Research Article

Effect of Mechanical Stress on Magnetic States and Hysteresis Characteristics of a Two-Phase Nanoparticles System

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In terms of the two-phase nanoparticles model, the effect of mechanical stress on the magnetic state of both uniaxial and multiaxial heterophase magnetic is investigated. The spectrum of critical fields of reversal of phases' magnetic moments was calculated and phase diagrams were drawn to assess the effect of mechanical stress on the degree of metastability of two-phase nanoparticles' magnetic states. By the example of epitaxial cobalt-coated γ -Fe₂O₃ particles, a theoretical analysis of the effect of uniaxial mechanical stress on the magnetization of a system of noninteracting heterophase nanoparticles is investigated. It was shown that tension reduced and compression increased coercive force H_c , while the residual saturation magnetization I_{rs} was not changed under the influence of mechanical stress.

1. Introduction

One of the factors that influence the magnetization is the heterophase of nanoparticles determining magnetic properties of nanodispersed materials. This heterophase may be specified by various factors. Thus, according to some authors (see, e.g., [1–5]), the formation of adjacent magnetic phases in natural magnetic materials is associated with oxidation or decomposition processes of a solid solution occurring in magnetic-ordered grains. For example, titanomagnetite being the main carriers of magnetic properties of natural materials can be exposed to decay [3] or oxidation [4, 5] processes. As a result of titanomagnetite collapse in nanoparticles, the extraction of both titanium depleted and enriched phases ultimately leads to a change in magnetic properties of a natural material. As an example of multiphase artificial magnetic materials can be nanoparticles with core/shell structure. Currently there are many different types of core/shell nanoparticles, such as metal/metal [6–11], nonmetal/metal [12–14], metal/polymer [15, 16], nonmetal/nonmetal, polymer/nonmetal, and polymer/polymer [17]. Development of technology of core/shell nanoparticles allowed for obtaining stable particles of nanometer scale [18–20], the particle size of 1–100 nm in high demand in many areas, particularly important to its

application in medicine [21] and the production of memory storage [22]. Core/shell nanoparticles containing metals (Fe, Co, gold, platinum, palladium, Mn, Zn, etc.) are used in the manufacture of materials for storage (magnetic memory) and creating the design of microwave and electromagnetic devices [23–25] as well as creating medicines [26], drug delivery [27–29], oncology [30], and biomedicine [31, 32]. Polymer and nonmetallic core/shell nanoparticles are a new class of materials for electronics, for example, the organic light-emitting diodes (OLEDs), organic photovoltaics (OPVs), sensors, and organic field effect transistors (OFETs) [33–35]. Carriers of magnetic memory elements' magnetic properties may be represented by iron particles [36–38] exposed to surface oxidation or magnetic nanoparticles coated with other material, such as cobalt-coated γ -Fe₂O₃ particles [39–41]. According to Mössbauer spectroscopy [36] oxidized coating of iron particles is a mixture of γ -Fe₂O₃ and Fe₃O₄, while the central part is composed of α -Fe. The thickness of the coating may be around 10% of the radius of the particles, and the amount of phase composed of iron oxides is more than 50%, according to other reports [37]. These particles can be either cubic, spherical [42, 43], or very extended [37]. Note that magnetic properties of these nanoparticles can change significantly once they transit to heterophase

state. Thus, an epitaxial cobalt coating of $\gamma\text{-Fe}_2\text{O}_3$ particles increases coercive force [41] that is associated by the authors [44, 45] with the interfacial exchange interaction. Interfacial exchange interaction can change not only the coercivity but also other magnetic characteristics [9, 46]. Amorphous magnetic materials obtained in a special way may also refer to multiphase ones. The process of phase formation in such magnets associates with amorphous material transition from metastable to equilibrium state (see, e.g., [47–49]). Nanocrystalline alloys' magnetic matrix that contains grains of other ferromagnets is of special interest here. Exchange interaction between the matrix and grains leads to collective phenomena, which causes a high performance of both magnetically soft and hard materials (e.g., Fe, Cu, Nb, Si, and B and Nd, Fe, and B, resp.). Along with the exchange interaction, there is mechanical stress at the interphase interface that is able to alter magnetic states of both phases due to size differences of neighboring phases' lattices. For example, the transformation in iron and its alloys $\gamma \rightarrow \alpha$ is accompanied by a grain extension (1.5%) and, as a consequence, by significant deformations. However, the heterogeneity of the interphase interface can lead to a significant reduction of internal stress associated with the $\gamma \rightarrow \alpha$ transformation [50]. It should be noted that the mechanical stresses can have a significant effect on the magnetic properties of nanoparticles [51–55].

Rather detailed theoretical study of magnetic states and magnetization processes of the system of uniaxial two-phase particles is presented in the works [39, 46, 51, 54, 56–59]. In this paper, we attempt to expand the model [58, 59] to multiaxis dual-phase particles and studies of the effect of mechanical stress on the magnetic states and magnetic properties of systems of such particles.

2. The Model of a Uniaxial Two-Phase Nanoparticle

In contrast to the model of two neighboring plane-parallel phases used in the works [56–59], we use a more plausible model of “inclusion.”

- (1) Homogeneously magnetized nanoparticle (phase (1)) with the volume $= V$ and the shape of an ellipsoid with the elongation $= q_1$ contains a uniformly magnetized ellipsoidal inclusion (phase (2)) with the volume $v = \varepsilon V$, whose long axis coincides with the long axis of the particle oriented along the axis OZ (see Figure 1).
- (2) The axes of crystallographic anisotropy $\langle k_A \rangle^{(1)}$ and $\langle k_A \rangle^{(2)}$ of both uniaxial ferromagnets are considered to be parallel to the axis OZ, and the vectors of phases' spontaneous magnetization $I_s^{(1)}$ and $I_s^{(2)}$ are located in the plane YOZ containing long axes of magnetic phases and make angles $\vartheta^{(1)}$ and $\vartheta^{(2)}$ with and the axis OZ, respectively.
- (3) The external magnetic field H is placed along the axis OZ as uniaxial mechanical stress σ .
- (4) Thermal fluctuations of phases' magnetic moments may be neglected.

FIGURE 1: An illustration of the model of a two-phase particle.

We exclude from consideration magnetoelastic interfacial interaction, which may be more or less justified only in case of highly disordered distribution of magnetic atoms in the boundary layer (see, e.g., [50]).

In the accepted approximation, free energy of nanoparticle F located in an external field H can be written as the sum of

- (i) crystallographic anisotropy energy:

$$E_A = -\frac{1}{4} \left\{ \left(I_s^{(1)} \right)^2 k_A^{(1)} (1 - \varepsilon) \cos 2\vartheta^{(1)} + \left(I_s^{(2)} \right)^2 k_A^{(2)} \varepsilon \cos 2\vartheta^{(2)} \right\} V, \quad (1)$$

- (ii) interaction energy of magnetic moment with its own magnetic field which in accordance with [66] can be represented as

$$E_m = \left\{ -\frac{\left(I_s^{(1)} \right)^2}{4} (1 - 2\varepsilon) k_N^{(1)} \cos 2\vartheta^{(1)} - \frac{\left(I_s^{(2)} \right)^2}{4} \varepsilon k_N^{(2)} \cos 2\vartheta^{(2)} + \frac{\varepsilon I_s^{(1)} I_s^{(2)}}{3} \left(k_N^{(2)} - k_N^{(1)} \right) \times \left(\sin \vartheta^{(1)} \sin \vartheta^{(2)} - 2 \cos \vartheta^{(1)} \cos \vartheta^{(2)} \right) \right\} V, \quad (2)$$

- (iii) energy of magnetoelastic interaction with uniaxial mechanical stress σ :

$$E_\sigma = -\frac{1}{4} \left\{ \left(I_s^{(1)} \right)^2 k_\sigma^{(1)} (1 - \varepsilon) \cos 2\vartheta^{(1)} + \left(I_s^{(2)} \right)^2 k_\sigma^{(2)} \varepsilon \cos 2\vartheta^{(2)} \right\} V, \quad (3)$$

- (iv) exchange interaction energy across the border, which according to [56] can be defined as

$$E_{\text{ex}} = -\frac{2A_{\text{in}}}{\delta} \cos(\vartheta^{(1)} - \vartheta^{(2)})s, \quad (4)$$

- (v) interaction energy of magnetic moment with an external magnetic field

$$E_H = -H \left[I_s^{(1)} (1 - \varepsilon) \cos \vartheta^{(1)} + I_s^{(2)} \varepsilon \cos \vartheta^{(2)} \right] V. \quad (5)$$

In (1)–(5) $k_A^{(1,2)} = K_1/(I_s^{(1,2)})^2$, $k_N^{(1,2)}$, dimensionless constants of anisotropy and phases' shape anisotropy are K_1 —the first anisotropy constant, and $k_\sigma^{(1,2)} = 2(\lambda_2^{(1,2)} + \lambda_3^{(1,2)})\sigma/(I_s^{(1,2)})^2$, $\lambda_2^{(1,2)}$, $\lambda_3^{(1,2)}$ —the second and third magnetostriction constants of a uniaxial crystal; V is the volume of a particle, s is the surface area that separates the phases, ε is the ratio of the inclusion volume to the volume of the whole particle, A_{in} is the constant of interfacial exchange interaction, and δ is the width of the transition region with the order of a constant lattice. Note that the shape anisotropy constant $k_N = 2\pi(1 - 3N_z)$ is expressed through the demagnetizing factor along the long axis N_z depending only on the elongation of the ellipsoid q :

$$N_z = \frac{\left[q \ln \left(q + \sqrt{q^2 - 1} \right) - \sqrt{q^2 - 1} \right]}{(q^2 - 1)^{3/2}}. \quad (6)$$

Analysis of two-phase particle's magnetic states is a procedure of minimization of the total free energy (F) of a nanoparticle:

$$F = \left\{ -\frac{(I_s^{(1)})^2}{4} K^{(1)} \cos 2\vartheta^{(1)} - \frac{(I_s^{(2)})^2}{4} K^{(2)} \cos 2\vartheta^{(2)} + I_s^{(1)} I_s^{(2)} (-\mathcal{U}_1 \sin \vartheta^{(1)} \sin \vartheta^{(2)} + \mathcal{U}_2 \cos \vartheta^{(1)} \cos \vartheta^{(2)}) - H \left[I_s^{(1)} (1 - \varepsilon) \cos \vartheta^{(1)} + I_s^{(2)} \varepsilon \cos \vartheta^{(2)} \right] \right\} V, \quad (7)$$

where the effective anisotropy constants $K^{(1,2)}$ and the position of are determined by the following formulas:

$$K^{(1)} = (1 - \varepsilon) (k_A^{(1)} + k_\sigma^{(1)}) + (1 - 2\varepsilon) k_N^{(1)} + \varepsilon k_N^{(2)}, \quad (8)$$

$$K^{(2)} = \varepsilon (k_A^{(2)} + k_\sigma^{(2)} + k_N^{(2)}).$$

The constants of an interfacial interaction $\mathcal{U}_1$ and $\mathcal{U}_2$ are expressed through the constants of magnetostatic and exchange interaction of the phases:

$$\mathcal{U}_1 = \varepsilon \left(\frac{(k_N^{(1)} - k_N^{(2)})}{3} + \frac{2sA_{\text{in}}}{\nu \delta I_s^{(1)} I_s^{(2)}} \right), \quad (9)$$

$$\mathcal{U}_2 = \varepsilon \left(\frac{2(k_N^{(1)} - k_N^{(2)})}{3} - \frac{2sA_{\text{in}}}{\nu \delta I_s^{(1)} I_s^{(2)}} \right).$$

Minimization of free energy (7) by $\vartheta^{(1)}$ and $\vartheta^{(2)}$ leads to a system of equations determining static states of phases' magnetic moments:

$$\begin{aligned} \frac{K^{(1)}}{2} \sin 2\vartheta^{(1)} + j \left[-\mathcal{U}_1 \cos \vartheta^{(1)} \sin \vartheta^{(2)} - \mathcal{U}_2 \sin \vartheta^{(1)} \cos \vartheta^{(2)} \right] \\ + \frac{H}{I_s^{(1)}} (1 - \varepsilon) \sin \vartheta^{(1)} = 0, \\ \frac{jK^{(2)}}{2} \sin 2\vartheta^{(2)} + \left[-\mathcal{U}_1 \sin \vartheta^{(1)} \cos \vartheta^{(2)} - \mathcal{U}_2 \cos \vartheta^{(1)} \sin \vartheta^{(2)} \right] \\ + \frac{H}{I_s^{(1)}} \varepsilon \sin \vartheta^{(2)} = 0, \end{aligned} \quad (10)$$

where $j = I_s^{(2)}/I_s^{(1)}$. The system of (10) allows to determine the main and metastable states of nanoparticles' magnetic moment.

3. Static States of the Magnetic Moment of Uniaxial Nanoparticle

The system of equations, within the accuracy of notations, transfers into the system of equations obtained earlier in [58, 59], so neglecting thermal fluctuations of the magnetic moment and in the absence of an external magnetic field when $K^{(1)} > 0$ and $K^{(2)} > 0$ (an "easy axis"), two-phase particles can be in one of the following states:

- (i) the first "(↑↑)-state": magnetic moments of both phases are parallel and directed along the axis OZ ;
- (ii) the second "(↑↓)-state": the phases are antiparallely magnetized, and the magnetic moment of the first phase $\mathbf{m}^{(1)}$ is directed along the axis OZ ;
- (iii) the third "(↓↓)-state": it differs from the first one in the antiparallel orientation of the phases magnetization relative to the axis OZ ;
- (iv) the fourth "(↓↑)-state": the magnetic moment of the second phase is directed along the axis OZ ; the magnetic moment of the first phase is directed against the axis OZ .

Besides, if the magnetostatic interaction between the phases dominates the exchange interaction $\mathcal{U}_2 > 0$, the second and fourth states are stable, while the first and third are metastable, since the free energy $F = \{-|K^{(1)}|(I_s^{(1)})^2 - |K^{(2)}|(I_s^{(2)})^2 - 4\mathcal{U}_2 I_s^{(1)} I_s^{(2)}\}V/4$ of a particle in these states is less than in the first and third, where $F = \{-|K^{(1)}|(I_s^{(1)})^2 - |K^{(2)}|(I_s^{(2)})^2 - 4\mathcal{U}_2 I_s^{(1)} I_s^{(2)}\}V/4$. Otherwise ($\mathcal{U}_2 < 0$), the first and third states are metastable.

The same states are realized in the case when the constants $K^{(1)}$ and $K^{(2)}$ have different signs. If $K^{(1)} < 0$ and $K^{(2)} < 0$

(an “easy plane”), a two-phase particle can be in one of the four states listed above with the phases’ magnetic moments perpendicular to the axis OZ .

Following (10), if at least one of the constants $K^{(1)}$ or $K^{(2)}$ is positive, then when the external magnetic field obtains a critical value H_c , the transition from one state to another will occur. For example, the critical field of the transition from the third state to the second or from the first state to the fourth is

$$H_c^{(3 \rightarrow 2)} = H_c^{(1 \rightarrow 4)} = \frac{|K^{(1)}| I_s^{(1)} - \mathcal{U}_2 I_s^{(2)}}{1 - \varepsilon}. \quad (11)$$

In much the same way we define the critical field of the transition from the third to the fourth (from the first to the second) state:

$$H_c^{(3 \rightarrow 4)} = H_c^{(1 \rightarrow 2)} = \frac{|K^{(2)}| I_s^{(2)} - \mathcal{U}_2 I_s^{(1)}}{\varepsilon}, \quad (12)$$

from the forth to the first (from the second to the third):

$$H_c^{(4 \rightarrow 1)} = H_c^{(2 \rightarrow 3)} = \frac{|K^{(1)}| I_s^{(1)} + \mathcal{U}_2 I_s^{(2)}}{1 - \varepsilon}, \quad (13)$$

from the second to the first (from the forth to the third):

$$H_c^{(2 \rightarrow 1)} = H_c^{(4 \rightarrow 3)} = \frac{|K^{(2)}| I_s^{(2)} + \mathcal{U}_2 I_s^{(1)}}{\varepsilon}, \quad (14)$$

from the third to the first and vice versa:

$$\begin{aligned} H_c^{(3 \rightarrow 1)} &= H_c^{(1 \rightarrow 3)} \\ &= \frac{|K^{(1)}| (I_s^{(1)})^2 + |K^{(2)}| (I_s^{(2)})^2 + 2(k_N^{(2)} - k_N^{(1)}) I_s^{(1)} I_s^{(2)}}{(1 - \varepsilon) I_s^{(1)} + \varepsilon I_s^{(2)}}, \end{aligned} \quad (15)$$

and from the second to the fourth and vice versa:

$$\begin{aligned} H_c^{(2 \rightarrow 4)} &= H_c^{(4 \rightarrow 2)} \\ &= \frac{|K^{(1)}| (I_s^{(1)})^2 + |K^{(2)}| (I_s^{(2)})^2 - 2(k_N^{(2)} - k_N^{(1)}) I_s^{(1)} I_s^{(2)}}{|(1 - \varepsilon) I_s^{(1)} - \varepsilon I_s^{(2)}|}. \end{aligned} \quad (16)$$

In the same way we can calculate the spectrum of critical fields when $K^{(1)} < 0$ and $K^{(2)} < 0$. In this case we can use the relations (11)–(16), having replaced $\mathcal{U}_2$ by $-\mathcal{U}_1$.

4. Magnetic States of Multiaxis Two-Phase Nanoparticle

Let nanoparticle’s magnetic phases be represented by the crystals of cubic symmetry with the constants of crystalline anisotropy of the first $K_1^{(1,2)}$ and the second $K_2^{(1,2)}$ order, respectively. Let us perform a solution of the problem of the

magnetic states of a multiphase particle in the framework of the following assumptions.

- (1) Crystallographic directions $[100]$, $[010]$, and $[001]$ of the phases coincide with the axes OX , OY , and OZ , respectively (see Figure 1), if the first constants $K_1^{(1,2)}$ of the phases’ crystalline anisotropy are positive. Otherwise ($K_1^{(1,2)} < 0$), we combine the “easy axis” (direction $[111]$) with the axis OZ .
- (2) We use the condition of magnetic uniaxiality of a multiaxis crystal’s grain [57, 67], the essence of which is that at a certain elongation its shape anisotropy prevails over the crystalline anisotropy. The process of magnetization of these particles is similar to magnetization of uniaxial particles with an effective constant determined by the total free energy. So for materials with $I_s > 350 \div 500$ (e.g., titanomagnetite or magnetite), the condition of magnetic uniaxiality is satisfied when the elongation $q \geq 1.2$.

As well as in Section 1, we build an expression for the total free energy, which includes

- (i) crystalline anisotropy energy, which, according to [62], depending on the sign of the constant of anisotropy has the following form:

$$\begin{aligned} E_A &= \begin{cases} \frac{1}{4} k_{A1}^{(1)} (I_s^{(1)})^2 (1 - \varepsilon) \sin^2 2\vartheta^{(1)} + \frac{1}{4} k_{A1}^{(2)} (I_s^{(2)})^2 \times \varepsilon \sin^2 2\vartheta^{(2)}, & k_{A1}^{(1)} > 0, \quad k_{A1}^{(2)} > 0, \\ \frac{1}{54} k_{A2}^{(1)} (I_s^{(1)})^2 (1 - \varepsilon) \sin^2 \vartheta^{(1)} (1 + 2 \cos 2\vartheta^{(1)})^2 + \frac{1}{54} k_{A2}^{(2)} (I_s^{(2)})^2 \varepsilon \sin^2 \vartheta^{(2)} (1 + 2 \cos 2\vartheta^{(2)})^2, & k_{A1}^{(1)} < 0, \quad k_{A1}^{(2)} < 0, \\ \frac{1}{4} k_{A1}^{(1)} (I_s^{(1)})^2 (1 - \varepsilon) \sin^2 2\vartheta^{(1)} + \frac{1}{54} k_{A2}^{(2)} (I_s^{(2)})^2 \varepsilon \sin^2 \vartheta^{(2)} (1 + 2 \cos 2\vartheta^{(2)})^2, & k_{A1}^{(1)} > 0, \quad k_{A1}^{(2)} < 0, \\ \frac{1}{54} k_{A2}^{(1)} (I_s^{(1)})^2 (1 - \varepsilon) \sin^2 \vartheta^{(1)} (1 + 2 \cos 2\vartheta^{(1)})^2 + \frac{1}{4} k_{A1}^{(2)} (I_s^{(2)})^2 \varepsilon \sin^2 2\vartheta^{(2)}, & k_{A1}^{(1)} < 0, \quad k_{A1}^{(2)} > 0, \end{cases} \end{aligned} \quad (17)$$

- (iii) energy of a grain in the field of mechanical stress

$$\begin{aligned} E_\sigma &= \frac{1}{2} (I_s^{(1)})^2 (1 - \varepsilon) \Lambda^{(1)} \sigma \sin^2 \vartheta^{(1)} \\ &\quad + \frac{1}{2} (I_s^{(2)})^2 \varepsilon \Lambda^{(2)} \sigma \sin^2 \vartheta^{(2)}, \end{aligned} \quad (18)$$

energy of magnetostatic interaction E_m , interfacial exchange interaction E_{ex} , and energy of the magnetic moment of

TABLE 1: Magnetic characteristics of two-phase nanoparticles.

	Fe ₃ O ₄	Fe _{2.44} Ti _{0.56} O ₄	CoFe ₂ O ₄	γ-Fe ₂ O ₃
I_s , G	480 [60]	151 [60]	425 [61]	350 [44]
K_1 , 10 ⁵ erg/cm ³	-1.06 [60, 61]	-0.7 [60]	27 [61]	0.46 [44]
K_2 , 10 ⁵ erg/cm ³	2.8 [60, 61]	-0.15 [60]	30 [61]	52 [56]
$\lambda_{100} \cdot 10^6$	-20 [60, 62]	170 [60, 62]	225 [63]	-5* [64]
$\lambda_{111} \cdot 10^6$	78 [60, 62]	92 [60, 62]	-335 [63, 65]	-5* [64]

* indicates isotropic constants of magnetostriction λ_s .

a particle in the external magnetic field E_H defined by the relations (2), (4), and (5), respectively. In the relations (17) and (18) $k_{A1}^{(1,2)} = K_1^{(1,2)} / (I_s^{(1,2)})^2$ and $k_{A2}^{(1,2)} = K_2^{(1,2)} / (I_s^{(1,2)})^2$ are dimensionless constant of crystalline anisotropy of the first or second order of the first or second phase, respectively, $K_1^{(1,2)}$ and $K_2^{(1,2)}$ are the anisotropy constants of the first and second order of a cubic crystal, and $\Lambda^{(1,2)} = 3\lambda_{100}^{(1,2)} / (I_s^{(1,2)})^2$, where $k_{A1}^{(1,2)} > 0$ and $\Lambda^{(1,2)} = 3\lambda_{111}^{(1,2)} / (I_s^{(1,2)})^2$, where $k_{A1}^{(1,2)} < 0$, $\lambda_{100}^{(1,2)}$ and $\lambda_{111}^{(1,2)}$ are the constants of phases' magnetostriction.

According to the second condition stated above, magnetic states of a multiaxis two-phase particle are equivalent to states of a uniaxial two-phase particle (see Section 2), whose effective anisotropy constants $K^{(1)}$ and $K^{(2)}$ are expressed through the constants of a multiaxial crystal:

$$\bar{K}^{(1)} = \begin{cases} k_{A1}^{(1)}(1-\varepsilon) + (1-2\varepsilon)k_N^{(1)} + \varepsilon k_N^{(2)} \\ \quad + \Lambda^{(1)}\sigma(1-\varepsilon), & k_{A1}^{(1)} > 0, \\ \frac{k_{A2}^{(1)}}{3}(1-\varepsilon) + (1-2\varepsilon)k_N^{(1)} + \varepsilon k_N^{(2)} \\ \quad + \Lambda^{(1)}\sigma(1-\varepsilon), & k_{A1}^{(1)} < 0, \end{cases} \quad (19)$$

$$\bar{K}^{(2)} = \begin{cases} \varepsilon(k_{A1}^{(2)} + k_N^{(2)} + \Lambda^{(2)}\sigma), & k_{A1}^{(2)} > 0, \\ \varepsilon\left(\frac{k_{A2}^{(2)}}{3} + k_N^{(2)} + \Lambda^{(2)}\sigma\right), & k_{A1}^{(2)} < 0. \end{cases}$$

Expressions (11)–(16) and (19) allow us to study the effect of mechanical stress and geometric (q and ε) and ($I_s^{(1,2)}$, $k_{A1,2}^{(1,2)}$, $\Lambda^{(1,2)}$, A_{in}) characteristics on the magnetic state of both single-axis and multiaxis crystallographic two-phase nanoparticles.

5. Diagrams of the Magnetic States of Heterophase Nanoparticles

For comparison, let us consider two types of two-phase nanoparticles, whose magnetic characteristics are presented in Table 1: relatively low coercitive nanoparticles of magnetite (first phase) that include titanomagnetite [3, 60] (Fe₃O₄-Fe_{2.44}Ti_{0.56}O₄) and high coercitive nanoparticles γ-Fe₂O₃ (second phase) epitaxially cobalt ferrite coated [39–41] (CoFe₂O₄-γ-Fe₂O₃).

Critical fields of magnetic reversal (11)–(16) depend essentially on the elongation q and the relative volume of one

of the ε particle's phases. Moreover, for some q and ε values the critical fields can be negative. Negativity H_c should be comprehended as the inability to implement the metastable state controlled by this critical field. This statement may be interpreted in the following way. If an ensemble is made up of particles with different relative sizes ε and elongation q of the second phase, the point A on the phase diagram $\{\varepsilon, q\}$ (see Figure 2) can be associated with a particle that at $H_c > 0$ (dark area) can be in either metastable or ground state and only in the ground state if $H_c < 0$ (light area).

Note that natural limitation of the volume of introduction (of the second phase) $-\nu \leq V$ and the relation between large semimajor axes of particles' ellipsoids and implementation $a_2 \leq a_1$ impose restrictions on the choice of q and ε . Indeed, if we use expressions for the volume and the long axis of the elongated ellipsoid ($\nu = (4\pi/3)qb_2^3$, $V = (4\pi/3)q_1b_1^3$, $a_1 = q_1b_1a_2 = qb_2$, b —minor axis of the ellipsoid), we can obtain the following relation $q_1\varepsilon \leq q \leq q_1/\sqrt{\varepsilon}$. Obviously, the range of permissible values of q and ε depends significantly on the elongation of nanoparticles q_1 (see Figure 2). The maximum value of $q = q_m$ and is limited with the value $q_m = 2\pi$, since the demagnetizing factor $N_z(q)$ decreases rapidly with q ($N_z(2\pi) \approx 0.02$) increasing.

In the study of the effect of mechanical stress on the magnetic states and magnetization processes of an ensemble of two-phase nanoparticles we use a dimensionless constant

$$k_\sigma = \begin{cases} \frac{3\lambda_{100}^{(1)}\sigma}{k_{A1}^{(1)}(I_s^{(1)})^2}, & k_{A1}^{(1)} > 0, \\ \frac{3\lambda_{111}^{(1)}\sigma}{|k_{A1}^{(1)}|(I_s^{(1)})^2}, & k_{A1}^{(1)} < 0. \end{cases} \quad (20)$$

As shown in Table 1, stress signs σ and k_σ match each other for the particles (Fe₃O₄-Fe_{2.44}Ti_{0.56}O₄), while negative values ($\sigma > 0$) correspond to tensile stress and positive values correspond to compression stress for cobalt-coated particles γ-Fe₂O₃.

Tables 2 and 3 show the basic diagram and the two-phase particles of the metastable states (CoFe₂O₄-γ-Fe₂O₃) and (Fe₃O₄-Fe_{2.44}Ti_{0.56}O₄), respectively, differing magnitude interfacial exchange interaction A_{in} compressive ($\sigma < 0$) or tensile ($\sigma > 0$) stresses. The particles, which were representative points in the phase diagram $\{\varepsilon, q\}$ in the region highlighted in the dark color, can be both in the ground and metastable states (with a parallel or antiparallel orientation of the magnetic moments of the phases). The points in the light

FIGURE 2: An illustration of the phase diagram $\{\varepsilon, q\}$. A two-phase ellipsoidal nanoparticle with the volume V and elongation q_1 containing ellipsoidal inclusion with the volume $v = \varepsilon V$ and elongation q corresponds to point A . Particles that represent the points in the dark area may be in either ground or metastable state, as their critical fields $H_c^{(1 \rightarrow 2)} > 0$ in the white area are in the ground state ($H_c^{(1 \rightarrow 2)} < 0$).

FIGURE 3: Dependence of the degree of metastability α_m of the system of nanoparticles CoFe_2O_4 - γ - Fe_2O_3 (a) and Fe_3O_4 - $\text{Fe}_{2.44}\text{Ti}_{0.56}\text{O}_4$ (b) on the value of the interfacial exchange interaction A_{in} and relative stress k_σ . Curves 1, 2, and 3 are calculated at $k_\sigma = -0.8$, $k_\sigma = 0$, and $k_\sigma = 0.8$, respectively.

area correspond to particles in its ground part in Table 1 as footnote. Please check state in which depending on the size of the interfacial exchange interaction A_{in} magnetic moments are parallel or antiparallel phase with each other. Figures in the second column 2 and the second row of Table 3 illustrate the effect on the metastability A_{in} biphasic nanoparticles $\sigma = 0$.

Let us define the degree of metastability α_m of the nanoparticles system as the ratio of the areas: the dark side of the chart to the general—dark and light. Figure 3 shows that

the metastable state is realized only at $A_{\text{in min}} \leq A_{\text{in}} \leq A_{\text{in max}}$, and the degree of metastability α_m reaches its maximum at a point $A_{\text{in}}^{(m)}$. And, if the magnetic moments of the phases are antiparallel in the ground state at $A_{\text{in}} \leq A_{\text{in}}^{(m)}$, then at $A_{\text{in}} > A_{\text{in}}^{(m)}$ they are parallel.

Table 2 and Figure 3(a) show that the stress does not affect the metastability of cobalt-coated particles γ - Fe_2O_3 . Such a weak dependence of the magnetic states on the stress is associated with high values of anisotropy and shape anisotropy. The effect of mechanical stress on the degree

TABLE 2: A diagram of the ground (light fill area) and metastable (dark fill area) states of the two-phase elongated ($q_1 = 3$) nanoparticles (CoFe_2O_4 - γ - Fe_2O_3) system located in the field of mechanical stress σ . Positive value $k_\sigma = 0.8$, that is, the right column of the table, corresponds to compressive stress; $k_\sigma = -0.8$ column at $k_\sigma = 0$ no stress (center column) corresponds to tensile stress. Table rows correspond to different values of interfacial interaction $A_{\text{in}} = \alpha \cdot 10^{-8}$ Erg/cm. Arrows indicate the state of parallel ($\uparrow\uparrow$) (the first and the third) and antiparallel ($\uparrow\downarrow$) (the second and the fourth) orientation of the magnetic moments of nanoparticles' phases.

TABLE 2: Continued.

FIGURE 4: Illustration of the calculation of the population of states. The dark region of the picture is area where nanoparticle may move from the first state to the second if the critical field is smaller than the magnetic field.

of nanoparticles ($\text{Fe}_3\text{O}_4\text{-Fe}_{2.44}\text{Ti}_{0.56}\text{O}_4$) metastability is more significant (see Table 3 and Figure 3(b)). Tension increases both the degree of metastability and the range of the constants of interfacial exchange interaction $[A_{\text{in min}}, A_{\text{in max}}]$ where metastable states are implemented (see Figure 3(b)); compression leads to the opposite effect. Described dependence of metastability on mechanical stresses was expected. Since in accordance with the relations (19), an increase in tensile stress increases the effective constant of anisotropy and, therefore, to expansion of the area of metastable states, while compressive stress reduces metastability (see Figure 3(b)).

6. Magnetization of the System of Two-Phase Nanoparticles

As shown in the previous section, a two-phase particle in one of the four magnetic states can be represented by a point on a diagram $\{\epsilon, q\}$ (see Figure 2). We assume that two-phase nanoparticles are uniformly distributed with respect to the relative volume of the second phase ϵ ($0 \leq \epsilon \leq 1$) and its elongation q ($0 \leq q \leq 2\pi$). Then the points representing these nanoparticles are distributed on the diagram $\{\epsilon, q\}$ uniformly. Therefore, to calculate the dependence of a number of particles $N_k(h)$ ($k = 1, \dots, 4$) located in each of the four magnetic states of the magnetic field h we should integrate it with respect to the diagram area $D(0 \leq H_c^{(i \rightarrow k)} \leq h)$ that satisfies conditions $0 \leq H_c^{(i \rightarrow k)} \leq h$ (see Figure 4).

So in the case of noninteracting nanoparticles the magnetic states vector population of the two-phase particles $\mathbf{N}(h) = \{N_1(h), N_2(h), N_3(h), N_4(h)\}$ is defined as

$$\begin{aligned}
 N_1(h) &= N_1(0) + \iint n_2 D(0 \leq H_c^{(2 \rightarrow 1)} \leq h) d\epsilon dq \\
 &\quad + \iint n_3 D(0 \leq H_c^{(3 \rightarrow 1)} \leq h) d\epsilon dq \\
 &\quad + \iint n_4 D(0 \leq H_c^{(4 \rightarrow 1)} \leq h) d\epsilon dq, \\
 N_2(h) &= N_2(0) + \iint n_3 D(0 \leq H_c^{(3 \rightarrow 2)} \leq h) d\epsilon dq \\
 &\quad + \iint (n_4 - n_2) D(0 \leq H_c^{(2 \rightarrow 4)} \leq h) d\epsilon dq \\
 &\quad - \iint n_2 D(0 \leq H_c^{(2 \rightarrow 1)} \leq h) d\epsilon dq,
 \end{aligned}$$

TABLE 3: Diagrams of the ground (light fill area) and metastable (dark fill region) states of the two-phase elongated ($q_1 = 3$) nanoparticles ($\text{Fe}_3\text{O}_4\text{-Fe}_{2.44}\text{Ti}_{0.56}\text{O}_4$) system characterized by an interfacial exchange interaction A_{in} in the field of mechanical stress σ . The notations are the same as in Table 2.

$$\begin{aligned}
N_3(h) &= N_3(0) - \iint n_3 D(0 \leq H_c^{(3 \rightarrow 1)} \leq h) d\varepsilon dq \\
&\quad - \iint n_3 D(0 \leq H_c^{(3 \rightarrow 2)} \leq h) d\varepsilon dq \\
&\quad - \iint n_3 D(0 \leq H_c^{(3 \rightarrow 4)} \leq h) d\varepsilon dq, \\
N_4(h) &= N_4(0) + \iint n_3 D(0 \leq H_c^{(3 \rightarrow 4)} \leq h) d\varepsilon dq \\
&\quad - \iint n_4 D(0 \leq H_c^{(4 \rightarrow 1)} \leq h) d\varepsilon dq \\
&\quad - \iint (n_4 - n_2) D(0 < H_c^{(2 \rightarrow 4)} \leq h) d\varepsilon dq.
\end{aligned} \tag{21}$$

$n_i = \{n_1, \dots, n_4\}$ here is the density of distribution on the diagram $\{\varepsilon, q\}$ of a number of particles in i -M state and $N_i(0)$ is the number of particles in one of the four equilibrium states at $h = 0$. $N_i(0)$ and n_i , and they are related as follows:

$$\begin{aligned}
N_1(0) &= \iint n_1 D(0 \leq H_c^{(1 \rightarrow 2)}, 0 \leq H_c^{(1 \rightarrow 3)}, \\
&\quad 0 \leq H_c^{(1 \rightarrow 4)}) d\varepsilon dq \\
&\quad + \iint n_2 D(H_c^{(2 \rightarrow 1)} \leq 0) d\varepsilon dq \\
&\quad + \iint n_3 D(H_c^{(3 \rightarrow 1)} \leq 0) d\varepsilon dq \\
&\quad + \iint n_4 D(H_c^{(4 \rightarrow 1)} \leq 0) d\varepsilon dq, \\
N_2(0) &= \iint n_1 D(H_c^{(1 \rightarrow 2)} \leq 0) d\varepsilon dq \\
&\quad + \iint n_2 D(0 \leq H_c^{(2 \rightarrow 1)}, 0 \leq H_c^{(2 \rightarrow 3)}, \\
&\quad 0 \leq H_c^{(2 \rightarrow 4)}) d\varepsilon dq \\
&\quad + \iint n_3 D(H_c^{(2 \rightarrow 3)} \leq 0) d\varepsilon dq \\
&\quad + \iint n_4 D(H_c^{(2 \rightarrow 4)} \leq 0) d\varepsilon dq, \\
N_3(0) &= \iint n_1 D(H_c^{(1 \rightarrow 3)} \leq 0) d\varepsilon dq \\
&\quad + \iint n_2 D(H_c^{(2 \rightarrow 3)} \leq 0) d\varepsilon dq \\
&\quad + \iint n_3 D(0 \leq H_c^{(3 \rightarrow 1)}, 0 \leq H_c^{(3 \rightarrow 2)}, \\
&\quad 0 \leq H_c^{(3 \rightarrow 4)}) d\varepsilon dq
\end{aligned}$$

$$\begin{aligned}
&\quad + \iint n_4 D(H_c^{(4 \rightarrow 3)} \leq 0) d\varepsilon dq, \\
N_4(0) &= \iint n_1 D(H_c^{(1 \rightarrow 4)} \leq 0) d\varepsilon dq \\
&\quad + \iint n_2 D(H_c^{(2 \rightarrow 4)} \leq 0) d\varepsilon dq \\
&\quad + \iint n_3 D(H_c^{(3 \rightarrow 4)} \leq 0) d\varepsilon dq \\
&\quad + \iint n_4 D(0 \leq H_c^{(4 \rightarrow 1)}, 0 \leq H_c^{(4 \rightarrow 2)}, \\
&\quad 0 \leq H_c^{(4 \rightarrow 3)}) d\varepsilon dq.
\end{aligned} \tag{22}$$

n_i can be defined from the condition that at $h = 0$ in the state of zero magnetization the first and third as well as second and fourth states of nanoparticles are equivalent:

$$\begin{aligned}
N_1(0) &= N_3(0), \quad N_2(0) = N_4(0), \\
N_1(0) + N_2(0) &= \frac{N}{2}, \\
n_1 + n_2 + n_3 + n_4 &= \frac{N}{\iint D(\{\varepsilon, q\}) d\varepsilon dq},
\end{aligned} \tag{23}$$

where N is the number of particles and $\iint D(\{\varepsilon, q\}) d\varepsilon dq$ is the total area of the diagram $\{\varepsilon, q\}$ (see Figure 2).

Relations (21)–(23) defining the population vector $\mathbf{N}(h)$ allow for calculating the value of magnetization of the two-phase nanoparticles system:

$$\begin{aligned}
I(h) &= c \left[(N_1(h) - N_3(h)) \left((1 - \varepsilon) I_s^{(1)} + \varepsilon I_s^{(2)} \right) \right. \\
&\quad \left. + (N_2(h) - N_4(h)) \left((1 - \varepsilon) I_s^{(1)} - \varepsilon I_s^{(2)} \right) \right],
\end{aligned} \tag{24}$$

where $c = NV/V_0$ is the volume concentration of the magnetic phase and V_0 is the volume that the system occupies.

7. The Effect of Mechanical Stress on the Magnetization Curve and Hysteresis Characteristics of an Ensemble of Cobalt-Coated Nanoparticles γ -Fe₂O₃

Calculation of the magnetization curve and hysteresis loop held with the help of relations (11)–(16) and (21)–(24) is shown in Figure 5.

Dependence of the coercive force H_c on the relative volume of cobalt coating $\tau = 1 - \varepsilon$ is determined not only by mechanical stress but also by the interfacial exchange A_{in} (see Figure 6).

It is easy to see that tension shifts magnetization curves to lower magnetic H , but compression has the opposite effect—magnetization curves shift to larger fields H relatively to the undeformed state. Thus, the mechanical stress does not affect the saturation magnetization I_s , which coincides with the

FIGURE 5: Dependence of magnetization of the elongated nanoparticles system (with the following characteristics: $q_1 = 3$, $A_{in} = 0$) on the magnetic field H , relative volume of the cobalt coating $\tau = 1 - \varepsilon$, and relative mechanical stress k_σ .

saturation remanence I_{rs} and decreases with increasing thickness of the membrane irritation. Recent experiments conducted on the core/shell nanoparticles of hematite/magnetite showed that I_s is almost independent of stress [68]. We obtained that the dependence of the saturation magnetization of the relative amount of cobalt coating τ is in qualitative agreement with experimental data [69]. I_s reduction system core/shell nanoparticles with increasing thickness of the shell due to the fact that with increasing τ is increased contribution less magnetic material and a magnetic role decline.

Dependence of the coercive force H_c on the relative volume of cobalt coating $\tau = 1 - \varepsilon$ is determined not only by mechanical stress but also by the interfacial exchange A_{in} (see Figure 6). In addition, the negative exchange interaction leads to a reduction in general, and the positive increases H_c relative to the state $A_{in} = 0$. We calculated the dependence of the coercive force of τ which is in qualitative agreement with experimental data [70]. Where shown, the dependence of the coercive force is not determined by the particle size and the relative thickness of coating core/shell nanoparticles.

We also note a qualitative agreement with experimental data [68]; we obtained depending H_c from mechanical stresses. In the same paper was received experimental linear dependence of the effective anisotropy constant of stress which is in good agreement with the results obtained by

us (see (19)). Dependence characteristics of the coercive force on the interfacial exchange interaction A_{in} are shown in Figure 7. Obviously, the nonmonotonic behavior $H_c = H_c(A_{in})$ is typical for nanoparticles with a large ($\tau = 0.9$) or low ($\tau = 0.1$) cobalt coating thickness and implemented in both positive and negative values A_{in} .

Since the coercive force is determined by the minimum of the range of critical fields, the nonmonotonic behavior H_c is related to the difference in the dependence of these fields on the exchange interaction constant A_{in} (see (10) and (11)–(16)). The above mentioned nonmonotonic behavior H_c was not observed in [39, 65, 66, 71], which is obviously associated with a narrower range of critical fields of magnetization reversal of two-phase particles than that in this study. A qualitative comparison of the results with the similar calculations presented in the works [8, 54, 69, 71, 72] shows that coercivity increases up to saturation along with the increase in the amount of CoFe_2O_4 phase, just as it was mentioned in those works.

8. Conclusions

In contrast to earlier models of two-phase nanoparticles [56–59, 67, 73] the model presented in this work allows for studying the effect of mechanical stress and magnetic field on the magnetic state of both uniaxial and multiaxial heterophase

FIGURE 6: Dependence of the relative coercive force $h_c = H_c(\sigma)/H_{cm}(\sigma = 0)$ of the elongated nanoparticles system ($q_1 = 3$, $H_{cm}(\sigma = 0) = 2947$ Oe) on the relative amount of cobalt coating τ , relative mechanical stress k_σ , and the interfacial exchange A_{in} .

FIGURE 7: Dependence of the relative coercive force $h_c = H_c/H_{c1}$ of the elongated nanoparticles system (with the following characteristics: $q_1 = 3$, $H_{cm} = 2947$ Oe) on the value of the interfacial exchange A_{in} , relative volume of cobalt coating τ , and relative mechanical stress, 1 - $\tau = 0.9$, 2 - $\tau = 0.1$, 3 - $\tau = 0.5$.

magnets. The estimated range of the critical fields of magnetization reversal of the phases' magnetic moments allowed for building phase diagrams and evaluating the effect of mechanical stress on the degree of metastability of nanoparticles' magnetic states. In the framework of our model, using an example of epitaxially cobalt-coated particles $\gamma\text{-Fe}_2\text{O}_3$ we

carried out a theoretical analysis of the effect of uniaxial stress on the magnetization process of the system of noninteracting heterophase nanoparticles. As it was shown tension leads to reduction and compression increases coercive force H_c , while the residual saturation magnetization I_{rs} does not change under the influence of mechanical stress. The coercive force

of the nanoparticles system changes in a nonmonotonic way as far as the interfacial exchange interaction increases and strongly depends on the thickness of cobalt coating.

The simulation of the effect of mechanical stresses on the magnetization of the system of two-phase single-domain noninteracting grains carried out in this study led to consistent results, which allows for using our model in analyzing magnetic properties of an ensemble of heterophase nanoparticles.

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