cdot \hat{a}_1 - 3(\hat{m}_2 \cdot \hat{r}_{12})(\hat{a}_1 \cdot \hat{r}_{12})$ is minimum. The whole behavior outlined above is very well reproduced by the full micromagnetic calculation which demonstrates the dipolar nature of the interaction between single domain particles.

In the case of particles large enough to present a vortex structure, the orientations of the effective moments of the particles are the vortex directions, \hat{v}_i , and the values of the moments correspond to the vortex cores magnetizations. We introduce the coefficient $\alpha_i = m_i / (M_s v_s)$. The value taken by α depends on both the characteristics of the particle and r_{12} and d_{112} through the polarization of the particle by the dipolar field of the second one. ($\alpha(1, 2) = \alpha(r_{12}, d_{112})$ and $\alpha_0 = \alpha(\infty)$). A simple approximation for the interaction energy is built by considering that each particle is in the dipolar field of the other. We have to take into account two contributions. The first one is given by (4), and the second one is twice the polarization energy of one sphere in the field of the second one. The second contribution has been introduced in (2) for one particle in a constant external field. The role of $m(o)$ is played by $M_s v_s \alpha_0$ while the induced moment in the direction of the dipolar field is $\vec{p} = p \hat{h}_{dip} = \chi H_{dip}(r_{12}) \hat{h}_{dip}$. We consider the case where the vortex \hat{v}_i is free to orient in the direction of the dipolar field due to particle $j \neq i$. This corresponds to either the absence of anisotropy or particles large enough for the vortex to be normal to the easy axis and $\hat{a}_1 = \hat{a}_2$. In this case we have $\vec{p} = (\alpha(1, 2) - \alpha_0) M_s v_s \hat{h}_{dip} = \Delta\alpha(1, 2) M_s v_s \hat{v}$ and adding twice the first term of (2) to the dipolar energy we get

$$E_{int}(1, 2) = \frac{\mu_0 (M_s v_s)^2}{4\pi r_{12}^3} \alpha_0 (\alpha_0 + \Delta\alpha(1, 2)) d_{112} \quad (6)$$

which coincides with the interaction energy between polar polarizable hard spheres. Then we have to calculate $\Delta\alpha(1, 2)$; for the simple case of two particles, introducing $u = \chi / (4\pi R^3)$ we get

$$\Delta\alpha(1, 2) = \alpha_0 \frac{-u d_{112}}{(r_{12}/R)^3 + u d_{112}} \quad (7)$$

A typical example of the interaction in terms of the distance r_{12} for $d_{112} = -2$ is displayed on figure 2. Similar results are obtained for other particles. The full calculation is compared to the analytical model given by (6). We consider two levels of approximation: either the simple dipolar interaction, where the polarization energy is neglected ($u = 0$) and the interaction corresponding to the dipolar polarizable spheres. Except in the vicinity of contact, ($r_{12} = 2R$) *equ.* (6) reproduces quite well $E_{int}(1, 2)$. The parameters needed, α_0 and χ are deduced from the magnetization curve of the isolated particle. We can therefore conclude that the interaction between spherical soft magnetic particles is of dipolar nature, and that the polarizability must be included when the particles present a vortex structure.

4 Concluding remarks

We can conclude that the leading interaction between soft magnetic nanoparticles corresponds as expected to the magnetic dipolar interaction. Then two situations must be distinguished according to the particle size: the interaction corresponds to the total magnetic moment without any polarizability contribution in the case of small particles which are in a single domain state when isolated, while a polarizability term must be included when the particles present a vortex structure. In this latter case, the value of the permanent moment, represented by the coefficient α_0 in the present work, is no more trivial, but can be determined as well as the relevant susceptibility χ from the isolated particle properties. We emphasize that here, the solution of the equation for the variation of $\Delta\alpha$ in terms of r_{12} and $d_{112}(1, 2)$ is quite obvious since there is only two particles, but this should be not the case for an assembly including a large number of particles where the interaction will present a n -body character. Finally, because of the dipolar nature of the leading interaction, we can confirm the behavior observed experimentally in [8] since the vortex of neighboring particles are expected to align themselves.

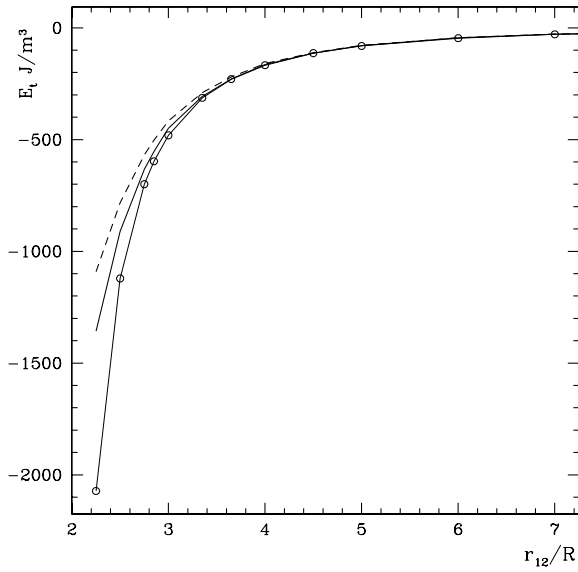


Figure 2: Interaction energy per unit volume between two spheres. $R = 35nm$; $K_1 = 0$; $d_{112} = -2$. Open circles: full calculation, from (3) (the thin line is a guide to the eye); solid line: equ.(6); dashed line: simple dipolar approximation, $u = 0$.

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