
**MAGNETISM
AND FERROELECTRICITY**

Stepwise Magnetization of Dispersed Ferromagnets due to Magnetic Interparticle Interactions

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Abstract—The main features of stepwise magnetization of dispersed ferromagnets caused by magnetic interparticle interactions are studied using a two-particle model. The ranges of values of the magnetic anisotropy constants of particles and of the dipole–dipole interaction between them are determined over which a reproducible jumpwise change in the magnetization of the system occurs in an external positive magnetic field. The proposed model is shown to explain the main specific features of the fine structure of the ferromagnetic resonance spectra.

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1. INTRODUCTION

Increasing interest in nanosized particles is due to their special features different from those of the bulk material. When studying real nanosized and nanostructured systems, one often deals not with an isolated nanoparticle but rather with an ensemble of interacting particles, which poses additional problems in interpreting the experimental data. In this connection, the most important problem is to study the collective behavior of magnetic particles in various disperse systems. Magnetic interparticle interactions play an important role and even can sometimes determine the behavior and properties of disperse magnetic structures in comparison with noninteracting, isolated, or nonmagnetic nanoparticles. The most studied effects are the changes in the coercive field [1, 2] and in the ferromagnetic resonance (FMR) spectra [3–6]; the “phase transition” from the disordered to an ordered magnetic state in a system of interacting small superparamagnetic particles [7]; and the magnetic phase decomposition of lanthanum manganites and related materials [8], upon which the material remains in the homogeneous chemical and structural state, but it is magnetically inhomogeneous. Interparticle interactions not only influence the fundamental properties of structures on an ultrasmall scale [1, 9–11], but they also expand the possibilities of producing nanostructural systems with preset characteristics, since they can cause self-assembling of magnetic nanoparticles [12, 13] into low-dimensional regular structures under certain synthesis conditions. The temperature of critical phenomena in this case is largely determined by the energy of magnetic dipole interaction.

One of the most important characteristics of dispersed magnets is the dependence of the magnetization of the system on external magnetic field. The process of magnetization of an isolated single-domain ferromagnetic particle with uniaxial magnetic anisotropy was studied long ago [14, 15]. However, the problem regarding the change in the magnetic moment of dipole-interacting magnetic particles has been solved only for certain particular cases and, as a rule, under the assumption that the ratio of the dipole–dipole interaction constant to the magnetocrystalline anisotropy constant is small [2, 16].

The effect of the magnetic interparticle interactions on the integrated characteristics of the process of magnetization of a disperse system has been studied theoretically in many papers [1, 3, 17, 18]. It has been shown, in particular, that the magnetic dipole–dipole interaction decreases the energy barrier to magnetization reversal and decreases the coercive force of the system in comparison with the case of noninteracting particles [1, 16] and that the magnetic relaxation rate decreases as the magnetic interparticle interaction increases [18].

In [4], using a simple model of two interacting identical magnetic dipoles, it was shown for the first time that, for certain parameter values, a qualitatively different process of particle magnetization reversal can occur. Specifically, the dipole–dipole interaction can cause the appearance of jumps in the magnetization and the corresponding hysteresis with variation in the magnetic field strength without reversing the field direction. A necessary condition for this effect to occur is that the different contributions to the total energy of the system

(Zeeman energy, magnetocrystalline anisotropy energy, dipole–dipole interaction energy) be of the same order of magnitude. Using the FMR method, it has been found experimentally that such processes in magnet-based disperse systems manifest themselves as additional narrow weak lines [4, 5]. These processes were studied further in the lanthanum manganites exhibiting colossal magnetoresistance [6]. It was shown that, by analyzing the additional narrow lines in the FMR spectra, one can obtain information on the spatial magnetic phase inhomogeneity of magnets exhibiting colossal magnetoresistance.

It is well known that hysteretic magnetization of a magnetic particle in positive magnetic fields can also occur in the absence of interparticle interaction for certain particle geometry and a certain strength of magnetocrystalline anisotropy of the particle. For example, magnetocrystalline anisotropy of a particle can cause hysteresis in positive fields in the case where an isolated particle has a cubic symmetry and an external magnetic field is applied along the (1,1,1) direction [19]. Numerical calculations showed that the probability of this phenomenon occurring in randomly oriented particles in a disperse sample is extremely low [4]. Another example of hysteresis in positive fields is the Barkhausen effect discovered in 1919. In this case, jumpwise changes in magnetization are due to retardation of domain wall motion by structural defects in ferromagnetic particles. The Barkhausen effect is observed in multidomain particles in low magnetic fields; under these conditions, magnetization occurs through domain wall motion. It should be stressed that the distinctive feature of the Barkhausen effect is irreproducibility of jumps in magnetization with varying external magnetic field. Steplike changes in magnetization in positive fields are also observed in single crystals with a chainlike structure, such as $\text{Ca}_3\text{Co}_2\text{O}_6$, in which the intrachain and interchain interactions are ferromagnetic and antiferromagnetic in character, respectively [20]. In this case, however, jumps in magnetization can occur only at low temperatures (≤ 10 K) and are associated with quantum mechanical tunneling, which is an additional relaxation channel during magnetization of the system.

In this paper, we numerically analyze the behavior of a disperse system consisting of magnetic particles in an external magnetic field. The particles are assumed to interact in pairs, with no restriction being imposed on the strength of the dipole–dipole interaction. The characteristics of jumps in magnetization are calculated as a function of the magnetic anisotropy of the particles and the strength of the dipole–dipole interaction between them. The proposed model is shown to adequately describe the main characteristics of the fine structure of the FMR spectrum observed experimentally in a disperse system in [4].

2. PHYSICAL MODEL

The energy of a system of two interacting uniformly magnetized ferromagnetic spherical particles having uniaxial magnetocrystalline anisotropy is written as

$$E(\theta_1, \varphi_1, \theta_2, \varphi_2) = -\mu\mathbf{H}(\mathbf{n}_1 + a\mathbf{n}_2) - K_a V[(\mathbf{n}_1\mathbf{e}_1)^2 + a(\mathbf{n}_2\mathbf{e}_2)^2] + K_d[(\mathbf{n}_1\mathbf{n}_2) - 3(\mathbf{n}_d\mathbf{n}_1)(\mathbf{n}_d\mathbf{n}_2)]. \quad (1)$$

Here, V and aV are the volumes of the first and second particles, respectively; $\boldsymbol{\mu}_1 = \mu\mathbf{n}_1$ and $\boldsymbol{\mu}_2 = a\mu\mathbf{n}_2$ are the magnetic moments of the particles; K_a is the magnetocrystalline anisotropy constant; $K_d = a(J_s V)^2/d^3$ is the dipole–dipole interaction constant, where J_s is the saturation magnetization; (θ_1, φ_1) and (θ_2, φ_2) are the angles defining the orientations of the magnetic moments $\boldsymbol{\mu}_1$ and $\boldsymbol{\mu}_2$, respectively, with respect to the external magnetic field; \mathbf{e}_1 and \mathbf{e}_2 are unit vectors directed along the easy axes of the first and second particles, respectively; and $\mathbf{d} = d\mathbf{n}_d$ is the position vector of one particle with respect to the other ($r_1 + r_2 < d$). Equation (1) can be expressed in terms of dimensionless variables $\varepsilon = E/K_a V$, $h = \mu H/K_a V$, and $k_d = K_d/K_a V = aJ_s^2 V/d^3 K_a$. The coordinate system is chosen so that the z axis is directed along the external magnetic field $\mathbf{H} = (0, 0, H)$ and the vector \mathbf{d} lies in the yz plane. The spherical coordinates of the vectors \mathbf{e}_1 and \mathbf{e}_2 and the position vector \mathbf{d} are designated as $(\theta_{e1}, \varphi_{e1})$, $(\theta_{e2}, \varphi_{e2})$, and (θ_d, φ_d) , respectively. The energy ε is minimized with the MATLAB program package using the BFGS method for the magnetic field h varying monotonically along the path $\infty \rightarrow 0 \rightarrow \infty$.

3. RESULTS AND DISCUSSION

If the interparticle interaction is weak ($k_d \ll 1$), the orientation of the magnetic moments of the particles varies monotonically as the external magnetic field decreases gradually to zero. Classical hysteresis is observed when a magnetic field is then applied in the opposite direction [14]. In the case of stronger interparticle interactions, the magnetic moment of the particles changes jumpwise with positive magnetic field at certain parameter values of the system. As an example, Fig. 1 shows the dependence of the angle θ_2 between the vectors $\boldsymbol{\mu}_2$ and \mathbf{H} on the magnetic field strength for several values of k_d and the following parameter values: $a = 0.2$, $\theta_{e1} = 67^\circ$, $\theta_{e2} = 134^\circ$, $\theta_d = 94^\circ$, and $\varphi_{e1} = \varphi_{e2} = \varphi_d = 0$. It can be seen that, over the range $0.116 < k_d < 0.431$, the direction of the magnetic moment $\boldsymbol{\mu}_2$ changes jumpwise with positive field without changing the sign of the field. Naturally, the direction of the magnetic moment of the larger particle $\boldsymbol{\mu}_1$ also changes jumpwise at the same value of the magnetic field. This example is not unique. Stepwise magnetization in positive fields (SWM PF) can occur at various values of k_d

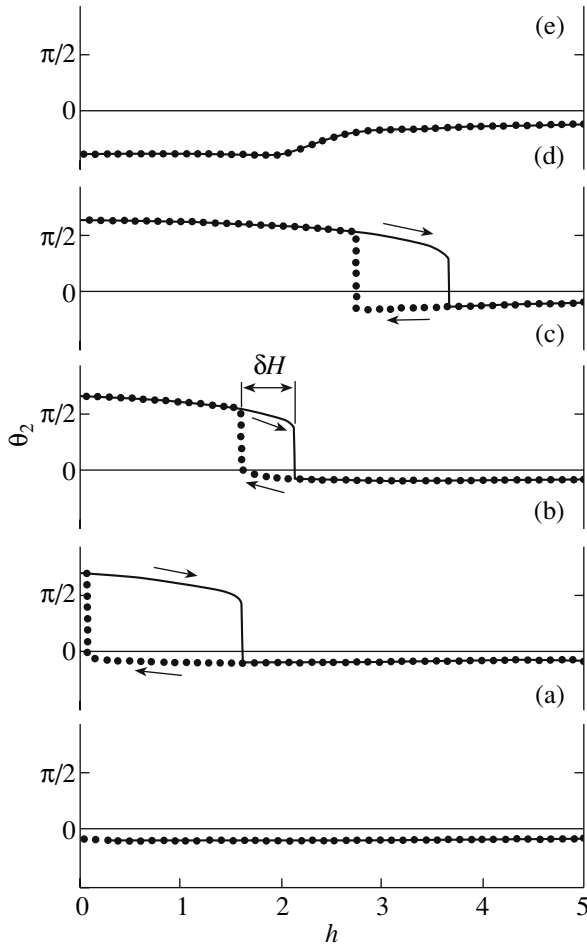


Fig. 1. Dependence of the angle θ_2 between the vectors $\boldsymbol{\mu}_2$ and \mathbf{H} on the external magnetic field h for a system of two interacting particles calculated for $a = 0.2$, $\theta_{e1} = 67^\circ$, $\theta_{e2} = 134^\circ$, $\theta_d = 94^\circ$, $\varphi_{e1} = \varphi_{e2} = \varphi_d = 0$, and various values of k_d : (a) 0.114, (b) 0.116, (c) 0.200, (d) 0.430, and (e) 0.432.

and a over certain ranges of values of the angles defining the orientation of the vectors \mathbf{e}_1 , \mathbf{e}_2 , and \mathbf{n}_d . This region of values of θ_{e1} , φ_{e1} , θ_{e2} , φ_{e2} , θ_d , and φ_d proved to be fairly complicated. Indeed, this can be seen from a cross section of this region for fixed values of certain angles. As an example, Fig. 2 shows the dependence of the field h_j at which the magnetization changes jumpwise on the angles θ_d and φ_d for $k_d = 1$, $a = 1$, $\varphi_{e1} = \varphi_{e2} = \theta_{e1} = 0$, and $\theta_{e2} = 40^\circ$.

In order to analyze magnetization reversal in a real disperse system, one should solve a many-particle problem. However, the strength of magnetic dipole–dipole interaction decreases rapidly with interparticle distance. Therefore, for diluted disperse magnets, a model of pairwise interaction of randomly oriented particles is a fairly good approximation. Using the Monte Carlo method, we found the magnetization jump probability distribution $\Delta N/N$ over magnetic-field values in positive fields for $a = 1$ and $k_d = 1$ and for $a = 0.2$ and

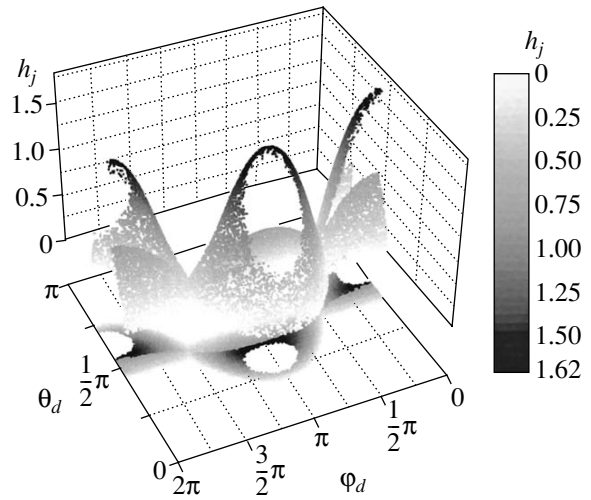


Fig. 2. Dependence of the magnetic field h_j ($h_j > 0$) in which a magnetization jump occurs on the angles θ_d and φ_d defining the orientation of the relative position vector \mathbf{d} of two particles calculated for $k_d = 1$ and $a = 1$. The vector \mathbf{e}_1 along the easy axis of the first particle is aligned with the direction of the external magnetic field ($\theta_{e1} = 0$, $\varphi_{e1} = 0$). For the second particle, $\theta_{e2} = 40^\circ$ and $\varphi_{e2} = 0$. The plot is calculated using the Monte Carlo method.

$k_d = 1.2$ (Figs. 3a and 3b, respectively). In the former case, the overall probability of SWM PF is 0.0937 ± 0.0005 . It can be seen from Fig. 3 that the maximum value of the magnetic field H in which magnetization jumps occur can noticeably exceed K_a/J_s . Figure 4 shows the dependence of the SWM-PF probability density $\rho_{\text{SWM-PF}}$ on the angle θ_d calculated for $a = 1$ and $k_d = 1$. It is seen that SWM PF occurs predominantly for pairs of particles whose relative position vector is perpendicular to the external magnetic field. For example, the SWM-PF probability density for $\theta_d = \pi/2$ is greater than that for $\theta_d \approx 0.1$ by a factor of about 50. The proportion of pairs of particles for which SWM PF is observed is plotted in Fig. 5 as a function of k_d . For $a = 1$ and $k_d = 1$, this proportion corresponds to the case considered above (Fig. 3a). Thus, according to our calculations, SWM PF occurs (at least for $0.1 \leq a \leq 1.0$) if the value of k_d lies in the range

$$0.5a \leq k_d \leq 1.5. \quad (2)$$

Jumps are absent at large values of k_d because the magnetic moments of the particles in this case are strongly correlated and exhibit a cooperative behavior, as a part of a single whole [21]. In the limiting case of weak interparticle interactions, the dipole–dipole interaction has almost no effect on the process of magnetization of the system and jumps occur only in negative fields [14]. In positive fields, magnetization jumps occur only if the competing contributions to the energy of the system are comparable in magnitude. In [2], a

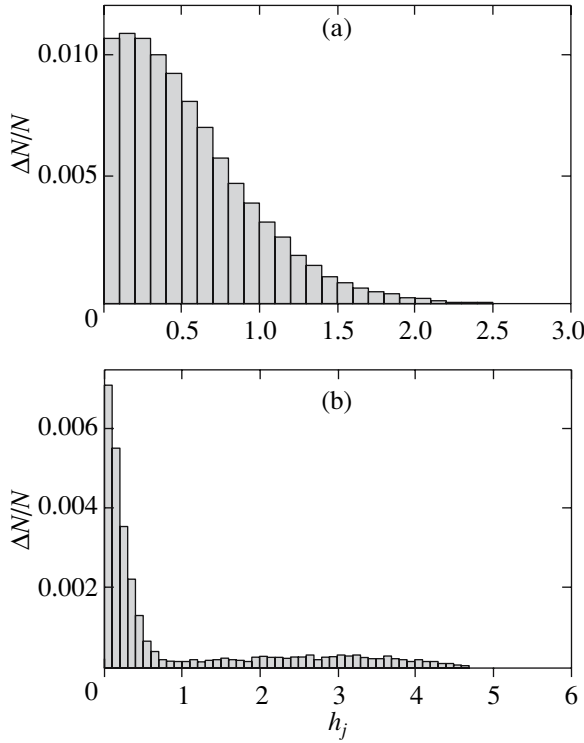


Fig. 3. Magnetization jump probability distribution $\Delta N/N$ with respect to the external magnetic field in a disperse system of randomly oriented magnetic dipoles interacting in pairs calculated for (a) $a = 1$ and $k_d = 1$ and (b) $a = 2$ and $k_d = 1.2$. The bar charts are obtained by averaging the results of 10^6 trials.

particular case of weakly interacting identical particles was considered, with the easy axes of the particles being parallel to the external field and $\theta_d = \pi/2$. It was shown that an increase in k_d leads to a decrease in the energy barrier that the system of interacting particles should overcome during magnetization reversal. At $k_d = 2/3$, the coercive field of a pair of particles vanishes. The range $k_d > 2/3$, wherein magnetization jumps can occur in positive fields, was not considered in [2].

The model of spherical particles imposes a geometrical restriction on the maximum value of k_d . Indeed, the minimum distance between the centers of spherical particles is $d_{\min} = r_1 + r_2$, where r is the radius of a particle. Therefore, the dipole–dipole interaction is limited by the condition $k_d^{\max} = 4\pi a J_s^2 / 3K_a(1 + a^{1/3})^3$. As shown above, SWM PF can occur if $k_d \geq 0.5a$; therefore, due to the finite size of particles, magnetization jumps can occur in positive fields if $4\pi J_s^2 / (3K_a(1 + a^{1/3})^3) \geq 0.5$. In the case where $4\pi a J_s^2 / 3K_a(1 + a^{1/3})^3 \geq 1.5$, the finite size of particles, according to condition (2), does not impose any restrictions on the occurrence of SWM PF.

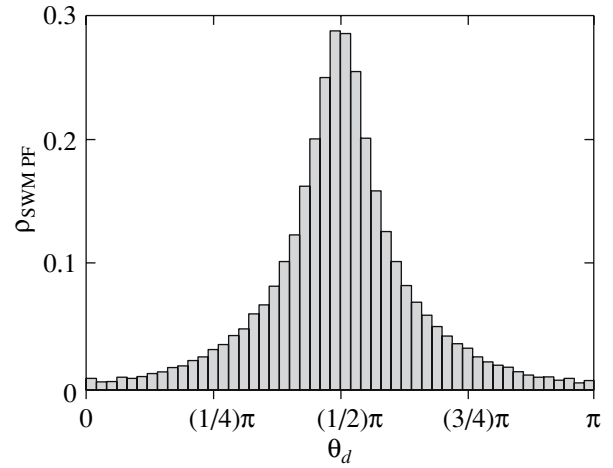


Fig. 4. Probability density $\rho_{\text{SWM PF}}$ of jumps in magnetization in positive magnetic fields as a function of angle θ_d for a disperse system of randomly oriented magnetic dipoles interacting in pairs calculated for $k_d = 1$ and $a = 1$. The bar chart is obtained by averaging the results of 10^6 trials.

It is well known that the process of magnetization of a disperse system depends significantly on the shape of the particles of which the system is comprised. Let us analyze the case where the magnetic anisotropy energy of a particle is mainly due to the shape anisotropy. We consider particles shaped like an oblong ellipsoid (with principal axes $l_{\parallel} > l_{\perp}$). We assume that $l_{\parallel} \approx l_{\perp}$ and, hence, the dipole–dipole interaction energy E_D^{el} of ellipsoidal particles is approximately equal to the corresponding energy E_D of spherical particles. In this case, the energy of the system is

$$\varepsilon_f = -h_f \mathbf{n}_h (\mathbf{n}_1 + a \mathbf{n}_2) - \left[(\mathbf{n}_1 \mathbf{e}_1)^2 + a \frac{\Delta N_2}{\Delta N_1} (\mathbf{n}_2 \mathbf{e}_2)^2 \right] + k_f [(\mathbf{n}_1 \mathbf{n}_2) - 3(\mathbf{n}_d \mathbf{n}_1)(\mathbf{n}_d \mathbf{n}_2)], \quad (3)$$

where $\varepsilon_f = 2E/J_s^2 V \Delta N_1$; $h_f = 2H/J_s \Delta N_1$; $k_f = 2aV/d^3 \Delta N_1$; $N_{\parallel 1}$, $N_{\perp 1}$, $N_{\parallel 2}$, and $N_{\perp 2}$ are the demagnetizing factors of the first and second particles, respectively; $\Delta N_1 = N_{\perp 1} - N_{\parallel 1}$ and $\Delta N_2 = N_{\perp 2} - N_{\parallel 2}$; and \mathbf{e}_1 and \mathbf{e}_2 are unit vectors parallel to the easy axes of the corresponding particles (which are directed along the principal axis a of the ellipsoid in the case under study). From comparing Eqs. (1) and (3), it follows that, in the case where the magnetic anisotropy of particles is mainly due to the shape anisotropy, SWM PF can also occur with characteristics coinciding with those determined above to within the definition of the dimensionless parameters (for $\Delta N_1 = \Delta N_2 = \Delta N$). For example, an analog of condition (2) is $(4/3)(a/\Delta N) \leq d^3/V \leq 4/\Delta N$. Moreover, estimates show that the finite size of particles does not impose any restrictions on the occurrence of SWM PF

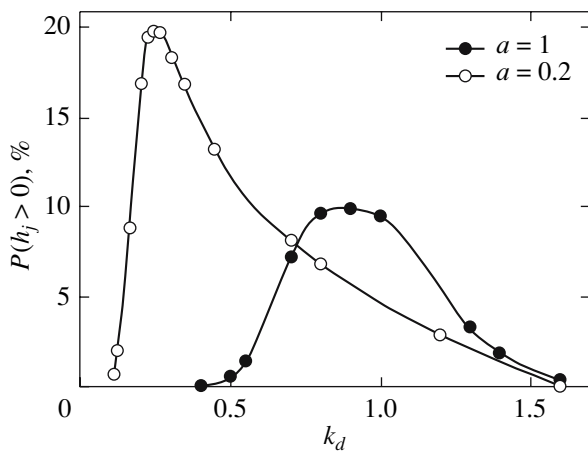


Fig. 5. Proportion of pairs of particles for which SWM PF occurs plotted as a function of k_d for $a = 1.0$ and 0.2 .

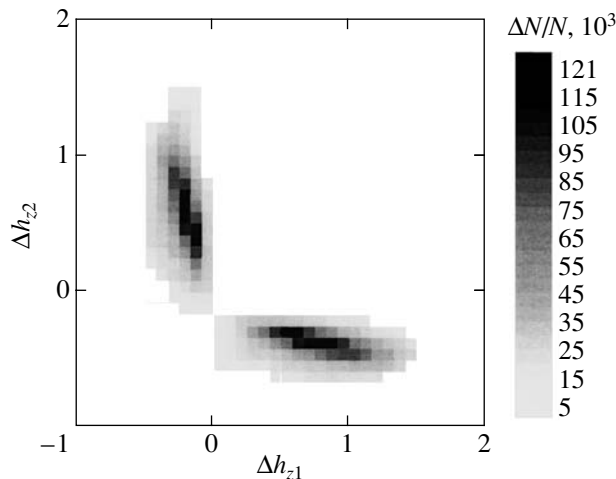


Fig. 6. Bar chart of the SWM-PF distribution over the changes in the local field projections on the direction of the external magnetic field calculated for two interacting particles for $k_d = 1$ and $a = 1$.

in this case. For $a = 0.2$ and $\Delta N = 2$, we find that $H_j^{\max} \geq 5J_s$. Thus, in the case where the magnetic anisotropy energy is mainly due to the shape anisotropy rather than the magnetocrystalline anisotropy (i.e., where $J_s^2 \Delta N / 2 \gg K_a$), the field H_j^{\max} below which SWM PF is observed increases significantly.

We carried out computer simulation of magnetization jumps in dispersed ferromagnets with inclusion of interparticle interactions in the limiting case of a zero temperature or of sufficiently large particles, where thermal fluctuations of the magnetic moments of the particles are negligible. Otherwise, the magnetic

moment of a particle exhibits a superparamagnetic behavior and the transition of the system from one local minimum to another with a decrease in the external magnetic field occurs at a higher field value. This effect, in turn, causes the SWM-PF distribution to broaden to higher magnetic fields.

It was shown experimentally in [4] that, due to the specific features of magnetization of a dispersed magnet, a fine structure arises in the FMR spectrum. The above analysis permits us to explain this effect. A jumpwise change in the magnetization causes a change in the local magnetic fields exerted on the particles. Figure 6 shows the changes in the projections of the local fields on the direction of the external magnetic field during SWM PF for a system of particles interacting in pairs ($k_d = 1$). It can be seen that the changes ΔH_z for the first and second particles differ noticeably. The process under study can change absorption in the FMR spectrum if, before or after the occurrence of a magnetization jump, the particle is in a local magnetic field corresponding to noticeable absorption. It is also necessary for the change in the local field for the particle in question not to be substantially smaller than the absorption line width. Moreover, since the hysteresis width during SWM PF (δH in Fig. 1) is usually greater than the modulation amplitude of the external magnetic field, the reproducible change in the FMR spectrum can be observed only if the values of the derivative of the absorption (measured experimentally) before and after a magnetization jump differ substantially. The above conditions are satisfied for most disperse magnets [22]. For example, the typical FMR absorption line width for Ni-, Co-, and Fe-based dispersions is $\sim 10^2$ – 10^3 G [22–24]. The change in the local field due to SWM PF $\sim \max(K_a/J_s, J_s)$ is of the same order of magnitude (Fig. 6). In the limiting case where the change in the local field due to SWM PF is greater than the absorption line width and the particle is under the resonance conditions before or after the magnetization jump, the absorption intensity changes jumpwise by a quantity equal to the contribution from this particle to the total FMR spectrum. It should be noted that stepwise magnetization of a pair of particles can cause a noticeable change in the local field exerted on a third particle that is located in the vicinity of the pair but does not have a significant effect on SWM PF. If the FMR absorption of the third particle is strong and the corresponding absorption line width is small, then an even small change in the local field can produce a significant contribution to the FMR fine structure.

Figure 7 shows the FMR fine structure measured for a model Ni-based disperse system. Based on the above results and the experimental data from [4], the FMR fine structure in low fields (up to ~ 1000 G) can be explained in terms of stepwise magnetization of single-domain particles and the FMR fine structure in high fields can be assigned to magnetization reversal in multidomain particles. Indeed, the two-particle model as

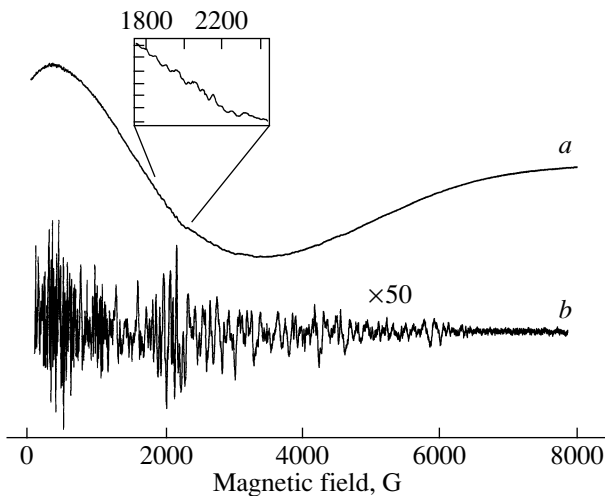


Fig. 7. (a) FMR spectrum of a model Ni-based sample obtained by ultrasonic Ni (5 wt %) dispersion in a molten paraffin; according to electron microscopy data, the particles range in size from 0.1 to 10 μm . (b) The FMR fine structure obtained by subtracting the broad smooth component.

applied to single-domain Ni particles predicts a high probability of SWM PF at fields of up to $\approx 2J_s = 1100$ G (Fig. 3). For almost spherical Ni particles, we have $k_d^{\max} \approx 4.2 > 0.5$ and, therefore, SWM PF can occur for any ratio between their volumes. However, according to condition (2), the geometrical restrictions decrease the probability of SWM PF events for spherical Ni particles with $a < 0.17$. Since not all of the particles that undergo SWM PF cause absorption in the field h_j of a magnetization jump, the intensity of the FMR fine structure will be proportional not only to the probability of SWM PF but also to the number of particles causing absorption in the field h_j . Thus, the fact that the intensity of the FMR fine structure does not increase significantly as the magnetic field decreases to zero (Fig. 7) can be due to a decrease in the total absorption of the particles in this field.

4. CONCLUSIONS

Thus, we have numerically analyzed the behavior of a disperse system consisting of magnetic particles (interacting in pairs) in an external magnetic field. It has been shown that the magnetization of the system exhibits hysteresis as the external magnetic field varies without reversing direction. This effect occurs for a certain geometry of particles over wide ranges of energies of magnetic anisotropy and dipole–dipole interaction between the particles. We have calculated the probability of this effect occurring in a disperse system of randomly oriented particles. The characteristic values of the external magnetic field that correspond to magneti-

zation jumps occurring in positive fields can exceed $\max(K_a/J_s, J_s)$. We have analyzed the conditions under which narrow lines of the FMR fine structure arise due to jumpwise changes in the resonance absorption conditions for particles coupled via dipole–dipole interaction. Based on the results obtained, we have qualitatively explained the main characteristics of the FMR fine structure observed experimentally in a model disperse system.

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