

# Physical properties of magnetic grains dispersed in anisotropic media

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**Abstract.** We calculate the dynamical response of a group of non-interacting magnetic grains dispersed in a medium that imposes on them an uniaxial anisotropy. Numerical results are obtained for a collection of grains that has its volume distribution given by a Gaussian function centered in the average volume  $V_0$ , and has a width  $\sigma V_0$ . We assume that the only effect of the medium on the particles is the anisotropy imposed on the grains to study the influence of the volume distribution ( $\sigma$  and  $V_0$ ) on the effective permeability. The results are analyzed through the calculation of numerical values for the components of the magnetic permeability and also by the analysis of the combined effect of the external dc magnetic field and the width of the volume distribution on the skin depth.

**PACS.** 75.40.Gb Dynamic properties (dynamical susceptibility, static susceptibility, spin waves, spin diffusion, dynamic scaling, etc.) – 75.50.Kj Amorphous and nanocrystalline magnetic materials; quasicrystals – 75.50.Tt Fine-particle systems; nanocrystalline materials

## 1 Introduction

The recent development of new technologies makes possible to synthesize small size particles tailored to fit different needs [1–9]. Despite the effort of a considerable number of researchers, many of the atypical properties observed in these systems are still waiting for physical justifications. It should be remarked that many of the features attributed to these particles are observed when they are forming an arrangement of grains. This fact suggests that (at least) some of the properties observed are not only due to attributes of single particles, but instead, they are features resulting of the collective behavior. Therefore, it is important to understand the physical properties of very small size particles, but it is also equally important (specially from the application standpoint) to understand the collective behavior of systems composed by a large number of these particles. The aim of this paper is to give a simple and useful procedure to obtain a description of physical characteristics of these systems.

The procedure described here may be used to obtain effective parameters to describe a large variety of systems and the main restriction is that the property under investigation do not distinguish the different parts of the system. In other words, the result obtained for the observable is reliable if it is not the response of a small number of par-

ticles. For example, this procedure should give a good description of the optical characteristics of the system if we are analyzing results obtained for radiation with a wavelength much larger than the cubic root of the average volume of the particles, but it should fail if the wavelength of the radiation is comparable with the diameter of the particles.

Our approach to study these systems is similar to one made by Skeff Neto et al. [10] many years ago. We propose that any physical property of these systems is the averaged value of the contributions of the individual particles. In the following we illustrate the use of this assumption through the study of some physical behavior of a collection of non-interacting magnetic grains.

To do that we assume that if the number of particles with volume between  $V$  and  $V + dV$  is proportional to  $f(V)dV$ , any physical property observed ( $O_{ob}$ ) in these systems is the average value of the property of the individual particles ( $O_P(V)$ ) i.e.,  $O_{ob} = \int O_P(V)f(V)dV$ .

To analyze the consequences of these assumptions, we will calculate the magnetic susceptibility of a collection of non interacting particles. We will obtain numerical results considering that the  $f(V)$  is given by

$$f(V) = \frac{1}{\Omega} \exp[-(V - V_0)^2/2(\sigma V_0)^2] \quad (1)$$

where  $\Omega$  is the normalization factor ( $\int f(V)dV = 1$ ) and  $\sigma$  measures the width of the gaussian distribution centered

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at the average volume  $V_0$ . We assume the grains are in a medium that imposes a preferential direction for their magnetic moment. We will take into account this effect by considering that each grain feels an uniaxial anisotropy along the direction  $\hat{\theta}_K$ . The anisotropy constant ( $H_A$ ) will measure the energy necessary to rotate the magnetic moment of the grain by  $\pi$  starting from the  $\hat{\theta}_K$  direction. Certainly real systems may have different interactions and consequently have a different dependence of the energy on the volume. (Usually the interaction of two systems depends on the characteristics of both systems as well as on the environment.) We chose one that makes the anisotropy energy proportional to the square of the component of the magnetic moment along  $\hat{\theta}_K$  and then it is proportional to the square of the volume of the grain. This choice is useful to illustrate the method and the result for different systems should be quantitatively different but should follow the general behavior depicted here.

If we assume that, in addition to the assumptions above, the system is in presence of a static magnetic field  $H_0\hat{z}$ , the energy of a grain with a volume  $V$  is given by:

$$E(V) = -\frac{H_A}{2M_0}(\mathbf{M}(V)\cdot\hat{\theta}_K)^2 - \mathbf{M}(V)\cdot(\mathbf{H}_0 + \mathbf{h}(t)) \quad (2)$$

where  $M_0 = |\mathbf{M}_0|$  is the absolute value of the magnetic moment  $\mathbf{M}_0$  of a grain with the average volume  $V_0$ ,  $H_A$  measures the strain to rotate the magnetic moment  $\mathbf{M}(V)$  of the grain that has a volume  $V$  from its anisotropy axis. The last term is the Zeeman energy, supplemented by the coupling of the magnetic moments to the externally applied field  $\mathbf{h}(t) = (\hat{x}h_x + \hat{y}h_y + \hat{z}h_z) \exp(-i\omega t)$ .

It should be remarked that if the static magnetic field  $H_0\hat{z}$  is not parallel to the anisotropy direction  $\hat{\theta}_K$ , the equilibrium configuration will be the result of a competition between the Zeeman and anisotropy energies. Assuming that  $\mathbf{M}(V)$  is proportional to  $V$ , the anisotropy has a quadratic dependence on the volume while the dependence of the Zeeman energy is linear. The result of this competition is that the grains with volume smaller than a critical value will be aligned with the field while the rest will be in some direction between  $\mathbf{H}_0$  and  $\hat{\theta}_K$ . As a consequence, the distribution of volume of the particles leads to a distribution of the resonance frequency which should be controlled by the form of  $f(V)$ .

## 2 The magnetic permeability

The magnetic susceptibility of an individual grain of volume  $V$  and magnetic moment  $\mathbf{M}(V)$  can be obtained from its equation of motion

$$\frac{1}{\gamma} \frac{d\mathbf{M}(V)}{dt} = \mathbf{M}(V) \times \mathbf{H}_{eff} \quad (3)$$

where  $\gamma$  is the gyromagnetic factor and  $\mathbf{H}_{eff}$  is the effective field felt by  $\mathbf{M}(V)$  which is obtained from the equation (2) and given by

$$\mathbf{H}_{eff} = \frac{H_A}{M_0}[\mathbf{M}(V)\cdot\hat{\theta}_K]\hat{\theta}_K + \mathbf{H}_0 + \mathbf{h}(t). \quad (4)$$

At this point we should say that we are considering that the internal energy of the grains is constant and the only modification induced by the environment (medium and external field) is on the direction of the magnetic moments.

We write  $\mathbf{M}(V)$  as a sum of its static value  $[\mathbf{M}(V)]_0$  (the value of  $\mathbf{M}(V)$  that gives the minimum to  $E(V)$ ) and its fluctuation  $\boldsymbol{\eta}(t)$  to rewrite the equation (3) as:

$$\frac{1}{\gamma} \frac{d\boldsymbol{\eta}(t)}{dt} = \boldsymbol{\eta}(t) \times \left\{ \frac{H_A M_K^0(V)}{M_0} \hat{\theta}_K + \mathbf{H}_0 \right\} + [\mathbf{M}(V)]_0 \times \left\{ \mathbf{h}(t) + \frac{H_A}{M_0} \eta_K(t) \hat{\theta}_K \right\}. \quad (5)$$

To make the equation (5) simpler we have defined  $\eta_K(t) = \boldsymbol{\eta}(t)\cdot\hat{\theta}_K$  and  $M_K^0(V) = [\mathbf{M}(V)]_0\cdot\hat{\theta}_K$ . Also, in equation (5) we have omitted the nonlinear terms as well as the term that determines the equilibrium configuration  $[\mathbf{M}(V)]_0 \times \left\{ \frac{H_A}{M_0} M_K^0(V) \hat{\theta}_K + \mathbf{H}_0 \hat{z} \right\}$  which is zero. This later condition leads to

$$H_0 \sin \theta + H_A \left[ v(\sin \theta \cos \theta \cos 2\theta_K) - \frac{\sin 2\theta_K}{2}(\cos^2 \theta - \sin^2 \theta) \right] = 0 \quad (6)$$

which allow us to obtain the equilibrium position of  $\mathbf{M}(V)$  (the angle  $\theta$  between  $\mathbf{M}(V)$  and the  $z$ -axis). In equation (6)  $\theta_K$  is the angle between  $\hat{\theta}_K$  and the  $z$ -axis, we wrote  $M(V) = M_0 V/V_0$  and we defined the dimensionless parameter  $v = V/V_0$ . A bit of algebra allow us to show that the equilibrium positions of the magnetic moments of the grains are given by the solution of the polynomial equation

$$v \sin 2\theta_K \xi^4 + 4 \left( v \cos 2\theta_K - \frac{H_0}{H_A} \right) \xi^3 - 6v \sin 2\theta_K \xi^2 - 4 \left( v \cos \theta_K + \frac{H_0}{H_A} \right) \xi + v \sin 2\theta_K = 0. \quad (7)$$

with  $\xi = \text{tg}(\theta/2)$ .

We assume  $\boldsymbol{\eta}(t) = (\hat{x}\eta_x + \hat{y}\eta_y + \hat{z}\eta_z) \exp(-i\omega t)$  to obtain, from equation (5):

$$\begin{aligned} -i\omega\eta_x/\gamma &= [H_0 + H_A v \cos(\theta_K - \theta) \cos \theta_K] \eta_y - M_0 v \cos \theta h_y \\ -i\omega\eta_y/\gamma &= -[H_0 + H_A v \cos(\theta_K - \theta) \cos \theta_K] \eta_x \\ &\quad + H_A v \cos(\theta_K - \theta) \sin \theta_K \eta_z + M_0 v \cos \theta h_x \\ &\quad - M_0 v \sin \theta h_z \\ -i\omega\eta_z/\gamma &= -H_A v \cos(\theta_K - \theta) \sin \theta_K \eta_y + M_0 v \sin \theta h_y. \end{aligned} \quad (8)$$

After some algebraic manipulations these equations can be rewritten to read:

$$\begin{pmatrix} \eta_x \\ \eta_y \\ \eta_z \end{pmatrix} = \Delta \begin{pmatrix} \frac{\omega_{xx}^2}{M_0} & i\omega v \cos \theta & -\omega_{xz}^2 \\ -i\omega v \cos \theta & \frac{(\omega_{xx}^2 + \omega_{zz}^2)}{M_0} & i\omega v \sin \theta \\ \frac{-\omega_{xz}^2}{M_0} & i\omega v \sin \theta & \frac{\omega_{zz}^2}{M_0} \end{pmatrix} \begin{pmatrix} h_x \\ h_y \\ h_z \end{pmatrix} \quad (9)$$

where

$$\begin{aligned}\Delta &= \frac{M_0}{\Omega_0^2 - \omega^2}, \\ \omega_{xx}^2 &= \gamma^2 M_0 v \cos \theta [H_0 + H_A v \cos \theta_K \cos(\theta_K - \theta)], \\ \omega_{zz}^2 &= \gamma^2 v^2 M_0 H_A \sin \theta \sin \theta_K \cos(\theta_K - \theta), \\ \omega_{xz}^2 &= \gamma^2 v^2 M_0 H_A \cos \theta \sin \theta_K \cos(\theta_K - \theta), \\ \text{and} \\ \Omega_0^2 &= \gamma^2 [H_A^2 v^2 \cos^2(\theta_K - \theta) + \\ & 2H_0 H_A v \cos \theta_K \cos(\theta_K - \theta) + H_0^2].\end{aligned}\quad (10)$$

Therefore, from equation (9) we can write  $\eta_i(V) = \sum_j \chi_{ij}(V) h_j$  where  $\chi_{ij}(V)$  is the correspondent element of the  $3 \times 3$  matrix displayed in the right hand side of the equation (9).

Then, we may define the effective permeability tensor  $\langle \mu_{ij} \rangle = \delta_{ij} + 4\pi \langle \chi_{ij} \rangle$  where the effective magnetic susceptibility  $\langle \chi_{ij} \rangle$  is given by:

$$\langle \chi_{ij} \rangle = \int_0^\infty \chi_{ij}(V) f(V) dV. \quad (11)$$

With the magnetic permeability on hand one may estimate several physical properties. We chose the magnetoimpedance to illustrate our calculation. This property, in the low frequency limit, is quite well described by the skin depth model [11,12] which has a simple dependence on the magnetic permeability. In the next section we present the calculation of the skin depth for the system described above.

### 3 The skin depth

As mentioned before the approach presented here should be reliable to analyze physical properties that do not “see” individual parts of the system. Instead, they are the response of a great number of particles. Therefore, in the limit where the approach is reliable (low frequency limit), we may neglect the displacement current to write Maxwell’s equations as:

$$\nabla \times \mathbf{H} = \frac{4\pi}{\rho c} \mathbf{E} \quad (12)$$

$$\nabla \times \mathbf{E} = -\frac{1}{c} \frac{\partial \mathbf{B}}{\partial t} = i \frac{\omega}{c} \bar{\mu} \cdot \mathbf{H} \quad (13)$$

where  $\rho$  is the conductivity,  $\omega$  is the frequency of the ac field. Therefore, in this limit we have:

$$\begin{pmatrix} \frac{i\rho(k_\perp c)^2}{4\pi\omega} + \mu_{xx} & \mu_{xy} & \mu_{xz} \\ \mu_{yx} & \mu_{yy} & \mu_{yz} \\ \mu_{zx} & \mu_{zy} & \frac{i\rho(k_\perp c)^2}{4\pi\omega} + \mu_{zz} \end{pmatrix} \begin{pmatrix} h_x \\ h_y \\ h_z \end{pmatrix} = 0 \quad (14)$$

where  $k_\perp$  is the component of the wavenumber perpendicular to the surface of the material. The system of equations above has a non-trivial solution if

$$k_\perp^4 - i \frac{4\pi\omega}{\rho c^2} B k_\perp^2 - \left[ \frac{4\pi\omega}{\rho c^2} \right]^2 C = 0, \quad (15)$$

where  $B = \mu_{xx} + \mu_{zz} - \frac{|\mu_{yz}|^2 + |\mu_{xy}|^2}{\mu_{yy}}$  and  $C = \mu_{xx}\mu_{zz} - |\mu_{xz}|^2 - \frac{\mu_{xx}|\mu_{yz}|^2 + \mu_{zz}|\mu_{xy}|^2 + 2\mu_{xy}\mu_{xz}\mu_{yz}}{\mu_{yy}}$ . We define  $k_\perp = \beta + i/\delta$ , where  $\delta$  is the skin depth which after some algebra can be written as:

$$\delta = 2 \left( -2\Re(k_\perp^2) + 2\sqrt{\Re(k_\perp^2)^2 + \Im(k_\perp^2)^2} \right)^{-1/2} \quad (16)$$

with

$$\Re(k_\perp^2) = \begin{cases} 0, & \text{if } B^2 > 4C \\ \frac{2\pi\omega\sqrt{B^2-4C}}{\rho c^2}, & \text{if } B^2 < 4C \end{cases} \quad (17)$$

and

$$\Im(k_\perp^2) = \begin{cases} \frac{2\pi\omega[B+\sqrt{B^2-4C}]}{\rho c^2}, & \text{if } B^2 > 4C \\ \frac{2\pi\omega B}{\rho c^2}, & \text{if } B^2 < 4C. \end{cases} \quad (18)$$

Since the magnetoimpedance is proportional to  $1/\delta$ , in the following section we analyze the behavior of this quantity with the width of the grain volume distribution and strength of the static external field. Our interest is to study the combined effects of the width of the grain volume distribution and strength of the static external field.

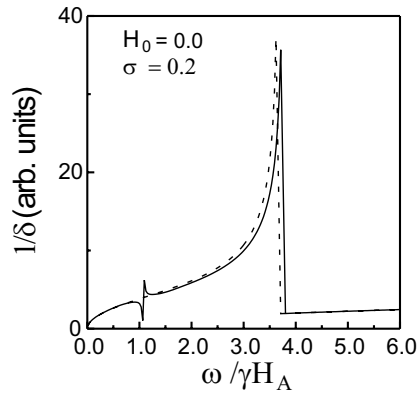
## 4 Numerical results

The calculations presented in the previous sections can be used to obtain numerical results for any geometry. However, to have more precise information of the influence of the combined effect of the external dc field and the distribution of the volume of the grains on the physical property under investigation, in the following we consider the external magnetic field parallel to the anisotropy direction.

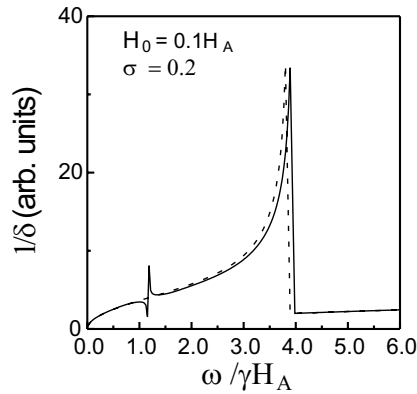
All quantities are depicted in dimensionless or arbitrary units since our goal is to show the relative modification of the property due to the collective behavior.

In Figures 1 to 6 we display the behavior of the inverse of the skin depth and one of the components of the magnetic permeability tensor (we chose  $\mu_{xx}$ ) for different values of the magnetic field and width of the grain volume distribution. For the sake of comparison we also plot in each figure (dashed curve) the behavior of the same property for a homogeneous material with the same physical parameters.

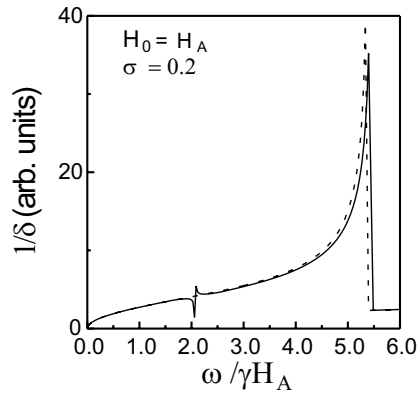
From Figures 1–3 (all of them are for  $\sigma = 0.2$ ) one can see that the presence of an external dc magnetic field modifies the frequency region where the magnetoimpedance has a singular behavior (as mentioned above, the magnetoimpedance is proportional to the inverse of the skin depth). Note that the region where is observed the biggest variation of the magnetoimpedance (which is due to the behavior of the product of the  $\mu_{ij}$  elements that appear in  $B$  and  $C$ ) moves to higher frequency when the intensity of the dc magnetic field is increased. The fluctuation of the magnetoimpedance around the averaged resonance frequency  $\Omega_0(V_0)$  is almost the same for all the fields investigated. The field dependence of the position of the region



**Fig. 1.** Inverse of the skin depth for  $H_0 = 0$  and  $\sigma = 0.2$ . The dashed line shows the result for a homogeneous system with the same physical parameters.



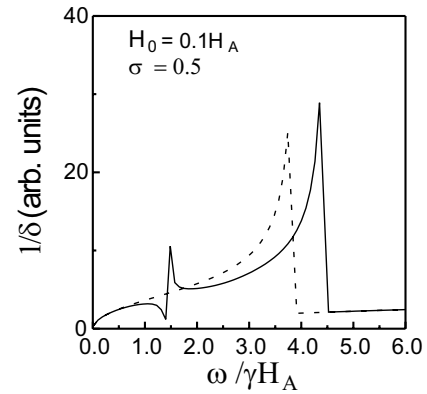
**Fig. 2.** The same of Figure 1 for  $H_0 = 0.1H_A$ .



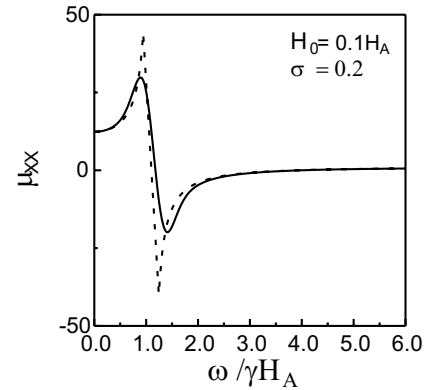
**Fig. 3.** The same of Figure 1 for  $H_0 = H_A$ .

where the abrupt behavior of  $1/\delta$  occurs is the only remarkable feature observed in these figures. It should be noted that in all of them the qualitative behavior of the magnetoimpedance is preserved.

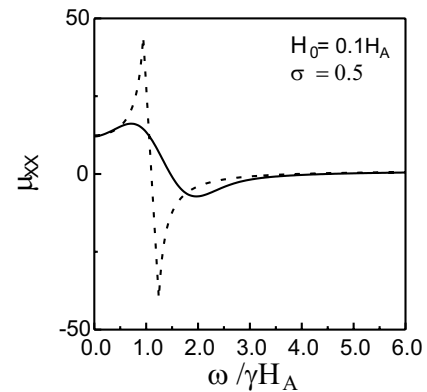
However, when we compare the results depicted in Figure 2 ( $H_0 = 0.1H_A$  and  $\sigma = 0.2$ ) with those in Figure 4 ( $H_0 = 0.1H_A$  and  $\sigma = 0.5$ ) we can see that the width of the grain volume distribution affects not only the position but also the form of the magnetoimpedance in the frequency region where it displays a singular behavior. It is also observed notable modifications in the shape of curves



**Fig. 4.** Behavior of the inverse of the skin depth for a magnetic field  $H_0 = 0.1H_A$  and  $\sigma = 0.5$ . The influence of the width of the volume distribution on the physical property can be seen when this figure is compared with Figure 2.



**Fig. 5.** The component  $\mu_{xx}$  of the permeability tensor for  $H_0 = 0.1H_A$  and  $\sigma = 0.2$ .



**Fig. 6.** The same of Figure 5 for  $\sigma = 0.5$ . Compare with the result for  $\sigma = 0.2$  to see the effect of the width of the volume distribution.

when it is compared with the one obtained for homogeneous system. Similar results can also be seen in Figures 5 and 6 where we show the frequency dependence of  $\mu_{xx}$  for different values of the external dc magnetic field. For homogeneous systems we only observe a very pronounced modification of the magnetic permeability near the resonance frequency. However, for a finite value of the grain

volume distribution width ( $\sigma = 0.2$ , for example) makes  $\mu_{xx}$  smoother in the neighborhood of the resonance frequency. By further increasing  $\sigma$  leads to an almost flat  $\mu_{xx}$  curve, corresponding to the superposition of resonance frequencies  $\Omega_0(V)$  distributed in a wide interval.

## 5 Final comments and conclusions

The results presented in this paper suggest that the width of the grain volume distribution can be used as an additional parameter to obtain a desired property of a sample composed by a collection of nanoparticles. It should be remarked that, if the material have its intrinsical physical properties with different dependence on the volume, the volume distribution of the particles will be of fundamental importance to have the sample with a specific property. In other words, specially in the case where the dependence on the volume of the physical properties of the grains is nonlinear, the volume distribution will play a fundamental to grow samples with a specific characteristic.

However, inter-particles interaction will be always present in these systems and have to be take into account for high density. This interaction should modify the results presented here and the main effect should be to smooth the response of the system to the external driving force since it should work as the viscosity in a fluid.

In this paper we are not aware of some specific material or experiment, but we hope this theoretical work encourages experimentalist to make experiments to confirm the results predicted here.

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