

## Hysteresis Characteristics of Interacting Fe/Fe<sub>3</sub>O<sub>4</sub> Core/Shell Nanoparticles

Anisimov Sergei<sup>a</sup>, Afremov Leonid<sup>b</sup>, Iliushin Iliac<sup>c</sup>

Department of Theoretical and Experimental Physics, Far Eastern Federal University, Vladivostok, Russian Federation

<sup>a</sup>e-mail: ahriman25@gmail.com, <sup>b</sup>afremov.ll@dvfu.ru, <sup>c</sup>futed@gmail.com

**Abstract**— In this work, we show our results of the theoretical study of the hysteresis characteristics of a system of interacting Fe/Fe<sub>3</sub>O<sub>4</sub> core/shell nanoparticles. We show that increase of the volumetric concentration of nanoparticles lead to decrease of the coercive field and remanent saturation magnetization. Results are in a good agreement with an experimental data.

**Keywords**— core/shell; coercive field; remanence saturation magnetization; magnetic interaction

### I. INTRODUCTION

Magnetostatic interactions are an important factor affecting the process of magnetization of the assembly of nanoparticles. For example, in an equilibrium distribution of  $N$  magnetic grains of size  $a$  in a nonmagnetic matrix, the field from one of the particles felt by the neighbor particle can be estimated as followed:  $H_m = 2M_s Na^3/R^3$ , where  $R$  – mean distance between particles,  $c = Na^3/R^3$  – volumetric concentration of the magnetic in the system. Usually, the magnetic record materials are made from magnetics with a high values of spontaneous magnetization  $M_s \sim 500 \div 1500$  G. Thus, at  $c \sim 0.1 \div 0.3$ , the magnetostatic interaction can significantly affect the magnetization of the low-coercivity nanoparticles. Many works are devoted to the theoretical [1-5] and experimental study [6-9] of magnetic interactions in systems of nanoparticles. In References [1-4], a method of designing the distribution function of the random interaction fields without a limitation on the type of interaction (magnetostatic, direct exchange, RKKY or any other interaction) is shown. Moreover, the distribution function is shown to depend not only on the location of the nanoparticles (dimension of an assembly) [4], but also on the concentration  $c$ . According to [4, 5], at low values of the volume concentration  $c$  of the dipole-dipole interacting magnetic nanoparticles ( $c < 0.1$ ), randomly distributed in the nonmagnetic matrix, random fields of magnetostatic interaction  $h$  are distributed according to Cauchy's law. At  $c > 0.1$ , the field distribution is normal.

In this work, a study of the effect of interaction on the coercive field and the remanent saturation magnetization of the Fe/Fe<sub>3</sub>O<sub>4</sub> core/shell nanoparticles has been carried out within the model of the two-phase nanoparticles [11].

### II. SELF-CONSISTENT EQUATIONS FOR THE MAGNETIZATION OF THE SYSTEM OF CORE/SHELL NANOPARTICLES

As has been noticed in the introduction, at low volume concentration of the magnetic nanoparticles  $c$  ( $c < 0.1$ ),

random fields of magnetostatic interaction  $h$  are distributed according to the Cauchy's law [4, 5]:

$$W_1(h, M, B) = \frac{1}{\pi} \frac{B}{B^2 + [h - \hat{h}(M)]^2}, \quad (1)$$

Where  $\hat{h}(M) = (N-8\pi/5) M(H)$  – mean (the most probable) interaction field. The distribution parameter  $B$  (intrinsic interaction field) and the magnetization of the system of nanoparticles  $M(H)$  are defined by the equations:

$$B = 5c \int [(1-\varepsilon)M_s^{(1)} + \varepsilon M_s^{(2)}] (n_1(t, h) + n_3(t, h)) + \\ |(1-\varepsilon)M_s^{(1)} - \varepsilon M_s^{(2)}| (n_2(t, h) + n_4(t, h)) W_1(h, M, B) dh, \quad (2)$$

$$M(H) = c \int [(1-\varepsilon)M_s^{(1)} + \varepsilon M_s^{(2)}] (n_1(t, H+h) - n_3(t, H+h)) + \\ |(1-\varepsilon)M_s^{(1)} - \varepsilon M_s^{(2)}| (n_2(t, H+h) - n_4(t, H+h)) W_1(h, M, B) dh, \quad (3)$$

Where  $N$  – demagnetization factor of the system along the external field  $H$ .

If  $c > 0.1$ , then the distribution of the interaction fields is normal:

$$W_2(h, M, B) = \frac{1}{\sqrt{\pi} B^2} \exp\left(-\frac{(h - \hat{h}(M))^2}{B^2}\right), \quad (4)$$

Here  $\hat{h}(M) = N M(H)$ , and the magnetization  $M(H)$  is defined by (3) (where necessary to change  $W_1(h, M, B)$  to  $W_2(h, M, B)$ ), and the intrinsic interaction field  $B$  is a solution of the following equation [4]:

$$B^2 = c \int [(1-\varepsilon)M_s^{(1)} + \varepsilon M_s^{(2)}]^2 (n_1(t, h) + n_3(t, h)) + \\ ((1-\varepsilon)M_s^{(1)} - \varepsilon M_s^{(2)})^2 (n_2(t, h) + n_4(t, h)) W_2(h, M, B) dh, \quad (5)$$

Where  $n(t, h) = \{n_1(t, h), n_2(t, h), n_3(t, h), n_4(t, h)\}$  – population vector of the states of two-phase nanoparticle components, of which  $n_i(t)$  can be considered as the probability to find a nanoparticle in one of four states: in the first «(↑↑)» and third «(↓↓)» states – with a parallel orientation of the magnetic moments of both phases, in the second «(↑↓)» and fourth «(↓↑)» states – with an antiparallel orientation of the magnetic moments of the phases (Fig. 1). According to [4], the population vector can be represented in vector form using the matrix exponential:

$$n(t, h) = \exp(w(h)t) n(0) + \int_0^t \exp(w(h)(t-\tau)) d\tau v(h), \quad (6)$$

Here, matrix elements  $w(h)$ ,  $v(h)$  and vectors  $n(t, h)$  and  $n(0)$  are expressed in terms of  $W_{ik}(h) = f_0 \exp(-E_{ik}(h)/k_B T)$  as followed:

$$W_{ik} = \begin{cases} -\sum_{j \neq i, j=1}^l W_{ij} - W_{4i}, & i = k \\ W_{ki} - W_{4i}, & i \neq k \end{cases}, \quad v = \begin{pmatrix} W_{41} \\ W_{42} \\ W_{43} \end{pmatrix},$$

$$n(t) = \begin{pmatrix} n_1(t, h) \\ n_2(t, h) \\ n_3(t, h) \end{pmatrix}, \quad n(0) = \begin{pmatrix} n_{01} \\ n_{02} \\ n_{03} \end{pmatrix},$$

Where  $f_0 = (10^8 \div 10^{10}) s^{-1}$ ,  $E_{ik} = E_{ik \max} - E_{i \min}$ , – the energy barriers separating  $i$ -th and  $k$ -th equilibrium states ( $E_{ik \max}$  – the smallest of the maximal values of the energy separating  $i$ -th and  $k$ -th states,  $E_{i \min}$  – energy of the  $i$ -th equilibrium state).

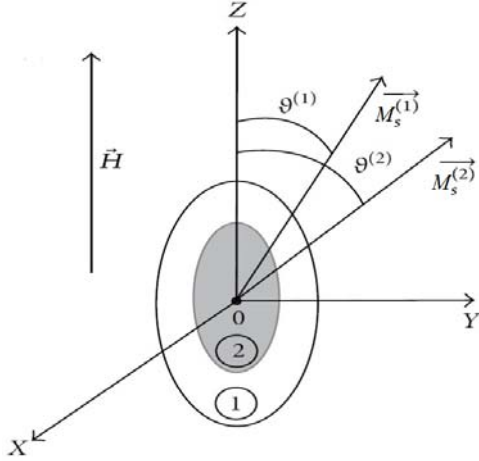


Figure 1. Illustration for the model of core/shell nanoparticles, used in our work [11]. Uniformly magnetized ellipsoidal nanoparticle (phase 1) with volume  $V$  and spontaneous magnetization  $M_s^{(1)}$  contain an ellipsoidal iron core (phase 2) with volume  $v = \varepsilon V$  and spontaneous magnetization  $M_s^{(2)}$ . External field  $H$  is applied along anisotropy axis, which is parallel to the long axes of the ellipsoids.

The system of self-consistent equations (2), (3), (5) along with a formula (6), which estimates the population vector  $n(t, H)$  allows us to calculate the magnetization  $M(H)$  of a system of an interacting core/shell nanoparticles.

### III. HYSTERESIS CHARACTERISTICS OF THE SYSTEM OF Fe/Fe<sub>3</sub>O<sub>4</sub> NANOPARTICLES

The coercive field  $H_c$  and the remanent saturation magnetization  $M_{rs}$  have been estimated from the hysteresis loops, and have been plotted using Equations (1) – (6). Since the system includes nanoparticles with magnetic moments which are susceptible to the thermal fluctuations, the contribution to the magnetization is given by the nanoparticles with a relaxation time  $\tau$  bigger than the measurement time  $t$  ( $\tau \geq t$ ). The calculation assumes that  $t=1$

second. Moreover, we took into account the dependence of the spontaneous magnetization and crystallography anisotropy of the iron and magnetite nanoparticles on the size. Because of the size distribution of the assemblies of synthesized nanoparticles, we have made an averaging of the magnetization  $M(H, b)$  over  $b$ . For the integration of  $M(H, b)$ , the lognormal distribution law has been used: with the mean size value  $\langle b \rangle$  and dispersion  $\sigma$  shown in the work [12].

$$F(B) = \frac{1}{B\sqrt{2\pi\sigma^2}} \exp\left(-\frac{(\log[b/\langle b \rangle])^2}{2\sigma^2}\right), \quad (7)$$

The dependence of the hysteresis characteristics  $H_c$  and  $M_{rs}$  on the intensity of the magnetic interaction in the system of Fe/Fe<sub>3</sub>O<sub>4</sub> are shown on the figures 2 and 3. The increase of the volume concentration  $c$  and the magnetostatic concentration lead to a “smoothing” of the hysteresis loop (Fig. 2), and result in a decrease of the coercive field  $H_c$  and the remanent saturation magnetization  $M_{rs}$  (Fig. 3). It is observed that the magnetostatic interaction has the most significant effect on the coercive field of the sample.

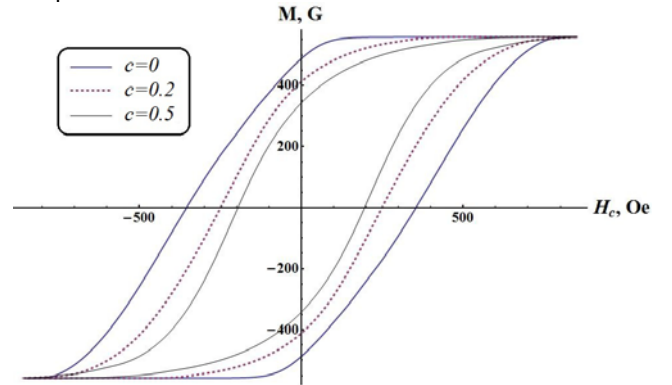


Figure 2. Hysteresis loops of three assemblies of interacting nanoparticles with different concentrations  $c$ .

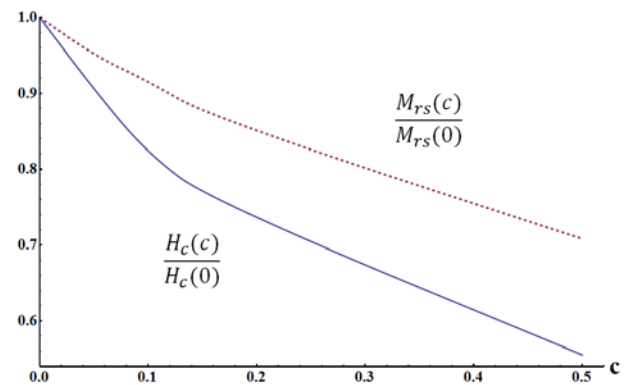


Figure 3. Dependence of the normalized values of the coercive field  $H_c(c)/H_c(0)$  and the remanent saturation magnetization  $M_{rs}(c)/M_{rs}(0)$  on the concentration of spherical magnetic nanoparticles in the sample.

The decrease of the hysteresis characteristics with the growth of the concentration  $c$  of magnetic particles in the

sample is due to the chaotization effect of the random interaction field on the distribution of the magnetic moments of the sample. According to our calculations, an increase of  $c$  leads to the sharp increase of the effective interaction field (parameter of the distribution function on the interaction fields)  $B$  at low concentrations ( $c < 0.1$ ) and to a monotonic increase at  $c > 0.1$  (Fig. 3).

#### IV. CONCLUSION

Modeling of the magnetostatic interaction effect on the hysteresis characteristics carried out within the model we have developed [11] has shown that chaotization effect of the magnetostatic interaction on the magnetic moment distribution lead to decrease of the coercive field and remanent saturation magnetization. Moreover, the increase of the volumetric concentration, and hence the magnetic interaction, lead to significantly larger decrease of the coercive field than remanent saturation magnetization

#### ACKNOWLEDGMENT

This work is supported by grant of Ministry of Education and Science: project № 559.2014.

#### REFERENCES

- [1] V.I. Belokon', K.V. Nefedev, "Distribution function for random interaction fields in disordered magnets: Spin and macrospin glass," *Experimental and Theoretical Physics*, Vol. 93, Issue 1, July 2001, pp. 136-142.
- [2] V.I. Belokon', S.V. Semkin, "Random-field method in the Ising model of a diluted ferromagnet," *Experimental and Theoretical Physics*, Vol. 75, Issue 4, October 1992, p. 680
- [3] L.L. Afremov, P.V. Kharitonov, "About magnetostatic interactions in the assembly of the growing monodomain nanoparticles," *AS USSR, Geophysics №2*, 1988, pp. 101-105.
- [4] L.L. Afremov, V.I. Belokon', Yu.V. Kirienko, K.V. Nefedev, "Magnetic properties of nanodisperse magnets," Vladivostok: Far Eastern Federal University Publishing, 2010, 120p.
- [5] V.P. Sherbakov, "About a distribution function of the molecular fields in the systems with random distribution of the interaction centers," *Magnetic Materials Physics*, Vol. 48(6), 1979, p. 1134.
- [6] J. Garcia-Otero, R. Porto, J. Rivas, "Henkel plots of single-domain ferromagnetic particles," *J. Appl. Phys.*, 87(10), 2000, pp. 7376 – 7381.
- [7] C.R. Pike, A. Fernandez, "An investigation of magnetic reversal in submicron-scale Co dots using first order reversal curve diagrams," *J. Appl. Phys.*, 85, 1999, pp. 6668 – 6676.
- [8] O. Hankel, "Remanenzverhalten und Wechselwirkungen in hartmagnetischen Teilchenkollektiven," *Phys. Status Solidi*, 7, 1964, p. 919.
- [9] R. Proksch, B. Moskowitz, "Interactions between single domain particles," *J. Appl. Phys.*, 75(10), 1994, pp. 5894 – 5902.
- [10] O.A. Goroshko, V.A. Ivanov, I.V. Soppa, "Observation of interaction fields in the assembly of single domain particles," *J. Magn. and Magn. Mater.*, 322, 2010, pp. 3385 – 3390.
- [11] L.L. Afremov, I.G. Ilyushin, "Effect of Mechanical Stress on Magnetic States and Hysteresis Characteristics of a Two-Phase Nanoparticles System", *Journal of Nanomaterials*, (2013), 15 p;
- [12] Maninder Kaur, John S. McCloy, Weilin Jiang, Qi Yao, You Quiang, "Size dependence of inter- and intracluster interactions in core-shell iron-iron oxide nanoclusters," *J. Phys. Chem. C*, 116, 2012, pp. 12875-12885.