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Theory of Magnetization of Two-Phase Superparamagnetic Particles: II. Modeling of Magnetization Processes

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Abstract—A model is built for fine chemically inhomogeneous two-phase ferrimagnetic particles. A technique is developed to calculate the total and remanent magnetizations and the coercivity of superparamagnetic particles.

INTRODUCTION

In the previous paper [1], the interphase magnetostatic interaction is shown to have a strong impact on the critical fields of magnetization reversal and, consequently, on the metastability of magnetic states in which a grain exists when placed in an external field **H**. The interaction of such grains in an ensemble should produce a similar result, and this may, in the final analysis, affect the magnetization process.

Quite a number of authors have dealt with the magnetostatic interaction observed in a system of chemically homogeneous superparamagnetic grains (see, for example, [2–6]). In our analysis of magnetic interaction in an ensemble of chemically inhomogeneous particles, we will use the random field method [3, 5].

1. THE INTERACTION-FIELD DISTRIBUTION FUNCTION

Consider an ensemble of $N + 1$ two-phase particles scattered at random in a nonmagnetic matrix and interacting as dipoles. Suppose that the particle of interest is located at the origin of the coordinate system. Given a volume particle distribution such that we can find a particle with a magnetic moment m_i in each element of volume dV_i, the probability that the projection of the interaction field H' onto the direction $\mathbf{l}_0$ selected by the applied field $\mathbf{H}$ in the interval H', $H' + dH'$ will be found at the origin is

$$\delta\left[H' - \sum_{i=1}^{N} \varphi_i(\mathbf{m}_i, \mathbf{r}_i)\right] dH',$$

where φ_i is the projection onto the direction $\mathbf{l}_0$ of the field set up by a particle of magnetic moment $\mathbf{m}_i$ at point $\mathbf{r}_i$. Then the total probability of finding the field in the interval H', $H' + dH'$ will be

$$W(H')dH' = \frac{1}{V^N}\int \delta(H' - \sum \varphi_i) \times \prod_{i=1}^{N} \tau(\mathbf{m}_i) d\mathbf{m}_i dV_i, \quad (1)$$

where $(dV_i/V)\tau(\mathbf{m}_i)d\mathbf{m}_i$ is the probability of finding a particle with a magnetic moment $\mathbf{m}_i$, $\mathbf{m}_i + d\mathbf{m}_i$ in a volume element dV_i, and $\tau(\mathbf{m}_i)$ is the particle distribution function in the specimen over the magnitude and direction of the magnetic moment $\mathbf{m}$. If the magnetic moment of the grain is aligned with or against the direction selected by the applied field, this distribution function may be expressed in terms of the occupancy of magnetic states N_i (see [1]) as

$$\begin{aligned}\tau(\mathbf{m}_i)d\mathbf{m}_i = \int \{ & N_{1i}(H' + H)\delta(m_i - m_1)\delta(\gamma_i) \\ & + N_{2i}(H' + H)\delta(m_i - m_2)\delta(\gamma_i) \\ & + N_{3i}(H' + H)\delta(m_i - m_1)\delta(\gamma_i - \pi) \\ & + N_{4i}(H' + H)\delta(m_i - m_2)\delta(\gamma_i - \pi)\} \\ & \times dm_i \frac{d\gamma_i}{2\pi} W(H')dH', \end{aligned} \quad (2)$$

where γ_i is the angle between the direction of $\mathbf{m}_i$ and $\mathbf{l}_0$; $(N_{1i}, N_{2i}, N_{3i}, N_{4i})$ is the state vector of the ensemble of two-phase particles; and

$$m_1 = a^3 q[I_{s1}(1 - \varepsilon) + I_{s2}\varepsilon],$$
$$m_2 = a^3 q[I_{s1}(1 - \varepsilon) - I_{s2}\varepsilon]$$

are the magnetic moments of the corresponding phases. The characteristic function $A(\rho) = \int W(H')\exp(i\rho H')dH'$,

which is the Fourier transform of the function $W(H')$, takes the form

$$A(\rho) = \frac{1}{V^N}\int\ldots\int\exp\left(i\rho\sum_i\varphi_i\right)\prod_i\tau(\mathbf{m}_i)d\mathbf{m}_i dV_i. \quad (3)$$

Using the normalization condition $\int\tau(\mathbf{m}_i)d\mathbf{m}_i = 1$ and assuming that, with N tending to infinity, the number n of particles per unit volume remains unchanged (the approximation of low concentrations), equation (3) may be recast as

$$A(\rho) = \left\{1 - \frac{n}{N}\int(1-\exp(i\rho\varphi)) \times\tau(\mathbf{m})d\mathbf{m}dV\right\}^N \longrightarrow e^{-c(\rho)}, \quad (4)$$

$$c(\rho) = n\int dV\int(1-\exp(i\rho\varphi))\tau(\mathbf{m})d\mathbf{m}, \quad (5)$$

$$\varphi = -\frac{m\cos\gamma(1-3\cos^2\nu)}{r^3}, \quad (6)$$

where ν is the angle between $\mathbf{l}_0$ and the radius vector of the particle.

Integration in (5) yields

$$c(\rho) = n\int[N_1 I_-(m_1,\rho) + N_2 I_-(m_2,\rho) + N_3 I_+(m_1,\rho) + N_4 I_+(m_2,\rho)]W(H')dH', \quad (7)$$

where

$$I_\pm(m,\rho) = \int\left\{1-\exp\left[\pm\frac{i\rho m}{r^3}(1-3\cos^2\nu)\right]\right\}dV. \quad (8)$$

Noting that $I_\pm(m,\rho) = bm|\rho| \pm iam\rho$, where $b \approx 5$, $a = 4\pi/3 - N + 4\pi/15$, and N is the demagnetizing factor, we obtain the Cauchy distribution function

$$W(H')dH' = \frac{dH'}{\pi B\left\{1+\left[\frac{(H'+aI)}{B}\right]^2\right\}}, \quad (9)$$

where the distribution parameter B and the magnetization I are defined by a set of equations of the form

$$B = bc\int\{(N_1+N_3)[I_{s1}(1-\varepsilon)+I_{s2}\varepsilon] + (N_2+N_4)|I_{s1}(1-\varepsilon)-I_{s2}\varepsilon|\}W(H')dH', \quad (10)$$

$$I = c\int\{(N_1-N_3)[I_{s1}(1-\varepsilon)+I_{s2}\varepsilon] + (N_2-N_4)[I_{s1}(1-\varepsilon)-I_{s2}\varepsilon]\}W(H')dH'. \quad (11)$$

Here, $c = na^3q$ is the volume concentration of the ferromagnetic material.

The above simultaneous equations, taken together with the relations defining the occupancy vector $\mathbf{N}$ (see equations (17) and (18) in [1]), make it possible to estimate the Cauchy distribution parameter B and to calculate the magnetization I.

2. MODELING THE MAGNETIZATION OF AN ENSEMBLE OF NONINTERACTING PARTICLES

At low particle concentrations (with c tending to zero), the interaction field distribution function $W(H')$ tends to $\delta(H')$, where $\delta(H')$ is the Dirac delta function. Integrating equation (11) yields an expression that defines the magnetization of an ensemble of identical noninteracting particles

$$I = c[(I_{s1}(1-\varepsilon)+I_{s2}\varepsilon)(N_1-N_3) + (I_{s1}(1-\varepsilon)-I_{s2}\varepsilon)(N_2-N_4)]. \quad (12)$$

Now, we take advantage of the spontaneous magnetization $I_s = I_s(x)$ and the crystallographic anisotropy $k = k(x)$ found by experiment in [7] for $Co_xZn_{2-x}Ba_3Fe_{24}O_4$ ferrites as functions of cobalt concentration x. They can be linearly interpolated as

$$I_s(x) = (-20x + 385)\ \text{G},$$

$$k(x) = 8.3 - 13.3x.$$

Assume that the first phase is depleted of and the second is enriched in cobalt, that is, $x_1 < x_2$.

2.1. Relaxation Time and Viscous Magnetization of an Ensemble of Interacting Particles

The three eigenvalues of the transformed transition matrix $\mathbf{W}$ (see [1]) found from the equation $\det|\mathbf{W} - \lambda\mathbf{E}| = 0$, where $\mathbf{E}$ is the unit matrix, may be interpreted as the inverse relaxation times τ_i. Here, the smallest τ corresponds to the "lifetime" of the maximally unstable state and the longest relaxation time τ_{max}, to the most stable state.

Figure 1a shows calculated curves of the reduced relaxation time τ_0 of a stable state ($\tau_0 = \tau_{max}f_0$) for an ensemble of two-phase particles plotted against the concentration of Co atoms in the first phase. A decrease in chemical inhomogeneity of the grain (an increase in x_1) is seen to cause a decrease in τ_0. This behavior of the relaxation time is related to a decrease in $I_s(x)$ and in the crystallographic anisotropy constant $k(x)$. An increase in the volume ε of the second phase is seen to be accompanied by a decrease in τ_0 because the potential barriers E_{ik} are lowered (see [1, Appendix I]). Therefore, the viscous magnetization of chemically homogeneous particles ($\varepsilon = 0$) increases faster than does that of inhomogeneous particles ($\varepsilon = 0.2$) (see Fig. 1b).

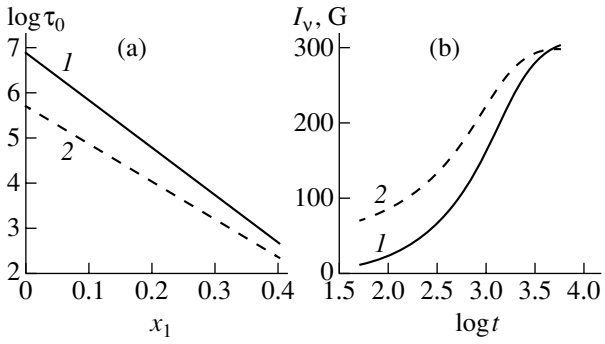

Fig. 1. Curves of (a) relaxation time τ_0 and (b) viscous magnetization plotted against cobalt concentration x_1 for an ensemble of (*1*) chemically homogeneous particles ($\varepsilon = 0$) and (*2*) of chemically inhomogeneous particles ($\varepsilon = 0.2$) with $x_2 = 0.5$, $a = 10$ nm, and $q = 1.5$.

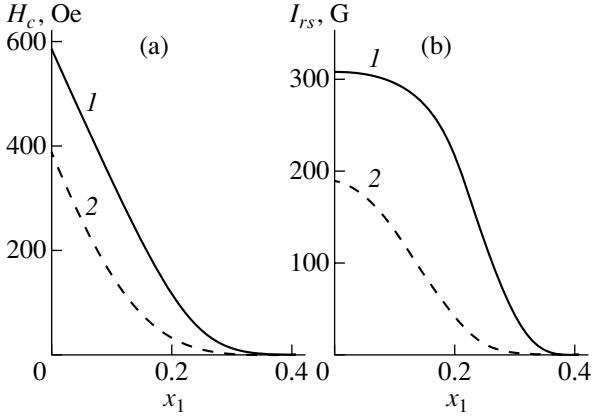

Fig. 2. Curves of (a) coercive force H_c and (b) remanent saturation magnetization I_{rs} plotted against cobalt concentration x_1 ($a = 10$ nm, $x_2 = 0.5$, and $q = 1.5$): (*1*) $\varepsilon = 0$ and (*2*) $\varepsilon = 0.2$.

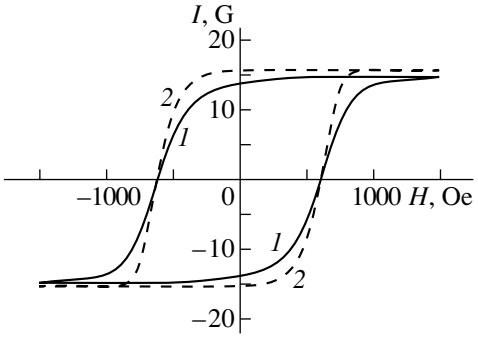

Fig. 3. Hysteresis loops for an ensemble of chemically homogeneous noninteracting particles (*1*) and interacting particles (*2*) with $q = 1.5$, $a = 10$ nm, and $x_1 = 0$.

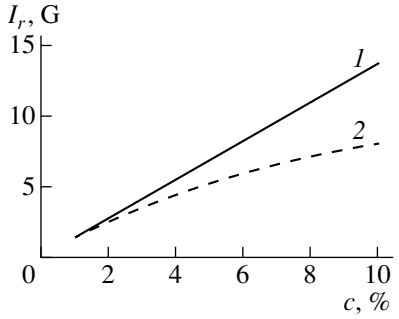

Fig. 4. Remanent magnetization I_r plotted against the volume concentration c of two-phase grains in an ensemble of noninteracting grains (*1*) and interacting grains (*2*) with $q = 1.5$, $a = 10$ nm, $x_1 = 0$, $x_2 = 0.5$, $H = 1000$ Oe, and $\varepsilon = 0.2$.

2.2. Hysteretic Properties and Remanent Magnetization of an Ensemble of Noninteracting Two-Phase Grains

When residing in a superparamagnetic state, fine particles can exhibit hysteresis only if the time elapsing after the removal of the applied magnetic field H or the time it takes for the field to fall off to $H = 0$ is shorter than the relaxation time. Naturally, as the grain size increases, such a situation can arise in the interval of reasonable spans of time (comparable with the observation time).

Figure 2 shows the coercive force H_c and the remanent saturation magnetization I_{rs} plotted against the concentration of the first phase, based on the modeling of hysteresis loops (see Fig. 3) with equation (12). As should be expected, an increase in the chemical inhomogeneity of the grain is seen to be accompanied by a decrease in H_c and I_{rs}, which is in turn associated with a decrease in the relaxation time τ_0.

3. EFFECT OF INTERACTION ON THE REMANENT MAGNETIZATION AND COERCIVITY OF AN ENSEMBLE OF CHEMICALLY INHOMOGENEOUS PARTICLES

Before we proceed to analyze the results of mathematical modeling as applied to the magnetization of an ensemble of interacting two-phase particles, it is worth noting that the effect of magnetostatic interaction is to randomize the magnetic moment distribution of particles. It is, therefore, natural to expect that in an ensemble of interacting particles the remanent magnetization should be lower compared to an ensemble of similar particles where the interaction is neglected. This assertion is illustrated in Fig. 4, which shows calculated curves of the remanent magnetization I_r of noninteracting particles (curve *1*) and of interacting particles (curve *2*).

This randomizing effect of the magnetostatic interaction brings on a decrease in the coercivity, this decrease in H_c being most significant in an ensemble of

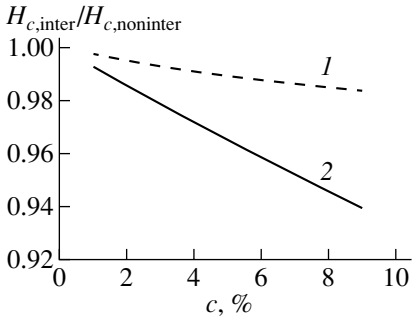

Fig. 5. Reduced coercive force $H_{c,\text{inter}}/H_{c,\text{noninter}}$ plotted against volume concentration c for chemically homogeneous particles (*1*) and inhomogeneous particles (*2*, $\varepsilon = 0.2$) with $q = 1.5$, $a = 10$ nm, $x_1 = 0$, and $x_2 = 0.5$.

chemically inhomogeneous particles (Fig. 5) whose relaxation time is shorter than that of an ensemble of chemically homogeneous particles.

The proposed model offers an insight into the magnetization and magnetic properties of a system of fine chemically inhomogeneous ferrimagnetic particles.

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