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The Metastable States of a Pseudo-Single-Domain Ferrimagnetic Grain

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Abstract—The basic metastable state of magnetic grains are analyzed in the framework of the developed model. It was shown that the grain may be in one of the three magnetic states, depending on its sizes and elongation: uniform magnetization, small (quasi-single-domain) and great (two-domain) nonuniform magnetic moment. The size of single domain a_0, the limiting size of a uniformly magnetized grain a_{0m} and the minimum a_1 and maximum a_{1m} sizes of the stable quasi-single-domain state were calculated. An important distinction of the results obtained is a nonmonotonic behavior of the critical sizes a_0, a_{0m}, a_1, a_{1m} with the increasing elongation of the grain.

The requirements for the reliability of paleomagnetic data constrain the choice of objects studied. The rocks containing magnetic minerals in the form of small, almost single-domain, particles are believed to be most acceptable. The remanent magnetization of such rocks is stable with respect to chemical, thermal, and other actions. However, the presence of the metastable magnetic states of the particles involved may substantially affect the preservation of the remanence, which plays an important role in the diagnostics of its type.

The magnetic properties of such particles were studied in many (largely theoretical) papers [1–10], which were based on one or another assumption on the internal magnetic structure. The general feature of all the proposed models is the use of a variational search for the total energy minimum. Depending on the accepted model, the variational parameters were the dimensions of the domains and their walls, the rotation angle of the spontaneous magnetization vector $\boldsymbol{I}_s$ in various grain areas, and other quantities. The development of computer facilities provided the changeover from a simple model [11], which represented domains and their walls by uniformly magnetized parallelepipeds, to models with a three-dimensional distribution of $\boldsymbol{I}_s$ [8, 12].

However, none of the studies could purport to fully account for the magnetism of these particles, since their objectives were to solve particular problems. The studies of the effects of an external field or shape anisotropy on the magnetization distribution in a grain were limited by the simplifying assumptions used in the quoted papers (specifically, the assumptions on the antiparallel orientation of the magnetic moments of domains [11] or on the magnetite uniaxiality [16]). Because of this, we make an effort to analyze the magnetic states in the framework of the model presented below, which is an extension of that previously proposed [16].

MODEL OF A TWO-DOMAIN PARTICLE

We consider a grain whose crystalline structure is represented by a mineral of cubic symmetry. The grain is of the shape of a rectangular parallelepiped with a base area of a^2 and height qa (the edges of the cube coincide with the crystallographic axes). We restrict ourselves to the case when the magnetic moment is distributed in plane $y0z$ (Fig. 1) by the law

$$\theta(x) = \begin{cases} \theta_1, & 0 \le x \le p_1 \\ \theta_1 + \dfrac{(\theta_2 - \theta_1)}{p_2}(x - p_1), & p_1 \le x \le p_1 + p_2 \\ \theta_2, & p_1 + p_2 \le x \le 1, \end{cases} \quad (1)$$

where θ_1 and θ_2 are the angles between the magnetic moment and axis $0z$ in the first and second domains, respectively, p_1 and p_2 are the widths of the domain and its boundary, respectively, and variables x, y, and z are normalized to a.

We study the equilibrium states of the magnetic moment of the grain by minimizing total energy E with respect to four parameters: θ_1, θ_2, p_1, and p_2. (In the subsequent discussion, we use the reduced energy defined as $\varepsilon = \dfrac{E}{qa^3 I_s^2}$.)

The total energy of the magnetic moment is composed of the **exchange energy**

$$\varepsilon_{ex} = A\frac{(\theta_2 - \theta_1)^2}{p_2 a^2 I_s^2}, \quad (2)$$

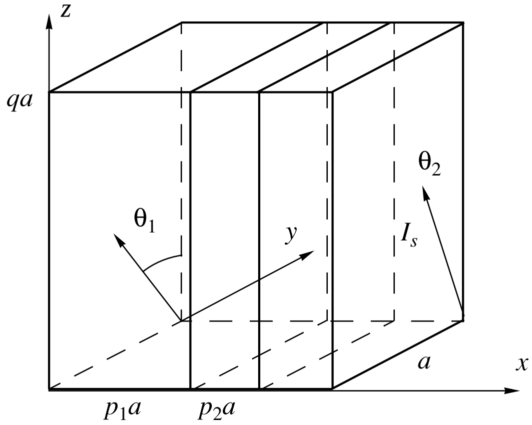

Fig. 1. Illustration of the model considered.

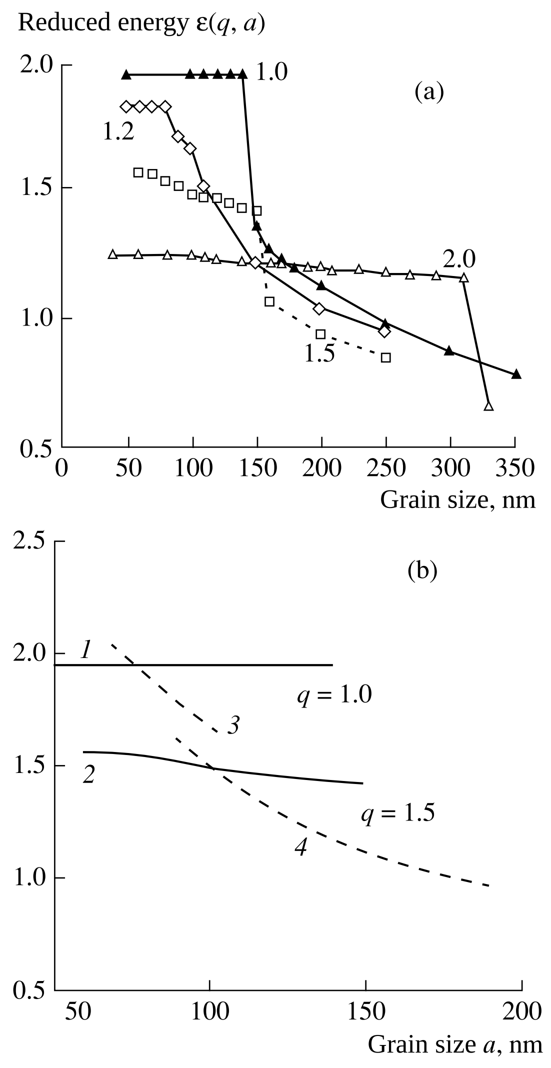

Fig. 2. Energy ε of a grain versus its size a and elongation q for various initial states. (a) The uniform distribution of the magnetic moment, (b) the uniform (curves *1* and *2*) and nonuniform (curves *3* and *4*) distributions of the magnetic moment.

the **crystallographic anisotropy energy**

$$\varepsilon_{\rm an} = \frac{K}{8I_s^2}\left\{1 - p_1\cos 4\theta_1 - (1 - p_1 - p_2)\cos 4\theta_2 - \frac{p_2(\sin 4\theta_2 - \sin 4\theta_1)}{4(\theta_2 - \theta_1)}\right\}, \quad (3)$$

the **magnetostatic energy**

$$\varepsilon_m = \frac{1}{2qa^3I_s^2}\int_S\int_S\frac{(\mathbf{I}_s(\mathbf{r}), ds)(\mathbf{I}_s(\mathbf{r}'), ds')}{|\mathbf{r} - \mathbf{r}'|} = \frac{1}{q}\int_0^1 dx\int_0^1 dx'\{f_1(x - x')\cos\theta(x)\cos\theta(x') + f_2(x - x')\sin\theta(x)\sin\theta(x')\} \quad (4)$$

and the **energy of the magnetic moment in external field H**

$$\varepsilon_H = -\frac{1}{qa^3I_s^2}\int_V(\mathbf{H}, \mathbf{I}_s)dV = -\frac{Hm(\theta_1, \theta_2, p_1, p_2, \varphi)}{I_s}. \quad (5)$$

Here, the following notation is used: A is the exchange constant, $\mathbf{I}_s$ is the spontaneous magnetization vector whose modulus is uniform over grain volume V, K is the crystallographic anisotropy constant, and φ is the angle between vector **H** and axis 0z. Furthermore,

$$f_1(\tau) = q\ln\left\{\frac{(\sqrt{q^2 + \tau^2} + q)\sqrt{1 + \tau^2}}{(\sqrt{1 + q^2 + \tau^2} + q)|\tau|}\right\} - \sqrt{q^2 + \tau^2} + \sqrt{1 + q^2 + \tau^2} - \sqrt{1 + \tau^2} + |\tau|,$$
$$f_2(\tau) = \ln\left\{\frac{(\sqrt{1 + \tau^2} + 1)\sqrt{q^2 + \tau^2}}{(\sqrt{1 + q^2 + \tau^2} + 1)|\tau|}\right\} - \sqrt{1 + \tau^2} + \sqrt{1 + q^2 + \tau^2} - \sqrt{q^2 + \tau^2} + |\tau|, \quad (6)$$

and

$$m(\theta_1, \theta_2, p_1, p_2, \varphi) = \left\{p_1\cos(\theta_1 - \varphi) + (1 - p_1 - p_2)\cos(\theta_2 - \varphi) + \frac{p_2[\sin(\theta_2 - \varphi) - \sin(\theta_1 - \varphi)]}{\theta_2 - \theta_1}\right\}, \quad (7)$$

where $m(\theta_1, \theta_2, p_1, p_2, \varphi)$ is the projection of the magnetic moment of the grain onto the direction of vector **H** and is reduced to qa^3I_s.

PRINCIPAL AND METASTABLE STATES

The **H** = 0 modeling of the magnetic moment distribution in magnetite grains (I_s = 485 A/m, A = 0.67×10^{11} J/m, $|K| = 1.36 \times 10^4$ J/m^3) of various sizes and elongations (see Figs. 2–4) identifies three types of magnetic states: a uniform magnetization state, a (pseudo-single-domain) state with a small nonuniformity of the magnetic moment, and a (two-domain) state with a greatly nonuniform magnetic moment.

Uniform Magnetization States

The dependence of the energy and magnetic moment on the size and elongation of the grain for each of its states is illustrated in Figs. 2 and 3. It follows that the state with uniform vector $\mathbf{I}_s$ is the principal one only in a size interval limited by single-domain size a_0, which changes nonmonotonically with the elongation of the grain: it decreases from 70 nm for isometric particles to 50 nm for particles with an elongation of q = 1.3 and reaches 150 nm for q = 3.0 (Fig. 4).

For $a > a_0$, the two-domain or pseudo-single-domain state is energetically more profitable than the uniform state (Fig. 2b). The latter, if it exists, remains stable up to maximum size a_{0m}. The dependence of a_{0m} on the grain elongation is presented in Fig. 4. We see that, as the elongation increases from q = 1 to q = 1.3 a_{0m} is halved, whereupon it increases, so that for q > 3.1, the uniform distribution of the magnetic moment can be presented in particles of any size (Figs. 3 and 4).

Nonuniform Magnetization States

The states with a weakly nonuniform distribution of $\mathbf{I}_s$ (Figs. 2 and 3) are equilibrium ones only at a range $a_0 < a < a_1$ (Fig. 4), but for $a_1 < a < a_{1m}$, they are metastable (here, a_1 is the pseudo-single-domain size and a_{1m} is the maximum pseudo-single-domain size). Note that the pseudo-single-domain state with q > 2.1 can exist in particles greater than a_0.

Furthermore, as the grain size increases, the zone of the nonuniform magnetic moment (the width of the domain boundary zone) remains unchanged, occupying approximately 2/3 of the grain volume.

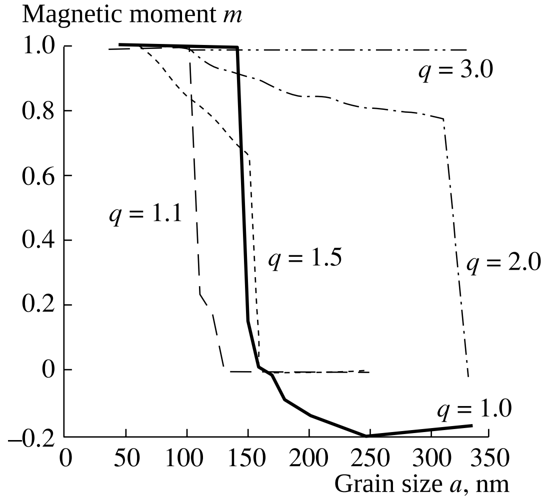

Fig. 3. Magnetic moment of a grain versus its size a and elongation q.

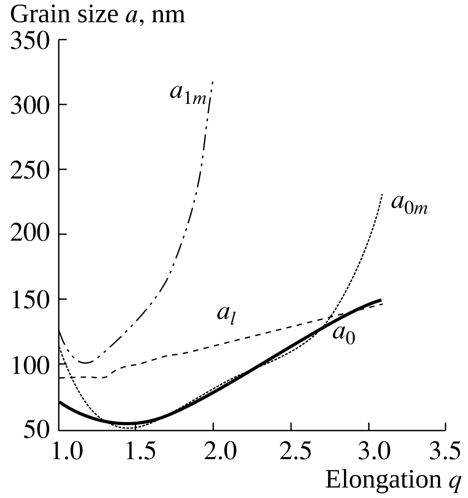

Fig. 4. The grain elongation q dependence of (*1*) single-domain grain size a_0, (*2*) maximum size a_{0m} for the uniform magnetization, (*3*) pseudo-single-domain size a_1, and (*4*) maximum pseudo-single-domain size a_{1m}.

ANALYSIS OF PRINCIPAL RESULTS

Single-Domain Dimension

The single-domain state dimensions calculated in our work (Fig. 4) are generally consistent with those given by Dunlop [13]. For example, the experimentally found size of a single-domain cubic particle is 50 [14] or 80 nm [15], and the theoretical estimates of a_0 are 80 [4, 16], 100 [8], 116 [17], and 120 nm [18]. Following [4, 19], an increase in grain elongation at the range $1.0 < q < 2.5$ results in greater a_0 in the size interval of 80 to 120 nm.

An important feature of our results is the nonmonotonic change in the single-domain sizes with increasing grain elongation (Fig. 4). Such a dependence $a_0 = a_0(q)$ is due to the nonmonotonic behavior of the effective anisotropy constant that results from the tensor summation of the crystallographic and shape anisotropy constants [20, 21].

The competition among various anisotropy types also affects the position of the "light" magnetization axis [20, 21], which can help to explain the dependence of the critical dimension on the grain elongation (Fig. 5). It is noteworthy that the orientation of the light axis in an isometric particle along the diagonal of its face (but not of the cube) relates to the constraints of the

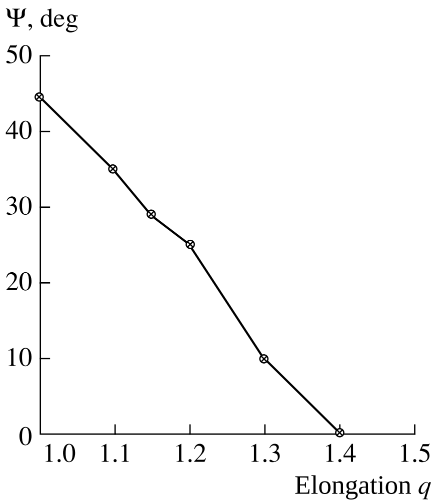

Fig. 5. The grain elongation q dependence of orientation angle Ψ of the light magnetization axis relative to the elongated grain edge.

model used (the magnetic moment can rotate only in plane $z0y$).

Metastable Single-Domain State

Maximum size a_{0m} of a grain with the uniform distribution of $\mathbf{I}_s$ qualitatively repeats the course of the effective anisotropy constant, in view of the nonmonotonous behavior of this constant. While the values of a_{0m} obtained for little elongated particles are close to those presented in the papers [2, 4, 16, 17], the dependence $a_{0m} = a_{0m}(q)$ found in [16] for $q > 1.2$ (when the shape anisotropy energy of the magnetite grain becomes comparable to the crystallographic anisotropy energy) is substantially different from the energy calculated by us. The elongation, for which maximum size a_{0m} is indefinitely large, falls into a range of somewhat greater q (ranging from 2.4 to 3.1, according to [16]), which significantly lowers a_{0m} for $1.2 < q < 3.0$.

Weakly Nonuniform State

Without going into terminological details, the state with a weakly nonuniform distribution of the magnetic moment can be identified either with the pseudo-single-domain state (a “twisting” mode) [22–24] or with a state characterized by the curling mode [16, 25]. Unlike the states described above [see also 16, 22, 25], this state can be realized along with the uniform or two-domain states and remains stable in a narrow interval of sizes $a_0 < a < a_1$. In this case, the values presented in Fig. 4 are twice as much as the critical size of the curling mode $a_c = 40$ nm [25].

Two-Domain State

The modeled two-domain magnetization distribution is not classical (two oppositely magnetized blocks are separated by a 180° domain wall). As the grain size increases, the observed angles θ_1 and θ_2, which determine the magnetic moment orientation in the first and second domains, deviate from 0° and 180°, respectively. Ultimately, this gives rise to the appearance of a small magnetic moment of the isometric particle for $a >$ 140 nm (see Fig. 3). A similar magnetization distribution, called “skirts” in [17], was studied on larger particles. For example, the skirts occurred in isometric ($q =$ 1.0) particles with $a = 180$ nm and in elongated ($q =$ 1.5) particles with $a = 450$ nm.

Calculations show that the two-domain state is metastable “from below,” in a range from $a \approx 90$ nm to $a = a_1$. For $a > a_1$, the two-domain state is energetically more profitable and exists along with the uniform (for $a < a_{0m}$) or pseudo-single-domain (for $a < a_{1m}$) magnetizations, becoming the only state for $a > a_{1m}$. The values of a_{1m} lie somewhat below those given in [4, 16] and, on the whole, are not in conflict with them.

The developed model and results from our analysis of the magnetic states can be used to model the magnetization of ensembles of particles whose sizes are comparable to the single-domain dimension, which is, in essence, the object of further studies.

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