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Contributions to study of ferromagnetic resonance in ferromagnetic particle systems with interactions

-SUMMARY-

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INTRODUCTION

An important category of magnetic measurements is essentially based on the superposition of a direct current (DC) field with an alternative (AC) field of small amplitude usually perpendicular to the dc field. In this way the shape of the energy minimum state determined by the dc field is examined.

Reversible susceptibility with the important case of transverse susceptibility (TS) [1, 2], magnetoimpedance measurements [3] and the very important case of ferromagnetic resonance[4] can be given as examples of this type of measurements. In DC/AC field measurements it is assumed that the oscillations of the magnetic moment around the minimum of the free energy given by the DC field are harmonic (the theories work within the linear approximation) The increase of the amplitude of alternating field causes higher harmonics of the induced signal, i.e. the transition to the nonlinear regime.

Recently, nonlinear phenomena in giant magnetoimpedance (GMI) measurements were also investigated. The resonance frequency dependence of the ac field amplitude, the influence of frequency on the critical curves, the dynamic hysteretic loop, ac field amplitude dependence of the coercive field and the chaotic behavior of the induced signal were studied [5].

To analyze the induced signal linearity, the Fourier trigonometric series are often used. In [6] the susceptibility is measured as the amplitude ratio of the first harmonic of the induced magnetization along a given direction and that of the ac field.

In [7] the interaction effects between the ferromagnetic system particles in the exchange interaction case are studied. Here the transverse susceptibility for the ferromagnetic and antiferromagnetic cases and the damping constant influence are calculated. For systems with magnetostatic interactions the distance between particles,the influence on the magnetization components and switching processes, was analyzed [8]

The synchronization is one of the characteristic processes of the nonlinear oscillators. In the ferromagnetic particles system in the ac/dc field measurements, the periodical variations of the magnetization components around the equilibrium directions can be considered oscillations to which the specifically synchronization study models can be applied. A general method for synchronization study is using the Kuramoto model [9]. We implemented this algorithm in our study.

In this thesis we focused on the magnetization processes of a Stoner-Wohlfarth particles system both without interactions and in the presence of the magnetostatic interactions, around the ferromagnetic resonance and without the temperature influence. To determine the equilibrium magnetic moments directions, the micromagnetic Landau-Lifshitz method is used. In the condition of resonance, the linear/nonlinear regime limit and the oscillations synchronization of the magnetic moments are analyzed.

I. THE FERROMAGNETIC PARTICLE DESCRIBED BY STONER – WOHLFARTH MODEL

The model consists of a material composed of ellipsoidal ferromagnetic identical particles without interactions where each particle is considered single domain, having uniaxial anisotropy, magnetostatic and anisotropy energy. This simple case represents an assembly in which the anisotropy axes of all the particles are headed in the same direction, being reduced at the case of a single particle, called Stoner-Wohlfarth particle. This is uniformly magnetized with the magnetization of saturation.

The stable orientations of the magnetic moment are due to the energetic equilibrium between the energy of the exterior field and the anisotropy energy. Imposing the derivatives of first and second order of the total free energy density to get cancelled, the equations of the equilibrium and stability lines are determined[10]:

$$(e) H_x = H_z tg\theta + H_K \sin\theta$$
$$(s) H_x = -\frac{H_z}{tg\theta} - H_K \frac{\cos 2\theta}{\sin\theta} \quad (1.1)$$

where θ is the angle between easy axis and external field direction (with this components H_x, H_z) and H_k is the anisotropy field.

These two equations represent the geometric place of the critical field which separates in the (H_z,H_x) plan the region where the function describing the free energy have two minima, of the region where this function has one minimum only. By solving these equations, the critical field equation is obtained.

$H_x^{2/3} + H_y^{2/3} = H_k^{2/3}$, astroid in (H_z, H_x) plan (1.2)

The algorithm to determine the magnetization's orientation at a state of equilibrium for a known value of the exterior field, is the following:

- the field is considered in the (H_x, H_z) plan with the origin in the center of the coordinate system;
- all the tangent lines to the astroid which pass through the peak of the magnetic field vector are traced;
- the tangent lines for which the condition of stability is respected are determined;
- the slope of the magnetization at equilibrium will be equal to the one of the tangent lines to the astroid which respects the stability condition.

To calculate the magnetic hysteresis loop of the single domain particle described by the Stoner-Wohlfarth model there needs to be found the positions of equilibrium corresponding to each value of the external field, then calculated the projection's value of the magnetization vector on the direction of the applied field[10].

The magnetic susceptibility is defined as the ratio between the magnetization induced to the sample and the variation of the inductive magnetic field [1]. The used method to determine the components of the tensor is called the method of the transverse susceptibility (TS) and it is a method to determine the anisotropy in ferromagnetic nanoparticle systems. It consists of the simultaneous appliance of the two magnetic fields:

- a static field- the parallel field (on the Oz direction)
- the second one, an alternative low magnetic field- the transverse field, on perpendicular direction on the first one.

The parallel component (***R**eversible **P**arallel **S**usceptibility* -RPS) is measured on the direction of the static applied field, while the transverse components (***R**eversible **T**ransverse **S**usceptibility*- RTS) are measured on the two perpendicular directions. When *Oz* is the DC field direction, the parallel component of the RS is expressed by:

$$\chi_P = \frac{dM_z}{dH_z}, H_x = H_y = 0 \tag{1.4}$$

and the perpendicular components RTS are expressed by:

$$\chi_{t1} = \left(\frac{dM_x}{dH_x}\right)_{H_y=0}, H_y = 0, \chi_{t2} = \left(\frac{dM_y}{dH_y}\right)_{H_z=0}, H_x = 0 \tag{1.5}$$

The TS method in the Stoner-Wohlfarth model is the start base in understanding the ferromagnetic particle systems, necessary for the onset of the studies presented in the next chapters.

II. FERROMAGNETIC RESONANCE. THE COMPLEX MAGNETIC SUSCEPTIBILITY

When a ferromagnetic sample is placed in a magnetic field, the magnetic moment of each particle will effectuate a motion of precession around the applied field, which in the presence of loss will be attenuated. The theoretical description of the ferromagnetic resonance signal is based on the analysis of the free total energy density of a single domain particle in the equilibrium position. The static analysis of this problem is known as Stoner-Wohlfarth model [11]. It gets started with Landau-Lifshitz-Gilbert(LLG) equation written in the Landau-Lifshitz form, which describes the magnetization processes taking place through processes of coherent rotation, [2, 12]:

$$\frac{d\vec{M}}{dt} = -\mu\gamma\left(\vec{M}\times\vec{H}\right) - \mu\frac{\gamma\alpha}{M}\left(\vec{M}\times(\vec{M}\times\vec{H})\right) \tag{2.5}$$

where γ is the gyromagnetic ratio , α is the phenomenological damping constant and $\vec{H}$ the effective field

The advantage of using the LLG equation to determine the equilibrium angles, especially in the case of a multiple coupled particles system, is that due to the fact that the free energy of the system has multiple minima, the evolution of those is described correctly by the dynamic equations; the method of minimization does not guarantee the correct determination of a state which corresponds to the energetic minimum.

The dynamics of the $\vec{M}$ magnetization vector consists of a precession motion, which, due to the presence of a small alternative signal h_{AC}, is subjected to small angular deviations $\Delta\theta$,$\Delta\varphi$ around the equilibrium position θ_0, φ_0. Solving the LLG equation in spherical coordinates we obtain the relations for the resonance frequency and the linewidth of the spectral line as in [4].

The complex susceptibility tensor in spherical coordinates, due to the ac field (h_θ, h_φ) is defined by the following relation [12, 13]

$$\begin{bmatrix} M_\theta \\ M_\varphi \end{bmatrix} = \overset{=}{\chi}\begin{bmatrix} h_\theta \\ h_\varphi \end{bmatrix} \Leftrightarrow \begin{bmatrix} M\Delta\theta \\ M\sin\theta_0\Delta\varphi \end{bmatrix} = \begin{bmatrix} \chi_{11} & \chi_{12} \\ \chi_{21} & \chi_{22} \end{bmatrix} \cdot \begin{bmatrix} h_\theta \\ h_\varphi \end{bmatrix} \tag{2.9}$$

In our study we calculated the components of the complex susceptibility tensor firstly in polar coordinates and then in Cartesian coordinates. The obtained relations are very general and can be applied for different types of anisotropy, different values and orientations of the applied field, if the expression of the free energy density is known.

For the particular case of the particle in Stoner-Wohlfarth model and by using the general relations of the transverse components of the susceptibility, we find again the reversible susceptibility formulas from Aharoni model [1], but this time the deduction is based on the LLG equation. This proves that at the limit of low frequency, applying the FMR theory the reversible susceptibility from Aharoni model is obtained.

While the Aharoni theory [1] offers information about the real part of the susceptibility only, the study of the transverse susceptibility by using the LLG equation is more general as it is effectuated by the study of magnetization dynamics in the assembly of AC/DC fields. If in the case of ferromagnetic resonance(FMR) there are measured absorptions of resonance in ferromagnetic materials at high frequencies [4], in TS method there is measured the variation of the magnetization on the ac field direction. To determine the equilibrium angles of the magnetization direction, we will not use the critical curve method, but we will integrate the Landau Lifshitz equation (the micromagnetic method).

So, we calculated the DC field dependencies of the real and imaginary parts of the transverse susceptibility, for some values of the low frequency of the alternative field when the easy axis direction are same with a ac field direction. The results of these simulations indicate that the maximums of the susceptibility are associated with the ferromagnetic resonance at zero frequency. One of the deficiencies of the theoretical TS method is that it cannot be used for the study of the AC field amplitude influence on the magnetization processes.

III. THE MICROMAGNETIC STUDY OF THE PROCESSES OF MAGNETIZATION OF STONER-WOHLFARTH PARTICLE

By calculating dynamic equilibrium angles of the magnetization vector direction in the presence of the alternative field, we studied the forced oscillations of the magnetic moment around the static equilibrium position determined by the DC field.

The AC field will produce a dynamic component of the magnetization due to the variations of polar and azimuthal angles around the static equilibrium angles. The (θ, φ) angles which determine the magnetization direction will be called dynamic equilibrium angles and with these values we calculated the components of the magnetization vector:

$$M_x = M_s \sin\theta \cos\varphi, M_y = M_s \sin\theta \sin\varphi, M_z = M_s \cos\theta \tag{3.1}$$

where M_s is the saturation magnetization of the particle.

The necessary component in the study of magnetization processes in the AC/DC fields is on the direction of the alternative field $m_x = M_x / M_s$. This is influenced by the amplitude and the frequency of the AC field.

The component of the dynamic susceptibility on the AC field direction is defined as follow: [6, 14]:

$$\overline{\chi}_{xx} = \frac{d\overline{m}_x}{d\overline{h}_{AC}} \tag{3.2}$$

In our study we have implemented two techniques to evaluate the susceptibility tensor in AC/DC measurements. The first step in both methods is to determine the equilibrium state in the presence of the DC field, θ_0, φ_0. By using these values in the general susceptibility formula [12] we obtain a first value for susceptibility tensor which can be named “theoretical” value (analytical method). The same tensor can be calculated numerically by using the (3.2) equation. This means that we introduce the AC field in the LLG algorithm and study the forced oscillations of the moment around the equilibrium state defined by the DC field. After a short transitory process the moment follows periodic oscillations. The amplitude of these oscillations is used to evaluate the susceptibility tensor (numerical method).

When the amplitude of the AC field is small enough we expect the difference between the two results to be very small. When the AC field amplitude is higher we expect the results to be different. The numerical method is able to give results even in the nonlinear range while the first is limited to the linear regime. We have compared the results for small AC field amplitudes only as a test for the numerical algorithm[15]. The differences between the results obtained by the two methods are less than 3.5%. So, the test shows that the method using numerical model reproduces almost exactly the same results as in the linear analytical case.

We analyzed the induced signal(the dynamic magnetization component) by using the trigonometric Fourier series [16]. In the linear regime (at small values of the AC field amplitude), the signal is sinusoidal and the fundamental mode energy is much greater than the sum of the energies of superior modes. Once the AC field amplitude increases, the values of the energy of the superior modes increase also, causing the transition into the nonlinear regime. The energy modes of the oscillation depends on the Fourier coefficients values. As the AC field amplitude increases, the higher-order coefficients increase starting with the second order coefficient, and the transition in the nonlinear regime occurs.

To describe the induced signal linearity, we have used the Total Harmonic Distortion parameter (THD). THD is defined as the ratio between the sum of the powers of the higher order harmonic and the power of the fundamental mode [17]. Using the AC field amplitude influence on the DC field dependence of the resonance frequency, we calculated the THD value which is the criteria for the transition towards the nonlinear regime: $THD = 2\%$ Based on this criteria, we calculated the DC field dependence of AC field amplitude corresponding to the linear/nonlinear regime limit[15].

We have shown that the AC field amplitude limit of the linear regime is lower at resonance and at switching points. The increase of the phenomenological damping constant or the AC field frequency cause the increase of the AC field amplitude corresponding to the transition in the nonlinear regime. This study offers a quantitative method to identify regions of nonlinear behavior of magnetic moments under simultaneous AC and DC field.

IV. THE MICROMAGNETIC STUDY OF THE MAGNETIZATION PROCESSES OF A STONER-WOHFART PARTICLES SYSTEM WITH MAGNETOSTATIC INTERACTIONS

This chapter presents the analysis of the magnetostatic interactions effects in the case of a single domain ferromagnetic particles system with uniaxial anisotropy by using the same micromagnetic method. The analysis of the effects of these interactions will be in the transverse susceptibility and the synchronization of the magnetic moments oscillations of the particles.

Initially, the system is considered to be two single domain ferromagnetic particles with uniaxial symmetry, which can be approximated with two magnetic dipoles with magnetostatic interactions [18].

The algorithm used to calculate the equilibrium angles of the magnetic moments of the particles is the same LLG algorithm by adding the interaction field. The particles system is coupled by the effective field, as the terms corresponding to each particle are not independent. The coupling between particles depends on the distance between them *d*, the coupling coefficient is considered to be $1/4\pi d^2$. Using the LLG algorithm we studied the transverse susceptibility of the system and the influence of the coupling on the magnetization processes.

We focused more on the case when the alternative field is applied on a single particle, analyzing the transfer of energy to the other one. The magnetization component of this particle will be the forced oscillations with a frequency determined by the AC filed, after the transitory regime. By interactions, a quantity of energy will be transferred from the first particle to the second one, which will have also a forced oscillation. We analyzed a series of characteristics of the magnetization’s oscillation for the second particle (amplitude, phase, frequency, duration for the transitory regime). We performed a study on the conditions in which the magnetization’s oscillations for each particle are produced with the same frequency and also with the difference of phase constant in time. In other words, the particles synchronize.

The *synchronization* is the process in which interactive, oscillating objects affect each other’s phases so that they spontaneously lock to a certain frequency or phase [19]. One of the most used models to study the synchronization phenomenon is the Kuramoto model[9]. The synchronization studies using the Kuramoto model are performed in the systems with large number of oscillators nearly identical, with different natural frequencies. The particles interact with each other and the synchronization result is expressed by a global parameter called the order parameter.

Our study is focused on the synchronization of the forced oscillations of a ferromagnetic particles system with uniaxial anisotropy and magnetostatic interactions[20] . For simplicity we consider a 1D chain of the ferromagnetic particles which are located at the same distance from each other. The analogy between the values of the two models is presented in the following table:

The parameter corresponding to the Kuramoto oscillators system	**The parameter corresponding to the S-W particles systems**
The phases	The azimuthal angles of dynamic equilibrium φ_i
The intensity of the coupling	The $1/d^3$ parameter
The intrinsic frequency of the particle	The *K1*constant of anisotropy of the S-W particle

For the synchronization study of the oscillations of the magnetic moments, first we calculated the phase's particles distributions by LLG method, then we used the Kuramoto algorithm to calculate the order parameter. We also evaluated the AC field amplitude and damping influence on the synchronization processes.

We have shown that the Kuramoto algorithm can be applied within certain limits for the study of the synchronization of the ferromagnetic particles system with magnetostatic interactions. In this way we determine the link between the distance between particles corresponding to the synchronization range and common frequency of the particles synchronized. The model limit is the appearance of the non-periodical forced oscillations due to the high ac field amplitude when these oscillations can become chaotic. Also, we observe that the increase of the phenomenological damping constant determines the decrease of the threshold of the synchronization start.

GENERAL CONCLUSIONS

During the scientific activity, we focused on three principal directions of studying: first of all, to understand the used methods I synthetized the theoretical aspects from the TS and LLG models, verifying the connection between the results obtained by proper simulations, also I built and tested an numerical calculations algorithm for the dynamics magnetization components of the particles; secondly I made a quantitative evaluation of the limit between the linear and nonlinear regimes of magnetic moments oscillation of the particles in ac/dc field; thirdly I implemented a model of studying the synchronization of the dynamics magnetizations components oscillations for a ferromagnetic particles system with magnetostatic interactions.

Thereby, I analyzed and compared the two methods of determining the direction of the magnetic moment equilibrium of a particle by using the critical curve and Landau-Lifshitz methods. Using the Landau-Lifshitz model I obtained the simulations of the resonance frequency and of the transverse susceptibility components. These were particularized at low frequency and led to the reverse susceptibility obtained by using the Aharoni theory. So, we concluded the generality of the Landau-Lifshitz method which will be used for all the simulations. The generality of the algorithm consists of the fact that it can also be used to study the influence of the ac field amplitude on the particles magnetization (and the transition in the nonlinear regime).

The increase of the ac field amplitude determines the perturbation of the signal's linearity by the appearance of the superior harmonics. The analysis of the induced signal's linearity was realized by using the Fourier series techniques. In the case of ferromagnetic resonance experiments, we applied this technique to analyze the linearity of the dynamic magnetization component on the ac field direction. As the signal linearity is perturbed, the superior order Fourier coefficients get higher. We studied the signal linearity by the total harmonic distortion parameter (THD). We created a criteria of transition in the nonlinear regime, calculating the limit value for the ac field amplitude which determines this transition.

In the last part of the thesis, we proposed a study of the synchronization of the magnetic moments oscillations of a ferromagnetic particle system in ac/dc fields. We implemented the algorithm described by the Kuramoto model of studying the synchronization of a high number of oscillators, by calculating the order parameter of the system. The type of the studied synchronization is a forced by the alternative applied field one and the results offer information about the minimal values of distances between the particles of the system necessary to begin the partial synchronization of the magnetic moments oscillations. Also, the results inform about the influence of the damping and the ac field amplitude on the level of synchronization and about the fact that in the resonance regions, the synchronization takes place very slow or not at all.

In conclusion, we consider that the studies which were presented can be continued for the cases in which due to an enough high amplitude of the alternative applied field, the induced oscillations become chaotic, and also, the extinction to the 2D particular systems.

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