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CHAIN MODEL OF GENERATION OF HEAT IN A SOLID DISPERSION OF SUBDOMAIN FERROPARTICLES ON EXPOSURE TO A VARIABLE FIELD

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The release of heat in a system of small ferromagnetic particles with magnetic hysteresis on exposure to a variable linearly polarized field has been studied. A chain model of the noncoherent process of magnetization reversal has been proposed; compared to the theory of absolute single-domainality, the model made it possible to bring computational results closer to experimental data.

Introduction. Systematic study of the processes of absorption of the energy of a variable magnetic field in dispersions of small ferromagnetic particles, which are magnetization-reversible following the hysteresis type, has been started in [1–3]. Such systems as heat sources are of interest in that they enable one to form three-dimensional heaters of adaptable shape and size, up to the cell size, which seems topical in the context of the trend toward miniaturization and of the development of nanosize technologies. Our direct interest expressed in this problem is linked to the study of the prospects for using hysteresis in small ferroparticles for local hyperthermia of malignant tumors [4]. Earlier [1, 2], the model of absolutely single-domain ellipsoids magnetization-reversed by coherent magnetization rotation was used for theoretical analysis of the dissipation of energy in ferrodispersions. A comparison of the results of such a calculation with experimental data [1, 3] obtained for needle-shaped submicron γ-Fe_2O_3 particles has revealed significant differences. They can partly be determined by the spread in particle shape and size and partly has a more fundamental nature related to the noncoherent character of magnetization reversal. Also, the disturbance of the single-domain state of microparticles in magnetization reversal is found in experiments on studying their magnetic properties (see [5]) and is investigated by micromagnetism methods [6]. However, the micromagnetic relations obtained for an individual particle are complex, and the model of absolute single-domainality dominates the literature on the theory of collective properties of ensembles of small ferroparticles. At the same time, as early as the mid-20th century it was proposed that magnetic microparticles be described by spherical chains in connection with the large differences in the values of the measured coercive force and that predicted by single-domain theory [7]. It was proposed that each sphere in the chain is magnetization-reversed in a coherent manner, and there is a nonrigid orientation link of a dipole nature between the orientations of spherical magnetic moments (this link allows the noncoherent process of magnetization reversal of the entire chain). In the present work, a variant of the chain model has been used for studying the orientation behavior and energy dissipation in a solid dispersion of ferroparticles with magnetic hysteresis. It has turned out that allowance for the noncoherent character of magnetization reversal within the framework of the chain model makes it possible to bring theoretical predictions sharply closer to measurement results.

Chain Model of Noncoherent Magnetization Reversal. We consider a linear chain consisting of three identical spherical particles which possess dipole magnetic moments $\mathbf{m}_k = m\mathbf{e}_k$ and a uniaxial crystalline magnetic anisotropy characterized by the energy density K. We assume that the directions of the easiest-magnetization axes of particles are coincident with the direction of orientation of the chain $\mathbf{n}$, and the magnetic field is applied along the direction $\mathbf{h}$: $\mathbf{H}(t) = H_0 \sin(\omega t)\mathbf{h}$. By virtue of the symmetric arrangement of the last particles, their dipoles have the same orientation $\mathbf{e}_2$ which can differ from the orientation $\mathbf{e}_1$ of the dipole of the central particle. The total magnetic energy of the chain involves the energy of dipoles in the external field, the energy of dipole interactions, and the overall energy of crystalline anisotropy:

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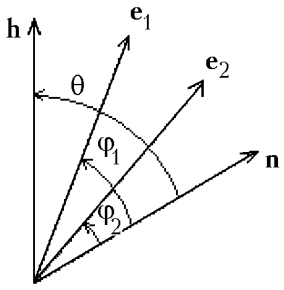

Fig. 1. Geometry of the problem.

$$U = -mH\,(\mathbf{e}_1 + 2\mathbf{e}_2)\,\mathbf{h} - KV_1\left[(\mathbf{e}_1\mathbf{n})^2 + 2\,(\mathbf{e}_2\mathbf{n})^2\right] + \frac{m^2}{d^3}\left[2\mathbf{e}_1\mathbf{e}_2 - 6\,(\mathbf{e}_1\mathbf{n})\,(\mathbf{e}_2\mathbf{n}) + \frac{1}{8}\,(1 - 3\,(\mathbf{e}_2\mathbf{n})^2)\right]. \quad (1)$$

In the system in question, we separate a plane formed by the directions of the field and the chain axis. We assume that the directions of dipole moments of particles remain in this plane in all cases. We stress the differences of the model proposed from the chain model [7]. First, in [7] (see also [8]), consideration has been confined to isotropic particles ($K = 0$); second, for the noncoherent process of magnetization reversal, a fan-shaped mode has been postulated: all dipoles in the chain make the same angle with the field, whereas any two neighboring dipoles are deflected from the plane of symmetry through the same angle in the opposite directions. We determine the process in question as a noncoherent plane magnetization-reversal mode. The system's state can be described in this case by an angle θ reckoned counterclockwise from the positive (arbitrarily) direction of the chain to the positive (arbitrarily) direction of the field and determined analogously by the angles φ_1 and φ_2 between the positive direction of the chain and the direction of the magnetic dipoles of the central and last particles respectively (Fig. 1). Introducing the energy scale $U_* = 2m^2/d^3$ and the notation $u = U/U_*$, $mH/U_* = p = 4H/(\pi I)$, and $KV_1/U_* = q = 4K/(\pi I^2)$, where I is the magnetization of particles, we represent the expression for energy (1) in the form

$$u = -p\cos(\theta - \varphi_1) - 2p\cos(\theta - \varphi_2) - q\,[\cos^2\varphi_1 + 2\cos^2\varphi_2]$$

$$+ \cos(\varphi_2 - \varphi_1) - 3\cos\varphi_1\cos\varphi_2 - \frac{3}{16}\cos^2\varphi_2\,. \quad (2)$$

The equilibrium positions of the magnetic moments are determined by the equations $\partial u/\partial\varphi_1 = \partial u/\partial\varphi_2 = 0$. In expanded form, they are

$$q\sin 2\varphi_1 - p\sin(\theta - \varphi_1) + \sin(\varphi_2 - \varphi_1) + 3\cos\varphi_2\sin\varphi_1 = 0\,,$$

$$\left(2q + \frac{3}{16}\right)\sin 2\varphi_2 - 2p\sin(\theta - \varphi_2) - \sin(\varphi_2 - \varphi_1) + 3\cos\varphi_1\sin\varphi_2 = 0\,. \quad (3)$$

The stability (minimum of energy) or instability (its maximum) of the stationary point is determined by the value of the second energy differential in it

$$d^2u = \frac{\partial^2 u}{\partial\varphi_1^2}(\Delta\varphi_1)^2 + 2\,\frac{\partial^2 u}{\partial\varphi_1\partial\varphi_2}\Delta\varphi_1\Delta\varphi_2 + \frac{\partial^2 u}{\partial\varphi_2^2}(\Delta\varphi_2)^2\,. \quad (4)$$

The quadratic form (4) is positive definite and the stationary point is the minimum point in the case where the necessarily real eigenvalues λ_i of the matrix of coefficients $u_{ij} = \partial^2 u/\partial\varphi_i\partial\varphi_j$ are all positive. The eigenvalues of the matrix of coefficients are the roots of the characteristic equation $\det\|u_{ij} - \lambda\delta_{ij}\| = 0$:

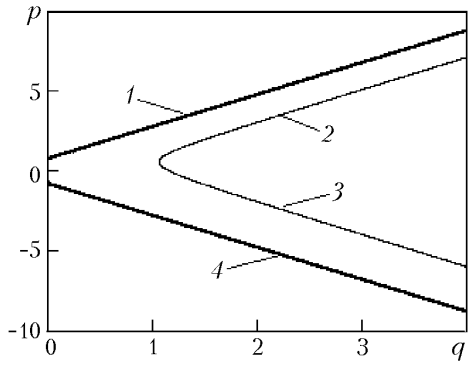

Fig. 2. Diagram of the regimes of magnetization reversal of the chain by a parallel field on the plane of dimensionless variables q–p: 1) switching of the orientations (φ_1, φ_2) of the dipoles of the central and last particles (π, π) $\rightarrow$ (0, 0); 2) (0, π) $\rightarrow$ (0, 0); 3) (0, π) $\rightarrow$ (π, π); 4) (0, 0) $\rightarrow$ (π, π).

$$\lambda_{1,2} = \frac{u_{11} + u_{22}}{2} \pm \sqrt{\frac{(u_{11} + u_{22})^2}{4} - u_{11}u_{22} + u_{12}^2} \ .$$

Clearly, the stability condition is reduced to the condition of positiveness of the smaller root of those two ($\lambda_2 > 0$), and the boundary of the stability region on the plane of variables p and q is determined by the equation $\lambda_2 = 0$, i.e.,

$$u_{11}u_{22} = u_{12}^2 \ . \tag{5}$$

We consider the process of magnetization reversal of the chain by a field directed along the chain axis. When the value of the field strength p is arbitrary and $\theta = 0$, the equilibrim equations (3) are satisfied by any pair of values (φ_1, φ_2) = (0, 0), (0, π), (π, 0), and (π, π). With allowance for the physical indistinguishability of both directions of the chain axis, the last two solutions replicate the first ones upon change in sign of p. If, in the initial state ($p = 0$), the dipoles of all the three particles have stable orientation in the positive direction of the chain axis ($\varphi_1 = \varphi_2 = 0$), magnetization reversal occurs upon the application of the counterfield ($p < 0$) at the instant of reaching condition (5) which leads to the quadratic equation

$$(p + 2q + 2)\left(p + 2q + \frac{19}{16}\right) = \frac{1}{2} \ . \tag{6}$$

The critical strength of the magnetization-reversal field is related to the smallest (in absolute value) root of this equation:

$$p_* = -0.778 - 2q \ . \tag{7}$$

One more stationary point ($\varphi_1 = 0$ and $\varphi_2 = \pi$) corresponds to a state in which the last particles are magnetized toward the central point. It can be stated a priori that this state is stable in a fairly weak field and for a fairly strong crystalline anisotropy. Let p be equal to 0; then Eq. (5) takes the form

$$q^2 - \frac{45}{32}q + \frac{9}{32} = 0 \ , \quad q_{1,2} \approx 0.242 \ , 1.167 \ .$$

The stability condition is $q > 1.167$ ($K > 0.917I^2$). In a nonzero field, Eq. (5) has the following roots:

$$p_{1,2} = \frac{19}{32} \pm \sqrt{4q^2 - \frac{45}{8}q + \frac{1513}{1024}} \ .$$

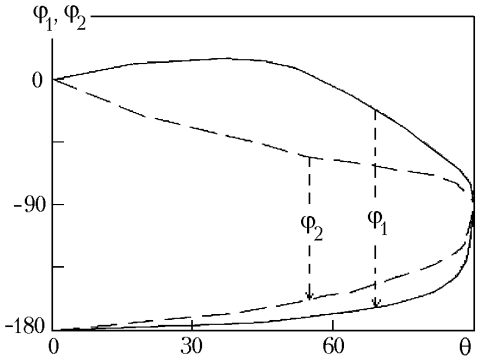

Fig. 3. Change in the orientations of the dipoles of the central (φ_1) and last (φ_2) particles in the chain (the particles are isotropic) at the instant of magnetization reversal as a function of the angle θ of orientation of the chain relative to the field. θ, deg.

The values of p hold for $q < 0.125$ and $q > 1.281$. The second of these conditions has a physical meaning. The critical values of the field of magnetization reversal of the chain in the positive (switching of the last dipoles) and negative (switching of the central dipole) directions are different. A diagram of the modes of magnetization reversal of the chain by a magnetic field that is parallel to its axis is presented in Fig. 2. As is seen, the modes of partial magnetization reversal are unattainable in practice, since the corresponding region of critical fields lies within the region of magnetization-reversal fields of the entire chain.

To illustrate the noncoherent character of magnetization reversal of the system in question, Fig. 3 shows the angles of orientation of the central and last dipoles relative to the axis of the chain of isotropic particles ($q = 0$) just before and after its magnetization reversal as functions of the angle between the chain and the magnetizing field. The critical field of magnetization reversal and the state of the system before and after the jump have been determined by numerical minimization of the energy (2) by the variables φ_1 and φ_2 for the prescribed value of θ and a growing value of p. The positive value of the angle φ_1 or φ_2 means that the corresponding dipole is to the left of the positive direction of the chain axis; the negative value is on the right. It is noteworthy that the last dipoles are deflected from the chain axis toward the field, whereas the central dipole, up to $\theta \approx 56$, is deflected in the opposite direction. The maximum difference between the dipole orientations at the instant of jump occurs for $\theta \approx 54$ and is about 60^o.

Coercive Force and the Absorption of Energy. When the chain is magnetization-reversed in the direction of its axis, the coercive force is determined by relation (7). Upon passage to dimensional quantities, it takes the form

$$H_c = 0.66I + \frac{2K}{I}. \tag{8}$$

In particular, for isotropic iron particles (I = 1700 G) relation (8) yields H_c = 1122 Oe compared to 1420 Oe in the case of a fan-shaped mode [7].

The energy absorbed by the chain in one act of magnetization reversal is equal to the difference of energies in the initial and final states. For the longitudinal field, we have

$$\Delta u_1 = u\,(p_*\,(q), q, \pi, \pi) - u\,(p_*\,(q), q, 0, 0)\,, \quad p_* = 0.778 + 2q\,.$$

The numerical results obtained from this relation are approximated, with an error of less than 1%, by the dependence

$$\Delta u_1 = 4.67 + 12q \quad (\Delta U_1 = 1.22I^2V + 4KV)\,, \tag{9}$$

where $V = 3V_1$ is the chain volume.

We consider the characteristics of magnetization reversal of the chain for an arbitrary angle of its orientation relative to the field. The parameters of the process are the coercive force p_c (field strength for which the projection of

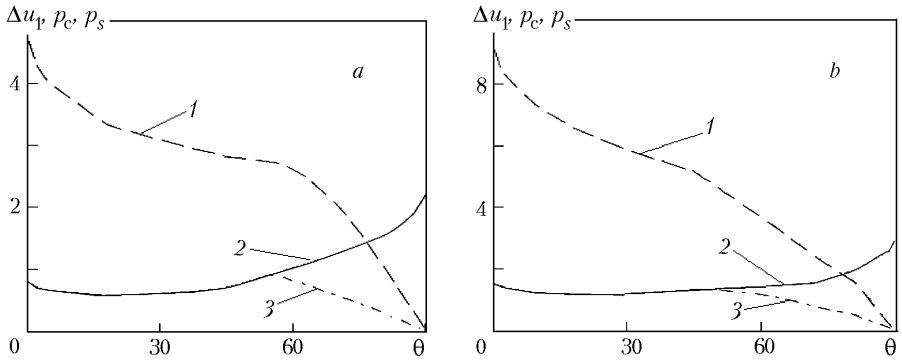

Fig. 4. Dimensionless absorbed energy Δu_1 (1), switching-field strength p_s (2), and coercive force p_c (3) of the chain vs. the angle θ of its orientation relative to the field: a) isotropic particles and b) anisotropic particles (q = 0.37). θ, deg.

the total magnetic moment of the chain onto the field direction vanishes), the switching-field strength p_s for which the irreversible transfer of the chain's dipole moments occurs, and the field energy absorbed on magnetic switching Δu_1. We carry out computations on the basis of the procedure of numerical minimization of the chain energy (2). The results for the case of isotropic particles are presented in Fig. 4a, and those for anisotropic particles with the parameters of a γ-Fe_2O_3 dispersion [1] (I = 400 G, $K = 4.7 \cdot 10^4$ erg/cm^3, and q = 0.37) are given in Fig. 4b. We note the characteristic difference of the resulting dependence of the coercive force on the angle of orientation of the chain from the analogous dependence for the model of fan-shaped magnetization reversal [7]. The small coercive-force maximum observed in our experiments in fieldwise orientation of the chain is absent from the model of [7]. The dependences $\Delta u_1(\theta)$ and $p_s(\theta)$ found numerically make it possible to compute the intensity of absorption of the variable-field energy in a disordered ensemble of chains. The average energy per chain, absorbed in one act of magnetization reversal, is determined by the relation

$$\langle \Delta u_1 (p) \rangle = \int_0^{\pi/2} f(\theta)\, \Delta u_1 (\theta)\, F(p, \theta)\, d\theta , \tag{10}$$

where $f(\theta) = \sin(\theta)$ is the normalized distribution function of the chain axes on the hemisphere $0 \le \theta < \pi/2$, and

$$F(p, \theta) = \begin{cases} 0 , & p < p_s (\theta) ; \\ 1 , & p \ge p_s (\theta) . \end{cases}$$

The power of energy dissipation in the disordered ensemble of chains with a volume concentration of ferromagnetic c on exposure to the field of frequency $f = \omega/2\pi$ is determined by the relation

$$W = \frac{2\pi}{9} cfI^2 \langle \Delta u_1 \rangle . \tag{11}$$

We compare the results for the chain model and results (obtained earlier [1]) of calculation from the model of ellipsoids magnetization-reversible in a coherent manner to the results of measurement [1] of the energy-absorption power in a 10% dispersion of needle-shaped γ-Fe_2O_3 particles in beeswax in a field of frequency f = 50 Hz. In Fig. 5, the heat-release power W measured in [1] as a function of the field amplitude H_0 is referred to the field frequency f and to the concentration of particles c. The quantity $\phi = W/(cf)$, called the characteristic specific thermal productivity (efficiency), of a ferrodispersion determines the energy absorbed in one cycle of variation in the field in a unit volume of ferroparticles. Curves 1 and 2 show results of calculation from formula (10) for I = 400 G at q = 0 and q = 0.37 ($K = 4.7 \cdot 10^4$ erg/cm^3). In the first case the crystalline anisotropy of particles is ignored; in the second case the crystalline-anisotropy constant is taken to be equal to its value for γ-Fe_2O_3. For absolutely single-domain ellipsoids of revolution, the absorption of energy is determined by the magnetization of a particle and by the effective-anisotropy constant. The

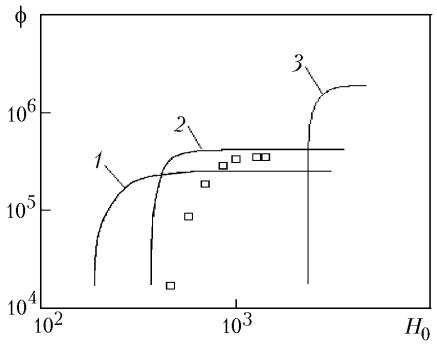

Fig. 5. Comparison of the dependence (measured in [1]) of the characteristic specific thermal productivity ϕ of a disordered system of needle-shaped γ-Fe_2O_3 particles (points) on the amplitude of the variable field H_0 to the results of computations from the chain model [1) isotropic particles and 2) anisotropic particles] and from the model of absolutely single-domain ellipsoids of revolution (3). ϕ, erg/cm^3; H_0, Oe.

latter, along with the crystalline-anisotropy constant, involves the form-anisotropy constant $K_f = 4\pi(N_b - N_a)I^2$, where N_a and N_b are the demagnetization factors along the ellipsoid axes (see [5]). The typical value of the ratio of the semiaxes of γ-Fe_2O_3 particles used for magnetic recording is 5:1. In this case we have $N_b - N_a \approx 0.44$ and $K_f = 8.8\cdot10^5$ erg/cm^3, and the effective-anisotropy constant is $K_{eff} = K + K_f \approx 9.3\cdot10^5$ erg/cm^3. The thermal productivity of the system of ellipsoids of revolution magnetization-reversible in a coherent manner with the indicated parameters, which has been calculated with the results of [1], is presented in Fig. 5 by curve 3. As we see, the values (predicted within the framework of this model) of the limiting ($H_0 \to \infty$) thermal productivity of the system and the amplitude of the field of "switching" of hysteresis differ by an order of magnitude from the values observed. Allowance for the noncoherent character of magnetization reversal within the framework of the chain model proposed substantially diminishes the disagreement between theory and experiment.

Conclusions. Thus, the chain approach proposing, undoubtedly, a very rough model of magnetic phenomena in small particles ensures an acceptable description of the quantities observed and can be a useful means for studying processes excited by an electromagnetic field in complex systems with small particles. In hyperthermia problems, the chain model creates prerequisites for a more realistic (compared to the model of absolute single-domainality) description of the regularities of energy dissipation in magnetic and mechanical codynamics of particles in soft matrices.

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NOTATION

c, volume concentration; d, particle diameter, cm; $\mathbf{e}_k$, unit vector in the direction of the magnetic moment of the kth particle; f, frequency, Hz; H, field strength, Oe; H_c, coercive force, Oe; **h**, unit vector in the direction of polarization of the field; I, (intensity of) magnetization, G; K, density of the energy (constant) of magnetic anisotropy, erg/cm^3; $m = IV_1$, magnetic moment of a particle, G·cm^3; **n**, unit vector in the direction of the axis of a particle chain; N_a and N_b, demagnetizing factors of an ellipsoid; p, dimensionless magnetic-field strength; p_s, dimensionless switching-field strength; $p_c = IH_c/K$, dimensionless coercive force; q, dimensionless anisotropy energy; t, time; U, energy, erg; U_*, energy scale, erg; $u = U/U_*$, dimensionless energy; ΔU_1, energy absorbed by the chain of particles in one act of magnetization reversal, erg; Δu_1, dimensionless energy absorbed in one act of magnetization reversal; V_1, particle volume, cm^3; $V = 3V_1$, volume of the chain of particles, cm^3; W, energy-dissipation power, erg/cm^3; ω, cyclic frequency, sec^{-1}; $\phi = W/(cf)$, specific thermal production of a ferrodispersion, erg·sec; θ, angle of orientation of the chain; φ, angle of orientation of the magnetic moment. Subscripts: 1, unit (one particle, one magnetization-reversal cycle), the central particle in the chain; 2, last particles in the chain; a and b, semiaxes of the ellipsoid of revolution; eff, effective; c, coercive; f, form, shape.

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