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Modeling of hysteresis characteristics of a dilute magnetic with dipole-dipole interaction of particles

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ABSTRACT

A modification of the method for calculating the distribution function of random fields of dipole-dipole interaction in dilute magnetics using the expansion in the Gram-Charlier series with the help of Bell polynomials has been carried out. On the basis of this method, a model has been developed that allows us to estimate the ensemble magnetisation, the volume fractions of particles in different magnetic states, the volume concentration of the ferrimagnetic and its effective spontaneous magnetisations on the basis of experimental data on hysteresis characteristics. The proposed approach allows us to take into account the particle size distribution and magnetic states. The model has several advantages, such as the possibility of taking into account the cluster distribution of particles and applicability to the limiting cases of thin layer and thin filament. Examples of partial verification of this model on objects of artificial and natural origin are given.

KEYWORDS

dilute magnetic • random fields of dipole-dipole interaction • method of moments • magnetization coercivity • lognormal distribution • magnetic states • effective spontaneous magnetization

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Introduction

A considerable number of works [1–7] are devoted to the theoretical description of the processes of remanent magnetization formation in various types of magnetic materials of both natural and artificial origin. Micromagnetic modeling, of this computer simulations [8–12], widely used for the theoretical study of magnetic structures, including those taking into consideration the dipole-dipole interaction between particles [13–17], firstly, often does not take into consideration the possible chemical heterogeneity of individual particles, secondly, their distribution by size, and, accordingly, by magnetic states [18,19], and thirdly, in the case of using software, there are often computational problems associated with the huge number of particles in real objects. Joint application of the micromagnetic approach and statistical methods enables partially solving these problems. The works of the authors [7,20] show the validity of such an approach for ensembles of magnetic particles randomly scattered in a "non-magnetic" matrix (dilute magnetic).

The purpose of this work is to modify the method developed earlier [21,22] for calculating the distribution function of random fields of dipole-dipole interaction in a dilute magnetic, and to apply this method to calculate the magnetization and to estimate the effective spontaneous magnetizations and ferrimagnetic concentration in the sample.

Method of moments for a cylindrically shaped sample

To find the distribution density of the dipole-dipole interaction fields of randomly scattered magnetic dipoles, we will use the approximation that *N* single-domain uniaxial spherical particles of diameter d_0 with magnetic moment m are randomly located in the volume *V*, which create a field on a fixed particle located at the origin of coordinates [21].

The field **h** generated by the test particle,

$$\mathbf{h}(\mathbf{m},\mathbf{r}) = \frac{3(\mathbf{mr})\mathbf{r}}{r^5} - \frac{\mathbf{m}}{r^3}, \quad (1)$$

where **r** is the radius vector of the test particle. Here and further in the paper all expressions are written in the CGS system, unless otherwise specified.

By averaging the fields over all values of the magnetic moments and radius vectors, it is possible to calculate the moments of the density distribution of the random fields of the dipole-dipole interaction *w*(**h**):

$$< h_k^n > = \frac{1}{V}\int \omega(\Omega)d\Omega \int_V h_k^n(\mathbf{m},\mathbf{r})dV, \quad (2)$$

$$\mu_n = N < h^n >, \quad (3)$$

where *k* is the index responsible for the projection (*k* = *x, y, z*), ω is the distribution density of magnetic moment orientations, and *V* is the volume of the sample excluding the central region with a diameter of $2d_0$.

For further analysis, the shape of a non-magnetic matrix in the form of a cylinder having height *d*, base radius *R*, and volume *V* was chosen (Fig. 1). This geometry allows further analyzing bulk samples as well as thin layers ($d \ll R$) and thin threads ($d \gg R$).

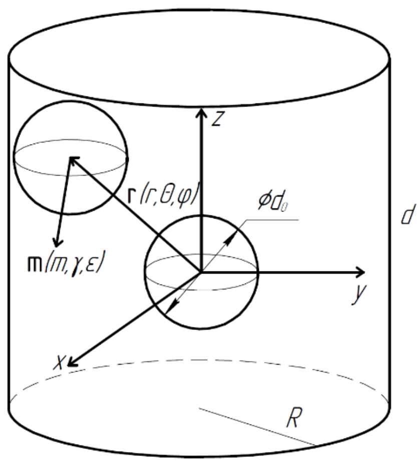

Fig. 1. Non-magnetic matrix of cylindrical shape with chaotic volume distribution of magnetic particles

The notations adopted in this model are as follows: the coordinates of the sample particle are spherical coordinates (*r*, θ, *φ*), the coordinates of the magnetic moment of the sample particle are (*m*, γ, ε), $\omega(\Omega) = \omega(\gamma, \varepsilon)\sin\gamma\, d\gamma\, d\varepsilon$. The two cases when the easy magnetization axes and the magnetic moments of the particles are parallel to the coordinate axes *Oz* and *Ox* (Eqs. (4) and (5), respectively) are of most interest:

$$\omega(\gamma,\varepsilon) = \frac{1}{\sin\gamma}[\alpha\delta(\gamma) + \beta\delta(\gamma-\pi)]\delta(\varepsilon), \quad (4)$$

$$\omega(\gamma,\varepsilon) = \frac{1}{\sin\gamma}[\alpha\delta(\varepsilon) + \beta\delta(\varepsilon-\pi)]\delta(\gamma-\pi/2), \quad (5)$$

where $\alpha + \beta = 1$ are the relative fractions of particles oriented along and against the direction of the coordinate axis, respectively. By substituting Eqs. (4) and (5) into Eq. (2), it is possible to analytically calculate the moments of the interaction field distribution [21]. The obtained expressions are cumbersome, but their use saves computational resources and provides an alternative to rather labor-intensive methods such as the Monte Carlo method.

There are at least two series expansions by moments of the probability density of a random normalized variable - the Gram-Charlier series expansion by orthogonal polynomials and the Edgeworth asymptotic series expansion [23]. In this paper, the Gram-Charlier series expansion is used, although the choice of the series expansion is not fundamental, since both series converge to the same values.

The Gram-Charlier series expansion of the probability density function is as follows:

$$f(x) = C_0\phi(x) + \frac{C_1}{1!}\phi'(x) + \cdots + \frac{C_n}{n!}\phi^{(n)}(x), \quad (6)$$

where $\varphi(x) = \exp(-x^2/2)/(2\pi)^{1/2}$ is the density of the standard normal distribution, $\varphi^{(n)} = (-1)^n He_n(x)\, \varphi(x)$, $He_n(x)$ is the Hermite polynomial of the n^{th} degree, C_n are constant coefficients defined as:

$$C_n = (-1)^n \int_{-\infty}^{+\infty} He_n(x) f(x) dx. \quad (7)$$

The Hermite polynomials (in the probabilistic definition) can be found through the recurrence relation:

$$\begin{aligned} &He_0(x) = 1,\\ &He_1(x) = x,\\ &He_n(x) = x \cdot He_{n-1}(x) - (n-1) \cdot He_{n-2}(x),\ n \geq 2. \end{aligned} \quad (8)$$

Thus, it is possible to analytically find any coefficient and any term of the series expansion of $\varphi(x)$ through reduced moments:

$$\nu_k = \frac{\mu_k}{\sigma^k} = \int_{-\infty}^{+\infty} x^k f(x) dx. \quad (9)$$

This method is very convenient for analytical calculation of the coefficients and further expansion of the density into a series with a predetermined number of expansion terms. In case it is necessary to dynamically determine the number of expansion terms in the program, the numerical calculation of Eq. (9), although possible, is difficult. Therefore, in our case another approach using the Bell polynomials is chosen.

The probability density function can be defined as follows [24]:

$$f(x) = \exp\left[\sum_{r=3}^{\infty} k_r \frac{\left(-\frac{d^r}{dx^r}\right)}{r!}\right]\phi(x). \quad (10)$$

The sum under the exponent can be rewritten through the complete Bell polynomials [25]:

$$\exp\left[\sum_{r=3}^{\infty} k_r \frac{\left(-\frac{d^r}{dx^r}\right)}{r!}\right] = \sum_{n=0}^{\infty} B_n(0,0,k_3,k_4,..,k_n)\frac{\left(-\frac{d^n}{dx^n}\right)}{n!}, \quad (11)$$

where k_n are the cumulants expressed through reduced moments by means of the incomplete Bell polynomials:

$$k_n = \sum_{i=1}^{n} (-1)^{i-1}(i-1)!\, B_{n,i}(\nu_1, \nu_2, \dots, \nu_{n-i+1}). \quad (12)$$

The Bell polynomials can be found through the recurrence relation [26]:

$$\begin{aligned} &B_{n+1,k+1}(x_1, x_2, \dots, x_{n-k+1}) = \sum_{i=0}^{n-k} C_n^i x_{i+1} B_{n-i,k}(x_1, x_2, \dots, x_{n-k-i+1}),\\ &B_{0,0} = 1,\\ &B_{n,0} = 0 \text{ for } n \geq 1,\\ &B_{0,k} = 0 \text{ for } k \geq 1, \end{aligned} \quad (13)$$

where C_n^i = $n!/[i!(n-i)!]$. The complete Bell polynomials are defined through the incomplete ones as a sum:

$$B_n(x_1, x_2, \ldots, x_n) = \sum_{k=1}^{n} B_{n,k}(x_1, x_2, \ldots, x_{n-k+1}). \quad (14)$$

Then the density of probability distribution is expressed through cumulants as follows:

$$f(x) = \left[\sum_{n=0}^{\infty} B_n(0, 0, k_3, k_4, \ldots, k_n) \frac{(-D)^n}{n!}\right] \phi(x). \quad (15)$$

Analytical expressions of moments (3) were previously obtained in [21]. Then the distribution density is found by Eqs. (6) or (10) with any necessary accuracy. For most problems, several first terms of the series are sufficient. Let the first unaccounted term of series (6) have the number n. Then the condition of its smallness can be written as follows:

$$\frac{1}{n!}\mu_n \ll 1 \text{ or } \frac{\mu_n}{n!\mu_2^{n/2}} \ll 1. \quad (16)$$

To estimate the accuracy, we discard the terms of order d_0/d and d_0/R in the expressions for the moments due to their smallness, then for a magnetic with volume concentration $c = Nv_0/V$ (v_0 is the particle volume) and spontaneous magnetization I_s the moments are:

$$\mu_{n,z}, \mu_{n,x} \approx 8\left(\frac{\pi}{6} I_s\right)^n c \frac{1}{n-1} \sum_{k=0}^{n} C_n^k \frac{(-3)^k}{2k+1}. \quad (17)$$

If we take μ_5 as the first discarded term, then, taking into consideration $\mu_{2,z}$, $\mu_{2,x} \approx (32/5)\cdot c\cdot(\pi I_s/6)^2$, the condition of a satisfactory approximation of the density distribution of the interaction fields will have the following form:

$$\mu_5 \ll 12400 \left(\frac{\pi}{6} I_s\right)^5 c^{5/2}. \quad (18)$$

From Eq. (18) we obtain a lower limit on the range of possible volume concentrations of the magnetic, at which the remaining quantity of terms of series (6) well approximates the distribution density of the interaction fields, namely $c \gg 0.003$. Thus, for larger concentrations, the first four moments of the distribution function are sufficient. Approximate values of the moments up to the fourth order inclusive, obtained taking into account Eq. (18) from the analytical expressions given in [21], are given in Table 1.

Table 1. Analytical expressions of the first four moments of the distribution function in approximation $2R > d$ ($\mathrm{tg}\,\theta_{max} = 2R/d$ determines the maximum value of the angle θ in Fig. 1): $\langle H_i \rangle$ - mathematical expectation, σ^2 - dispersion, μ_3 and μ_4 - central moments of the corresponding orders

Moment	Orientation of the external field and easy axes	
	Parallel to the base of the cylinder (coordinate axes Ox or Oy)	Perpendicular to the base of the cylinder (coordinate axis Oz)
$\langle H_i \rangle$	$\frac{4\pi}{3} c I_s \left(1 - \frac{3}{2}\cdot\cos\theta_{max}\right)\zeta(x_0)$	$-\frac{8\pi}{3} c I_s \left(1 - \frac{3}{2}\cdot\cos\theta_{max}\right)\zeta(x_0)$
σ^2	$\left(\frac{4\pi}{3}\right)^2 c I_s^2 \frac{1}{10}\left[1 - \frac{45}{32}\cdot\left(\frac{d_0}{d}\right)^3\right]$	$\left(\frac{4\pi}{3}\right)^2 c I_s^2 \frac{1}{10}\left[1 - \frac{15}{4}\cdot\left(\frac{d_0}{d}\right)^3\right]$
μ_3	$\left(\frac{4\pi}{3}\right)^3 c I_s^3 \frac{1}{280}\left(1 + 10\cdot\left(\frac{d_0}{d}\right)^6\right)\zeta(x_0)$	$\left(\frac{4\pi}{3}\right)^3 c I_s^3 \frac{1}{280}\left(1 - 67\cdot\left(\frac{d_0}{d}\right)^6\right)\zeta(x_0)$
μ_4	$\left(\frac{4\pi}{3}\right)^4 c I_s^4 \frac{1}{1120}\left[1 - 20\cdot\left(\frac{d_0}{d}\right)^9\right]$	$\left(\frac{4\pi}{3}\right)^4 c I_s^4 \frac{1}{1120}\left[1 - 264\cdot\left(\frac{d_0}{d}\right)^9\right]$

In Table 1, dimensionless magnetization ζ of an ensemble of particles is equal to the difference between the relative number of magnetic moments oriented along and against external field *H* directed along the chosen axis:

$$\zeta = \alpha - \beta = \int_{-H_0}^{\infty} W(H_i - H)dH_i - \int_{-\infty}^{-H_0} W(H_i - H)dH_i, \tag{19}$$

where H_0 is the magnetization reversal field of the particle, $W(H_i - H)$ is the distribution density of the magnetostatic interaction field projections on the chosen coordinate axis, which can be expressed through $f(x)$:

$$W(H_i - H) = f(x)/\sigma, \text{ where } x = \frac{H_i - H - \langle H_i \rangle}{\sigma}. \tag{20}$$

Equation (19) can be reduced to the following form:

$$\zeta(x_0) = 1 - 2\int_{-\infty}^{-x_0} f(x)dx, \text{ where } x_0 = \frac{H_0 + H + \langle H_i \rangle}{\sigma}. \tag{21}$$

Expression (21) can be simplified by substituting Eqs. (6) and (15):

$$\zeta(x_0) = 1 - 2\int_{-\infty}^{-x_0} \phi(x) \sum_{n=0}^{\infty} A_n He_n(x)\, dx, \tag{22}$$

where $A_n = (-1)^n C_n/n! = B_n(0, 0, k_3, k_4, \dots, k_n)/n!$ is a field-independent coefficient that depends only on the distribution moments. In a similar way, the functions $Z(x_0)$, independent of the distribution moments, can be introduced:

$$Z_n(x_0) = \int_{-\infty}^{-x_0} \phi(x) He_n(x) dx. \tag{23}$$

This function can be simplified to the following system:

$$Z_n(x_0) = \begin{cases} \frac{1}{2} - \frac{1}{2}\operatorname{erf}\left(\frac{x_0}{\sqrt{2}}\right), n = 0 \\ (-1)^n \phi(x_0) \cdot He_{n-1}(x_0), n \geq 1 \end{cases}, \tag{24}$$

and the magnetization is simplified to the following expression:

$$\zeta(x_0) = 1 - 2\sum_{n=0}^{\infty} A_n \cdot Z_n(x_0). \tag{25}$$

A model of interacting particles with effective spontaneous magnetization

In a real dilute magnetic, the particles are distributed by sizes and magnetic states, may have different crystallography and directions of easy axes, be chemically inhomogeneous, etc. In the case of an ensemble of stable single-domain particles, we can take a lognormal size distribution and, having estimated the average particle size, calculate the ensemble magnetization in an external field. If the dispersion of the distribution is large and the ensemble includes particles in different magnetic states, the concept of effective spontaneous magnetization, which takes into consideration possible magnetic and/or chemical inhomogeneity of the particles, can be introduced to estimate the hysteresis characteristics of the ensemble.

By using the approximation of lognormal particle distribution by volume [27,28], the fractions of particles in different magnetic states can be calculated (Fig. 2). In modeling, the range of particles is divided into 5 intervals: superparamagnetic (SP), single-domain (SD), pseudo-single-domain (PSD), and multi-domain (MD), with the range of superparamagnetic particles containing unblocked superparamagnetic ones that do not contribute to the remanent magnetization, as well as superparamagnetic particles blocked by magnetostatic interaction that contribute not only to the saturation magnetization but also to the remanent magnetization [6,29]. The probability density of the lognormal distribution is written as:

$$\varphi(x) = \frac{1}{x\sigma\sqrt{2\pi}} \exp\left(-\frac{(\ln(x-\mu))^2}{2\sigma^2}\right), \tag{26}$$

where $x = v/v_p$ is the particle volume reduced to the characteristic volume, σ is the standard deviation, and μ is the mathematical expectation of the corresponding Gaussian distribution. The fraction of particles in the volume range from x_1 to x_2 is equal to:

$$n_i = \int_{x_1}^{x_2} \varphi(x)dx \,/\, \int_{x_{\min}}^{x_{\max}} \varphi(x)dx, \quad (27)$$

where x_1 and x_2 are the lower and upper limits of the range of volumes of a group of particles in a certain magnetic state, x_{min} (d = 0) and x_{max} ($d = d_{max}$) are the minimum and maximum relative volumes of particles, respectively, and $x_2 \leqslant x_{max}$.

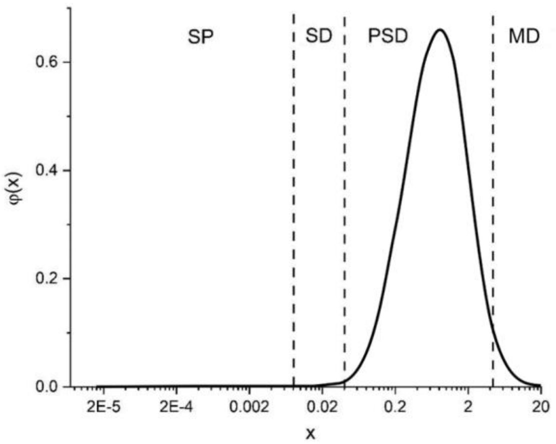

Fig. 2. Lognormal distribution of relative volumes of particles [29]

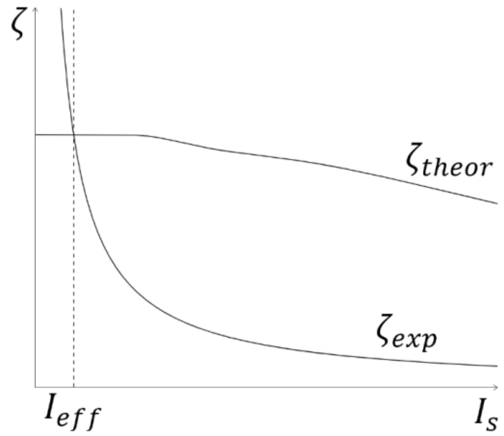

Fig. 3. Graphical determination of the value of effective spontaneous magnetization ($I_{s\,eff}$ or $I_{rs\,eff}$)

The average relative volume of each particle group is found as:

$$x_i = \int_{x_1}^{x_2} x \cdot \varphi(x)dx, \quad (28)$$

and the average absolute volume as:

$$v_i = v_p \frac{x_i}{n_i}, \quad (29)$$

where $v_p = \pi d_p^3/6$, d_p is the characteristic size of the particle.

The volume concentration of particles in each of the states is equal to:

$$c = N\frac{v_{average}}{V} = \frac{N}{V}(n_{sp}v_{sp} + n_{bsp}v_{bsp} + n_{sd}v_{sd} + n_{psd}v_{psd} + n_{md}v_{md}), \quad (30)$$

where N is the number of ferrimagnetic particles in a sample of volume V, $v_{average} = n_{sp}v_{sp} + n_{bsp}v_{bsp} + n_{sd}v_{sd} + n_{psd}v_{psd} + n_{md}v_{md}$ is the average volume of ferrimagnetic particles in various magnetic states with corresponding average volumes [6]. If the particles are grouped into clusters in the sample volume, which is essential to account for the dipole-dipole interaction, we can introduce the volume concentration of ferrimagnetic η in a cluster of volume V_{cl} and concentration of clusters c_{cl} in the sample. Then $c = \eta c_{cl}$ if the interaction between clusters can be neglected. If the interaction cannot be neglected, then, in the first approximation, we can discard the cluster distribution and consider the particle distribution to be homogeneous over the sample volume (see Eq. (30)).

To find the value of the effective spontaneous magnetization, it is necessary to bring the theoretical dimensionless magnetization determined by Eq. (25) into agreement with the experimental value:

$$\zeta_{exp} = \frac{M}{cI_s}, \quad (31)$$

where M is the saturation magnetization M_s or the saturation remanence M_{rs}, and c_s and c_{rs} are the corresponding volume concentrations of particles contributing to M_s or M_{rs}. In our model, two values correspond to the spontaneous saturation magnetization of ferroparticles: $I_{s\ eff}$ and $I_{rs\ eff}$. For chemically homogeneous particles, $I_{s\ eff}$ coincides with the spontaneous magnetization of the material, whereas for chemically inhomogeneous particles it is some averaged value. The value of $I_{rs\ eff}$ takes into consideration the magnetic and chemical heterogeneity, as well as the peculiarities of the crystallography of both individual particles and the ensemble as a whole. Besides, in our model, volume concentration c_{rs} does not include fractions of truly superparamagnetic (unblocked) and multidomain particles.

Since, in general, finding the effective spontaneous magnetizations is reduced to solving the integral equation (see Eqs. (23-25) and expressions for odd moments in Table 1), it is easier to find the solution graphically by specifying the range of possible values of I_s. The point of intersection of the theoretical ζ_{theor} (25) and experimental ζ_{exp} (31) curves provides the value of the effective spontaneous magnetization I_{eff}, corresponding to $I_{s\ eff}$ or $I_{rs\ eff}$ (Fig. 3).

The effective spontaneous magnetization by the remanence $I_{rs\ eff}$ takes into consideration the changes that occur in the magnetic state of the ensemble and its constituent particles when the external magnetic field decreases from saturation to zero. The value of $I_{rs\ eff}$ is influenced by a number of factors, namely, the distribution of particles by size and, consequently, by magnetic states, the scatter of the directions of the crystallographic axes of particles relative to the external field, and the chemical heterogeneity of the whole ensemble and individual particles. In addition, in our approximation of magnetostatic interaction due to a large number of particles, the distribution of random fields of the dipole-dipole interaction is considered unchanged, and unblocking of a part of magnetic moments of superparamagnetic particles is related only to the reduction of the external field.

To check the consistency of the values of the effective spontaneous magnetizations with the experimental data, we can use the following evaluation formula:

$$\frac{M_{rs}}{M_s} = \frac{c_{rs} I_{rs}}{c_s I_s}. \quad (32)$$

Here c_{rs} is the volume concentration of particles participating in the creation of the remanent magnetization (for simplicity we can neglect the concentrations of truly superparamagnetic and multidomain particles, see Eq. (30)); the concentration of particles contributing to the saturation magnetization, $c_s = c$.

Conclusion

The modified method for calculating the distribution function of dipole-dipole interaction fields in a dilute magnetic of cylindrical shape taking into consideration the lognormal distribution of particles by sizes (and, consequently, magnetic states) allows, on the basis of experimental values of hysteresis parameters and structural characteristics of individual particles, their clusters, and the ensemble as a whole, calculating the volume fractions of particle concentrations in different magnetic states, the volume concentration of ferrimagnetic in the sample, and its effective spontaneous magnetizations.

The approach presented in this paper has a number of advantages. In the case of chaotic distribution of magnetic particles in a “non-magnetic” matrix, the model allows moving from micromagnetic calculations of the interaction of each of the particles with one another to the distributions of random interaction fields. In addition, due to the decomposition of the distribution function into an infinite series, any necessary accuracy can be achieved, and the moments of this function are obtained in analytical form. The model allows taking into consideration cluster distribution of particles. If the concentration of clusters is small, the dipole-dipole interaction between them can be neglected, otherwise the distribution of particles in the sample can be considered homogeneous (the entire sample is one cluster). Moreover, the choice of the cylindrical volume allows the method to be used in the limiting cases of thin threads and thin layers. The drawback of the model is the simplifying assumptions about the spherical shape of the particles and their crystallographic uniaxiality. However, this disadvantage is partially compensated by the fact that the model assumes the field of magnetization reversal of a single particle $H_0 = H_{cr}$, where H_{cr} is the experimental value of the coercivity of remanence.

Partial verification of the model described in this paper was carried out when calculating the hysteresis characteristics of objects of both artificial and natural origin [6,7,20,29,30]. To estimate the hysteresis characteristics of two-phase chemically inhomogeneous particles and their ensembles, one can use the approach developed by the authors in [6,7] and the program for micromagnetic modeling [31,32].

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