Journal of ELECTRICAL ENGINEERING, VOL 66. NO 7/s, 2015, 124-127

MAGNETIZATION PROCESSES OF ULTRA-HARD MAGNETIC NANOCOMPOSITES – MODELING AND SIMULATIONS

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We present models and simulations, based on the Stoner-Wohlfarth and the two-level energetic model, of the magnetization process for high-coercive magnetic nanocomposites. In the investigated system, the magnetic objects with extremely high coercivity are "frozen" after the first magnetization and being a source of the so-called quenched internal magnetic field. A model accounting a presence of randomly oriented internal magnetic field that influences objects contributing to magnetization processes of the composite. The results of simulations are in good agreement with experimental data and confirm correctness of the applied model.

Keywords: hard magnet, modelling and simulations, random field, Stoner-Wohlfarth model

1 INTRODUCTION

Magnetic materials are very important in nowadays technologies. New and continuously increasing requirements can be fulfilled by modern nanostructured magnetic composites containing different phases characterized by different magnetic properties. In the field of hard magnets, interactions between the phases are especially important and can lead to an appearing of new and unique properties [1,2].

Recently, we reported ultra-high coercivity (about 3.6 T) in Fe-Nb-B-Tb type of bulk nanocrystalline alloys prepared by vacuum suction casting technique [3]. The alloys consists nanograins of hard magnetic $Tb_2Fe_{14}B$ phase as well as relatively soft like $TbFe_2$ and Fe phases. Moreover, a specific microstructure including dendrite-like branches with high shape anisotropy can be a source of additional type of magnetic object.

An example of magnetic hysteresis loop for $(Fe_{80}Nb_6B_{14})_{0.88}Tb_{0.12}$ alloy is presented in Fig. 1. In this case the loop is complex, asymmetric, enclosed and different during first and second run. Such shape suggests a presence of soft, hard and ultra-hard magnetic components as was schematically shown in Fig. 2. The objects with extremely high anisotropy are disordered for as cast material (their total magnetisation is equal to zero), however a firs magnetic saturation lids to alignment along external magnetic field direction. Next the ordered objects are "frozen" and they cannot change magnetization, being a source of the so-called quenched internal magnetic field. This field can express magnetic interactions between the objects and their surroundings.

From the both, scientific as well as application point of view, it is a key challenge to understand the role of interactions with ultra-hard phases and consequently, magnetization process in such materials. In this work, we propose the Stoner-Wohlfarth and the two-level energetic model accounting a presence of randomly oriented internal magnetic field that influences objects contributing to magnetization processes of the composite. The aim is to study a role of interactions between magnetically hard and ultra-hard particles as well as qualitative analysis of the applied model in context of the experimental data.

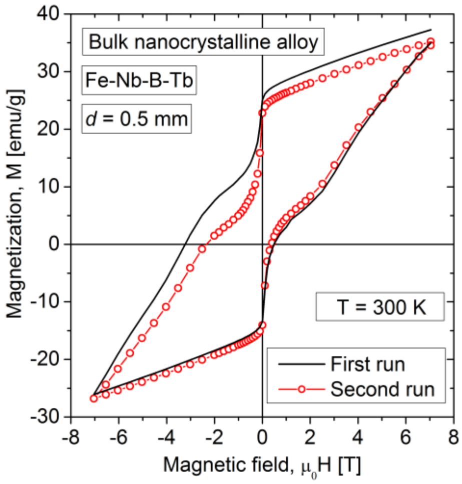

Fig. 1. The firs and the second run of magnetic hysteresis loop for the $(Fe_{80}Nb_6B_{14})_{0.88}Tb_{0.12}$ bulk nanocrystalline alloy.

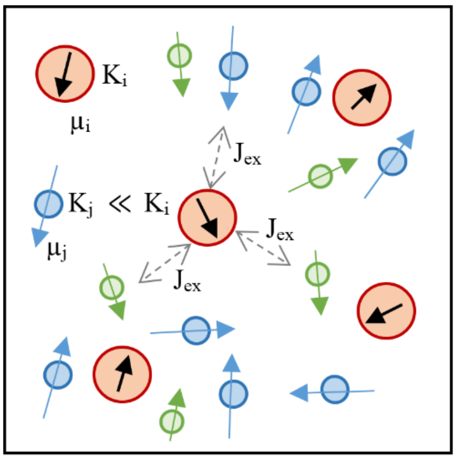

Fig. 2. The schematic presentation of soft (green), hard (blue) and ultra-hard (red) magnetic objects with their magnetic moment μ and anisotropy coefficient K in discussed material.

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ISSN 1335-3632 © 2015 FEI STU

2 THEORETICAL BACKGROUND AND CALCULATON PROCEDURE

The magnetic properties of the no-interacting, single domain nanoparticles (at temperature $T = 0$ K) describes so-called the *Stoner-Wohlfarth (S-W) model* [4]. The key point of the model is the energy calculation of each magnetic object as a function of θ (angle between the applied field and the object axis Z) including magnetocrystalline anisotropy, shape anisotropy and interaction with magnetic field. Figure 3(a) presents three examples of such function for different ratio of magnetic energy and anisotropy coefficient. Base on $E(\theta)$ minimums, the state and in consequently the direction of object magnetisation, can be calculated.

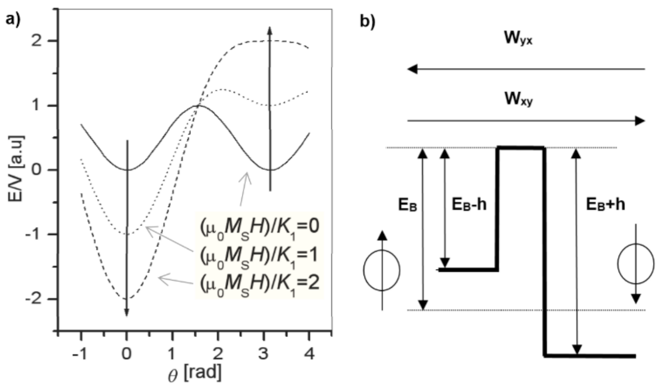

Fig. 3. (a) — an example of $E(\theta)$ dependences based on Stoner-Wohlfarth model, (b) — diagram of the two-level model, [5].

Dynamic of the system in high temperature can be discussed in a frame of co-called *Two-level energetic model* [6]. Two magnetic direction of object corresponds to two states, let say X and Y, separated by the blocking energy E_B and factor h related to energy interaction between magnetic object and magnetic field. Note that, such barrier is asymmetric as shown in Fig. 3(b). The frequency of jumps between states is expressed by factors W_{XY} (from X to Y) and W_{YX}:

$$W_{XY} = W_0 \exp\left(-\frac{E_B - h}{\mathrm{k}_B T}\right), \tag{1}$$

$$W_{XY} = W_0 \exp\left(-\frac{E_B + h}{\mathrm{k}_B T}\right), \tag{2}$$

where W_0 is the jump frequency for $T \to \infty$. Let assume that total amount of magnetic object is a sum of objects in state X and Y, *ie* $N = N_X + N_Y$, then

$$N_X = N\tau W_{YX} + \left(N_{X0} - N\tau W_{YX}\right)\exp\left(-\frac{t}{\tau}\right), \tag{3}$$

where $\tau = 1/(W_{XY} + W_{YX})$ is the time constant, t is the time of measurement and $N_{X0} = N_X(t = 0)$.

Prepared system consists one (in some cases) or two type of sets of spherical magnetic object corresponding to soft and hard magnetic phases. Both of them are characterized by linear distributions of anisotropy coefficient K_1 and K_2 as well as magnetic moment μ_1 and μ_2 for the first and the second phases, respectively. The magnetic objects of ultra-hard phase were not directly included in the simulation, however their impact on surroundings was modeled by additional random field H^{RF} introduced into the system. Finally, the energy of i-th magnetic object can be calculated by the formula

$$E_i = K_i \sin(\theta_0 - \theta)^2 - \mu_i \mu_B \mu_0 (H^{EX} + H_i^{RF}). \tag{4}$$

The values of θ as well as θ_0 are an angle between Z-axis of the system and magnetization axis as well as easy magnetization axis of magnetic object. The model consists uniform distribution of θ_0 from 0 to π for each type of magnetic object. Moreover, H^{EX} is external magnetic field and H_i^{RF} is additional field randomized (during each step) for i-th magnetic object based on normal distribution described by average value $< H^{RF} >$ as well as standard deviation σ_{RF}. It is important to note, that $< H^{RF} >$ starting from 0 at the beginning of hysteresis loop simulation and increasing up to the fixed value (described by $< H^{RF} >$ parameter) for the last point on virgin magnetization curve. Such situation (schematically shown in Figure 4) corresponds to impact of ultra-hard magnetic object, which are ordering during first magnetization and "frozen" after that. Let emphasize, that the introduce of random field to the system supplements the S-W model for interactions between objects, which is not supported by standard S-W approach.

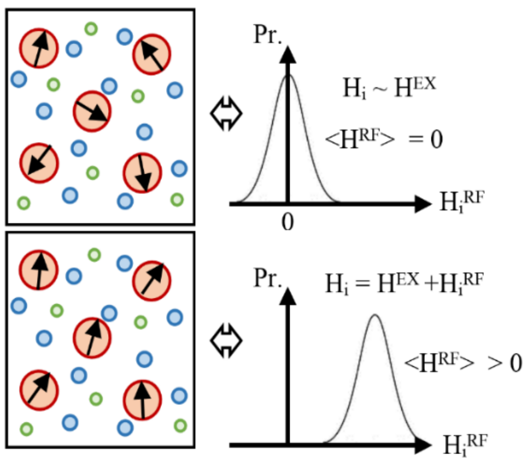

Fig. 4. The correlation between state of ultra-hard magnetic object and random field distribution as well as field H_i "feeling" by i-th simulated object.

Finally, the simulation procedure can be summarized as follows

Step 1. Initialize magnetic object in random state and set random field with $<\boldsymbol{H^{RF}}> = \mathbf{0}$.

Step 2. Calculate energy of each object using formula (4) and random value of $\boldsymbol{H_i^{RF}}$ base on normal distribution.

Step 3. Find minimums of $E_i(\theta)$ and calculate N_X base on eq. (1), (2) and (3).

Step 4. Calculate the average value of magnetic moment $<\mu^i>$ for each object

$$<\mu^i>=\mu_i\mu_B(N_Y\cos\theta_Y+N_X\cos\theta_X) \qquad (5)$$

where $\boldsymbol{\theta_Y}$ and $\boldsymbol{\theta_X}$ are the value of θ for Y and X state energy minimum, respectively.

Step 5. Calculate the total average magnetic moment of the system normalized to maximum value for fully saturated material and update average value of random field $<\boldsymbol{H^{RF}}>$ as well as H^{EX}.

Step 6. Repeat the procedure starting from step 2 for full simulation of hysteresis loop double run at room temperature in $\boldsymbol{\mu_0 H^{EX}}$ range from -1T to +1T including 100 points and 5000 magnetic objects.

3 RESULTS AND DISCUSSION

Figure 5 shows simulated hysteresis loops obtained for one magnetic component and four different combination of random field parameters. In this case all magnetic objects are characterized by anisotropy coefficient K_1 and magnetic moment μ_1 equal to 1 eV and 10^4 μ_B, respectively. As may expect, simulations without additional random field leads to classical – symmetrical, one-component hysteresis loop.

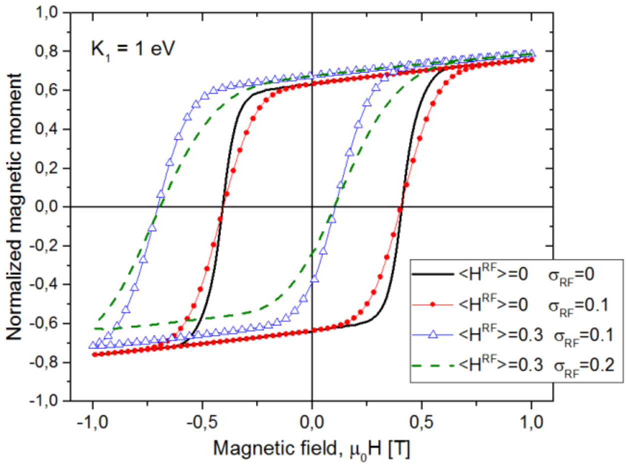

Fig. 5. The simulated hysteresis loops obtained for one component system with different parameter of additional random field H^{RF}

On the other hand, positive average vale of random filed $<\mu_0 H^{RF}>$ = 0.3 T shifts hysteresis loops to the left direction as well as provides asymmetries in X axis. Such behaviour can be easy explain taking into account the total magnetic field $H_i = H^{EX} + H_i^{RF}$ "feeling" by i-th object. Additional value of H_i^{RF} increasing the total magnetic field in I and IV quadrant of coordinate system and consequently, accelerates the ordering of magnetic object according to field direction. In the second half of coordinate system the situation is opposite. It is important to note, that the biggest absolute value of $\mu_0 H_i$ in the III quarter is equal to 0.7 T, therefore some of the magnetic object are not fully saturated. This situation is manifested by asymmetrical values of positive and negative magnetic saturation. Moreover, increasing of standard deviation for random field distribution σ_{RF} leads to larger dispersion of total magnetic field "feeling" by each object and consequently, increases the smoothness of obtained hysteresis loops.

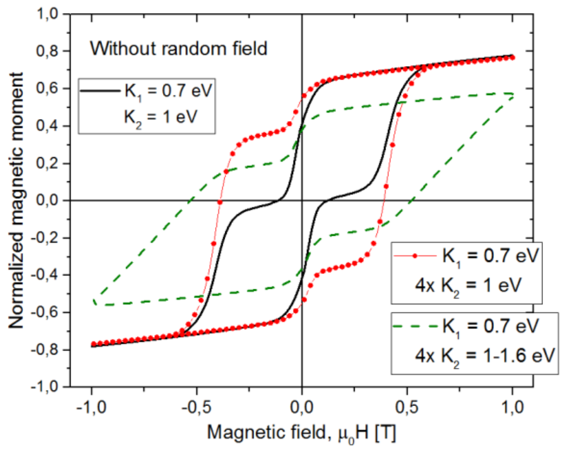

Fig. 6. The simulated hysteresis loops for the system including two type of magnetic component with different proportion of anisotropy coefficient and amount of objects

In order to perform the quantities analysis of experimental measurements (see Fig. 1) the more complicated, multi-component systems were also studded. Figure 6 presents the example of hysteresis loops obtained for sets of two kind of magnetic object corresponds to soft and hard magnetic phases. For all disused cases, the anisotropy coefficient of soft component K_1 = 0.7 eV were chosen, while the hard one is represented by K_2 = 1 eV or the linear distribution from 1 to 1.6 eV for last case. In order to improve the clarity of multi-component impact, the additional random field was turn off. The simulated hysteresis loops present classical shape characteristic for double-phases materials with non-interacting magnetic objects.

The proportion between amount of soft and hard component (1:1 and 1:4 for presented examples) corresponds to height of the slope in II and IV quarter of coordinate system. The most interesting example includes the wide distribution of anisotropy for hard component. In this case, the magnetic objects changing their magnetization direction "one by one" according to magnetic field increase in contrast to rapid slopes for well-defined, narrow distribution of magnetic object anisotropy. It is important to note, that applied external magnetic field (+/- 1 T) is not sufficient to fully saturate all magnetic object with anisotropy coefficient up to1.6 eV. Such situation was chosen for further analysis due to the well correlation to the experimental data.

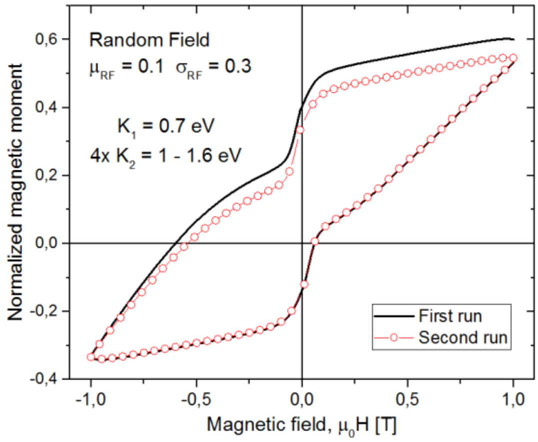

Fig. 7. The first and the second run of simulated hysteresis loop obtained for the system consists two type of magnetic component and additional random field

Figure 7 shows the magnetic hysteresis loop simulated based on the system consists two kind of magnetic component - as previous, soft phase (K_1 = 0.7 eV) and hard phase (K_2 = 1 - 1.6 eV) with proportion of amount 1:4, respectively. The average value of additional random field $<\mu_0 H^{RF}>$ = 0.1 T as well as standard deviation of Gaussian distribution σ_{RF} = 0.3 were chosen experimentally. The obtained hysteresis corresponds to all characteristic features of measured $M(H)$ curves presented in Fig. 1. The shift provided by additional random field combined with double-component system characterised by wide distribution of anisotropy coefficient simulated in relative narrow range of magnetic field leads to asymmetric, not fully saturated and opened, smooth double-phases hysteresis loop typical for Fe-Nb-B-Tb nanocrystalline bulk alloys obtained by vacuum suction casting technique. Especially important is a comparison of the first and the second run of simulation for the same system similar to the measurement procedure performed for real material. In both cases the second run is not identical with the previous one for upper part of $M(H)$ curve, which indicates that some of the magnetic object are “frozen” after first magnetization and do not take direct part in the further magnetisation procedure.

4 CONCLUSIONS

In this work the extension of the Stoner-Wohlfarth and Two-level models were proposed and tested. It is especially important to note, that the introduction of an additional, random field supplements the standard S-W model for interactions between ultra-hard and others, more soft, magnetic objects. In relation to performed simulations, the main conclusions can be summarized as follows:

(i) The applied model including additional random field increasing during first magnetization as well as two type of magnetic object corresponds to soft and hard magnetic phases leads to the results of performed simulations which are in good agreement with the experimental data for the Fe-Nb-B-Tb type of balk nanocrystalline alloys.

(ii) The obtained results confirm correctness of the applied model in relation to magnetic systems with ultra-hard interacting magnetic object and allow for quantities analysis of asymmetric, not fully saturated and opened, multi-phases hysteresis loops.

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Received 30 November 2015