
ORDER, DISORDER AND PHASE TRANSITIONS
IN CONDENSED MEDIA

THE INFLUENCE OF MAGNETIC FIELD AMPLITUDE ON THE
MAGNETIZATION REVERSAL KINETICS OF MAGNETIC NANOPARTICLES

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Abstract. The influence of magnetic field amplitude on magnetization reversal kinetics and the magnetic hyperthermia effect produced by a single-domain ferromagnetic particle immobilized in a non-magnetic medium has been theoretically investigated. The calculation results, based on the mathematically regular Kramers theory, show that the dissipation W of alternating magnetic field energy in the particle can increase with field amplitude faster than according to the quadratic law $W \sim H_0^2$. This conclusion, at least in principle, explains recent experiments on magnetic hyperthermia in systems of immobilized particles, where the dependence was discovered $W \sim H_0^\gamma, \gamma > 2$.

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1. INTRODUCTION

Composite systems consisting of magnetic nanoparticles embedded in a non-magnetic carrier medium attract great interest from researchers and practitioners due to their rich set of unique physical properties, which find active applications in many modern and promising industrial and biomedical technologies. Reviews of works on this topic can be found, for example, in [1–5]. In particular, the magnetic hyperthermia method for cancer treatment is based on introducing magnetic nanoparticles into a diagnosed tumor area and heating these particles with an alternating magnetic field to temperatures (typically above 41–42°C) at which tumor cells die [6–14]. The first theoretical work [15] on magnetic hyperthermia was based on phenomenological Debye equations for the remagnetization of magnetic nanoparticles using the approximation of linear dependence of particle magnetization on the external field. Cases of particles immobilized in an external medium and particles freely rotating in a viscous fluid were considered. In works [16], the remagnetization kinetics of immobilized ferromagnetic particles was studied using statistical physics methods based on the Fokker-Planck equation for the orientation distribution function of the particle's magnetic moment. Estimates of the

particle's magnetization relaxation time in a constant magnetic field are provided. Theoretical studies of the remagnetization kinetics of a stationary particle in an alternating field were conducted in [17] for arbitrary values of magnetic anisotropy parameter and field amplitude. Within this approach, to determine the statistically average magnetization of the particle, it is necessary to solve, strictly speaking, an infinite system of coupled differential equations, and justifying the possibility of truncating it while maintaining a finite number of equations represents a separate problem. Analysis shows that the number of equations in the system that must be retained to obtain results with acceptable accuracy rapidly increases with increasing particle magnetic anisotropy parameter and magnetic field amplitude. Therefore, the task of deriving compact, sufficiently convenient equations for particle remagnetization kinetics remains relevant, even if under certain restrictions on the system's physical parameters.

One of the most important characteristics of magnetic hyperthermia as a physical phenomenon is the dependence of the intensity of W magnetic field energy dissipation (heat generation intensity) on the frequency and amplitude of the magnetic field. For small H_0 field amplitudes, where the approximation of linear dependence of magnetization on the field

holds true, the relation $W \sim H_0^\gamma$ is valid. An increase in the field leads to a slower than linear dependence of the particle system magnetization on the field, therefore it is natural to expect that this should lead to a slower than quadratic dependence of energy dissipation on the field amplitude. However, unexpectedly, recent experiments [18] showed a faster than quadratic dependence $W \sim H_0^\gamma$, $\gamma > 2$.

The aim of this work is, firstly, to derive a compact, convenient equation for the magnetization reversal kinetics of a ferromagnetic particle in alternating fields, which can be used in a fairly wide range of field amplitudes. Secondly, based on this equation, to show that a faster than quadratic dependence of the W energy dissipation intensity on the alternating field amplitude H_0 is quite possible and is not a consequence of methodological or other experimental errors.

For this purpose, we will consider a single-domain uniaxial ferromagnetic particle immobilized in a non-magnetic medium. Note that the immobilization of magnetic nanoparticles often occurs, for example, when they are introduced into biological tissues [19, 20]. We limit ourselves to analyzing strong magnetic anisotropy of the particle, i.e., the energy of this anisotropy is assumed to be much greater than the thermal energy of the system. Since, as is known, the magnetic anisotropy energy is proportional to the particle volume, this means that the particle size is not very small. For example, it can be shown that for magnetite particles, often used in experiments and applications, the magnetic anisotropy energy exceeds thermal energy at room temperatures when the particle diameter is greater than 16–18 nm (estimates of physical characteristics of magnetite particles can be found, for example, in [21]). Note that the surface features of the nanoparticle can make an additional contribution to its magnetic anisotropy energy. Therefore, in reality, the strong anisotropy approximation may hold true even for particles with a diameter significantly smaller than the mentioned 16–18 nm. Finally, we will neglect gyromagnetic effects, which manifest only at high field frequencies (on the order of gigahertz), significantly exceeding the range acceptable for many technological applications.

2. MATHEMATICAL MODEL AND BASIC APPROXIMATIONS

Consider a stationary ferromagnetic particle (see illustration in Fig. 1) placed in an oscillating magnetic field parallel to the particle's easy magnetization axis. Note that in *in vitro* experiments, the direction

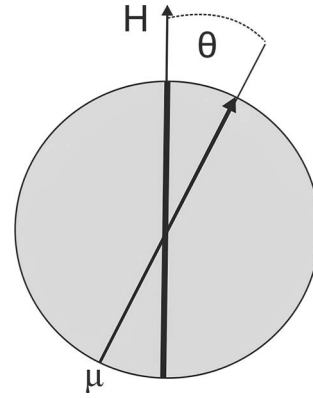


Fig. 1. Illustration of the particle under consideration. Thick line – easy magnetization axis of the particle

of particles' easy magnetization axes can be fixed along a chosen direction if the polymerization of the magnetic composite is carried out in a sufficiently strong constant field.

Let us denote μ – a unit vector directed along the magnetic moment of the particle (see Fig. 1) and $f(\mu)$ – a normalized to unity distribution function (probability density) over the orientations of vector μ . This function can be found as a solution to the Fokker-Planck equation, which, taking into account the axial symmetry of the problem, can be written as [16, 17, 22]

$$2\tau_D \frac{\partial f}{\partial \tau} = -\frac{1}{\sin \theta} \frac{\partial}{\partial \theta} j. \quad (1)$$

Here j is the probability flux density in space μ :

$$\begin{aligned} j(\theta) &= -\sin \theta \left(f \frac{\partial u}{\partial \theta} + \frac{\partial f}{\partial \theta} \right) = \\ &= -\sin \theta e^{-u} \frac{\partial}{\partial \theta} (f e^u). \end{aligned} \quad (2)$$

Parameter τ_D is the characteristic time of rotational diffusion of the particle's magnetic moment, determined by its internal crystal structure; θ is the polar angle of deviation of vector μ from the particle's magnetic anisotropy axis (hence, from the direction of the oscillating field, see Fig. 1); u – and dimensionless, with respect to absolute temperature $k_B \Theta$ particle energy, Θ is the absolute temperature in kelvins. Energy u can be represented as

$$\begin{aligned} u &= -\sigma \cos^2 \theta - h \cos \theta, \\ h &= \mu_0 \frac{m}{k_B \Theta} H, \quad \sigma = \frac{K_V p}{k_B \Theta}, \end{aligned}$$

Here μ_0 is the magnetic permeability of vacuum; m – is the absolute value of the particle's magnetic moment; k – is the particle's magnetic anisotropy parameter; v_p and d_p are the volume and diameter of the particle; h is the ratio of Zeeman interaction energy of the particle with magnetic field H to thermal energy $k_B\Theta$, which can be considered as dimensionless magnetic field; σ – is the dimensionless parameter of particle's magnetic anisotropy. Note that for magnetite particles typically used in medical and biological applications, by order of magnitude $\tau_D \sim 10^{-9}$ s (see, for example, [21]).

In general case, the solution of equation (1) in finite analytical form has not been obtained. Here we consider the case of strong magnetic anisotropy, when strong inequalities are satisfied $\sigma \gg 1$, $\sigma \gg h$. Note that no other restrictions on the magnitude of magnetic field are assumed.

Within the accepted approximations in space μ there are two potential wells separated by a potential barrier, i.e., function $u(\theta)$ at $0 \leq \theta \leq \pi$ has two minima and one maximum.

The potential minima correspond to $\theta = 0$ and $\theta = \pi$; they are equal to respectively.

$$\begin{aligned} u_0 &= -\sigma - h, \\ u_\pi &= -\sigma + h. \end{aligned} \quad (4)$$

The value θ_{\max} of angle θ , corresponding to maximum u , is determined from equation $\cos \theta_{\max} = -h/2\sigma$; the potential value at the maximum point equals

$$u_{\max} = \frac{h^2}{4\sigma}. \quad (5)$$

Let's introduce probabilities P_0 and P_π that vector μ belongs to potential wells $0 \leq \theta \leq \pi/2$ and $\pi/2 \leq \theta \leq \pi$ respectively. They can be determined as follows:

$$\begin{aligned} P_0 &= \int_0^{\max} f(\mu) \sin \mu \, d\mu, \\ P &= \int_0^{\max} f(\mu) \sin \mu \, d\mu. \end{aligned} \quad (6)$$

Obviously, the normalization condition $P_0 + P_\pi = 1$ must be satisfied. To find P_0 , we integrate both sides of equation (1) in the same way as in the

first integral (6). After simple transformations, we obtain

$$2\tau_D \frac{\partial P_0}{\partial t} = -j(\theta_{\max}). \quad (7)$$

To find $j(\theta_{\max})$, we will use the basic ideas of Kramers' classical theory of Brownian particle diffusion through a potential barrier (see also [23]). Following this method, we rewrite equation (2) as

$$j(\theta) \frac{e^u}{\sin \theta} = -\frac{\partial}{\partial \theta} (f e^u) \quad (8)$$

and integrate both sides of (8) over θ from 0 to π :

$$\int_0^\pi j(\theta) \frac{e^u}{\sin \theta} d\theta = -[f_\pi e^{u_\pi} - f_0 e^{u_0}], \quad (9)$$

$$f_0 = f(0), \quad f_\pi = f(\pi).$$

Since $\sigma \gg 1$, function $u(\theta)$ near its maximum has the form of a high sharp peak:

$$u_{\max} - u_{0,\pi} \gg 1, \quad \frac{d^2 u}{d\theta^2} \Big|_{\theta_{\max}} \gg 1.$$

Therefore, we can evaluate the integral in the left part of (9) using standard steepest descent method considerations:

$$\begin{aligned} \int_0^\pi j(\theta) \frac{e^u}{\sin \theta} d\theta &\approx j(\theta_{\max}) \frac{e^{u_{\max}}}{\sin \theta_{\max}} \sqrt{\frac{\pi}{\Omega}}, \\ \Omega &= -\frac{1}{2} \frac{d^2 u}{d\theta^2} \Big|_{\theta_{\max}}. \end{aligned} \quad (10)$$

In linear approximation for small ratio h/σ we obtain $\Omega = \sigma$ and $\sin \theta_{\max} = 1$. Consequently,

$$\int_0^\pi j(\theta) \frac{e^u}{\sin \theta} d\theta \approx j(\theta_{\max}) \sqrt{\frac{\pi}{\sigma}} e^{u_{\max}}. \quad (11)$$

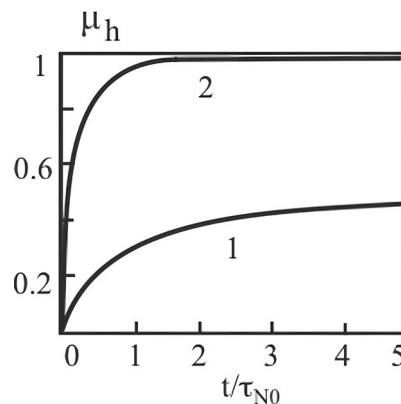


Fig. 2. Statistically averaged projection μ_h of vector μ as a function of time. Numbers near curves: 1 – $h = 0.5$; 2 – $h = 2.5$

Combining (9) and (11), we obtain

$$j(\theta_{max}) \approx - \left(\sqrt{\frac{\pi}{\sigma}} e^{u_{max}} \right)^{-1} \left[f_{\pi} e^{u_{\pi}} - f_0 e^{u_0} \right]. \quad (12)$$

Using (12) in equation (7), considering relations (4) and (5), replacing for simplification the approximate equality sign " \approx " with the equality sign "=", within the approximation $\sigma \gg 1$ we arrive at equation

$$\begin{aligned} \frac{\partial P_0}{\partial t} &= \frac{1}{2\tau_I} \left[f_{\pi} e^h - f_0 e^{-h} \right], \\ \tau_I &= \tau_D \sqrt{\frac{\pi}{\sigma}} \exp \left\{ \sigma + \frac{h^2}{4\sigma} \right\}. \end{aligned} \quad (13)$$

Let's now determine the relationship between probability P_0 and values f_0, f_{π} of probability density f at points $\theta = 0$ and $\theta = \pi$ of potential minima u . For this, as in Kramers' classical theory, we consider that the vector μ transition through a high potential barrier is a relatively rare phenomenon, and the characteristic time between two such transitions is much longer than the time of thermodynamic equilibrium establishment in each potential well, which is of the order of τ_D . Therefore, at any moment we can consider the state inside such a well as practically equilibrium, described by the Boltzmann distribution function. Obviously, this does not mean equilibrium distribution between potential wells. Thus, in each of the potential wells $0 \leq \theta \leq \pi/2$ and $\pi/2 \leq \theta \leq \pi$, according to Boltzmann distribution we have

$$f(\theta) = f_0 \exp\{u_0 - u(\theta)\} \quad (14)$$

and

$$f(\theta) = f_{\pi} \exp\{u_{\pi} - u(\theta)\}. \quad (15)$$

Substituting (14) into the first relation (6), we obtain

$$\begin{aligned} P_0 &= f_0 \int_0^{\theta_{max}} \exp\{u_0 - u(\theta)\} \sin \theta d\theta = \\ &= f_0 \int_0^{\theta_{max}} \exp\{-\sigma(1 - \cos^2 \theta)\} \exp\{-h(1 - \cos \theta)\} \times \\ &\quad \times \sin \theta d\theta. \end{aligned} \quad (16)$$

Since $\sigma \gg 1$, function $\exp\{-\sigma(1 - \cos^2 \theta)\}$ has a sharp maximum at $\theta = 0$. Due to inequality $\sigma \gg h$ exponential $\exp\{-h(1 - \cos \theta)\}$ changes with θ much slower than $\exp\{-\sigma(1 - \cos^2 \theta)\}$. Consequently, using the steepest descent method, we can represent

$$\begin{aligned} P_0 &\approx f_0 \int_0^{\theta_{max}} \exp\{-\sigma x^2\} dx = \\ &= f_0 \frac{1}{2\sigma} \left(1 - \exp\{\sigma \theta_{max}^2\} \right) \approx f_0 \frac{1}{2\sigma}. \end{aligned} \quad (17)$$

Similarly

$$P_{\pi} \approx f_{\pi} \frac{1}{2\sigma}. \quad (18)$$

Below for convenience in equations (17) and (18) we will use the equality sign "=" instead of the approximate equality " \approx ".

Due to the normalization condition $P_0 + P_{\pi} = 1$, using (17) and (18), we arrive at the equality

$$f_0 + f_{\pi} = 2\sigma. \quad (19)$$

Combining relations (17), (18), and (19) with (13), we obtain the equation

$$\begin{aligned} \frac{\partial f_0}{\partial t} &= \frac{1}{\tau_{II}} \left[\sigma e^h - f_0 \operatorname{ch} h \right], \\ \tau_{II} &= 2 \frac{\tau_I}{\sigma} = 2\tau_D \sqrt{\frac{\pi}{\sigma^3}} e^{\sigma + \frac{h^2}{4\sigma}}. \end{aligned} \quad (20)$$

The statistically average projection of the unit vector μ on the field direction \mathbf{H} can be calculated as follows:

$$\mu_h = \mu_{h0} + \mu_{h\pi}, \quad (21)$$

where

$$\begin{aligned} \mu_{h0} &= \int_0^{\theta_{max}} f(\theta) \cos \theta \sin \theta d\theta, \\ \mu_{h\pi} &= \int_{\theta_{max}}^{\pi} f(\theta) \cos \theta \sin \theta d\theta. \end{aligned} \quad (22)$$

Taking into account (14), (15), and (19), equation (22) can be rewritten as

$$\begin{aligned} \mu_{h0} &= f_0 \int_0^{\theta_{max}} \exp\{-\sigma(1 - \cos^2 \theta)\} \exp\{-h(1 - \cos \theta)\} \times \\ &\quad \times \cos \theta \sin \theta d\theta \approx f_0 \frac{1}{2\sigma}, \end{aligned} \quad (23a)$$

$$\begin{aligned} \mu_{h\pi} &= f_{\pi} \int_{\theta_{max}}^{\pi} \exp\{-\sigma(1 - \cos^2 \theta)\} \exp\{h(1 + \cos \theta)\} \times \\ &\quad \times \cos \theta \sin \theta d\theta \approx -f_{\pi} \frac{1}{2\sigma}. \end{aligned} \quad (23b)$$

Here, as in (17), (18), the saddle point method for evaluating definite integrals is used again.

Substituting (21), (23) into (20), we arrive at the equation

$$\frac{\partial \mu_h}{\partial t} = \frac{1}{\tau_{II}} [\text{sh } h - \mu_h \text{ ch } h] \quad (24)$$

or

$$\begin{aligned} \frac{\partial \mu_h}{\partial t} &= \frac{1}{\tau_N} [\text{th } h - \mu_h], \\ \tau_N &= \frac{\tau_{II}}{h} = \tau_{N0} \frac{h^2}{e^{4\sigma}}, \\ \tau_{N0} &= 2\tau_D \sqrt{\frac{\pi}{\sigma^3}} e^{\sigma}. \end{aligned} \quad (25)$$

3. RESULTS AND DISCUSSIONS

If h is time-independent, for example, after a step change in the field, parameter τ_N is constant and plays the role of characteristic Neel relaxation time μ_h to its equilibrium value $\text{th } h$; τ_{N0} is the value of τ_N at $h = 0$. The obtained estimate τ_{N0} coincides with that obtained in the limit of very high magnetic anisotropy of the particle. As seen from (25), τ_N decreases with increasing magnetic field h (recall, strong inequality $h \ll \sigma$ is assumed to hold), which corresponds to the conclusions of [16, 17].

When h varies with time t , the value τ_N depends on t and cannot be considered as relaxation time. Some calculation results for $\mu_h(t)$ after a step change of h from zero to some constant values are shown in Fig. 2. Increasing the final field value h leads to faster relaxation of μ_h to its equilibrium value. Note that after the field is turned off, the relaxation time μ_h to zero will be τ_{N0} , i.e., longer than the relaxation process time when increasing the field. Thus, the processes of system magnetization after field increase and demagnetization after its decrease (turnoff) are generally characterized by different relaxation times.

Let us now consider the case of oscillating field $h = h_0 \cos \omega t$, where $h_0 \ll \sigma$ is its amplitude. Solution (24) with initial condition $\mu_h = 0$ at $t = 0$ has the form

$$\begin{aligned} \mu_h(t) &= \frac{V(t)}{\tau_N} \int_0^t \frac{(\text{sh } h \cos \omega t')}{V(t')} \times \\ &\times \exp \left\{ \left[-\frac{(h_0 \cos \omega t')^2}{4\sigma} \right] \right\} dt', \end{aligned} \quad (26)$$

$$V(t) = \exp \left\{ \left[-\frac{1}{\tau_{N0}} \int_0^t \exp \left\{ \left[-\frac{(h_0 \cos \omega t')^2}{4\sigma} \right] \right\} \times \text{ch}(h_0 \cos \omega t') dt' \right] \right\}. \quad (26)$$

We will define the effective complex susceptibility χ at the signal frequency as follows:

$$\begin{aligned} \chi'(\omega, h_0) &= \frac{1}{h_0} \lim_{T \rightarrow \infty} \frac{1}{T} \int_0^T \mu \cos \omega t dt, \\ \chi''(\omega, h_0) &= \frac{1}{h_0} \lim_{T \rightarrow \infty} \frac{1}{T} \int_0^T \mu \sin \omega t dt. \end{aligned} \quad (27)$$

Note that the susceptibility defined in this way determines the relationship between the average vector μ_h of particle magnetic moment orientation and dimensionless field h .

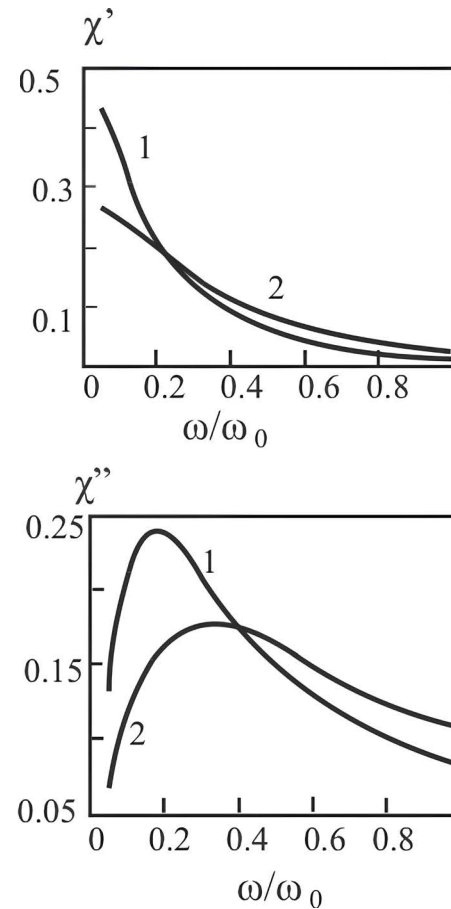


Fig. 3. Real and imaginary parts of effective susceptibility χ versus angular field frequency ω . Numbers next to curves: 1 — $h_0 = 0.5$; 2 — $h_0 = 2$. Parameter $\omega_0 = 2\pi / \tau_{N0}$

The measured material susceptibility, which relates the magnetization of the entire composite and the dimensional field H , is proportional to χ with coefficient

$$\mu_0 \frac{m^2}{v_p k_B T} \varphi,$$

where φ is the volume concentration of particles.

Some calculation results χ' and χ'' are shown in Figs. 3 and 4.

Both parts χ' and χ'' of susceptibility decrease with field amplitude h_0 when the angular frequency ω is significantly less than ω_0 , and increase at relatively high values of ω . To our knowledge, this circumstance has not been previously noted in literature.

It is well known that an alternating magnetic field \mathbf{H} causes heating of ferromagnetic particles, used particularly in cancer therapy (magnetic hyperthermia method). The physical reason for this

heating is the dissipation of magnetic field energy during particle remagnetization.

The average value of dissipation energy over time $T \gg \{2\pi/\omega, \tau_{N0}\}$ can be determined from general thermodynamic considerations (see, for example, [15]):

$$W = -\mu_0 \frac{m}{T} \int_0^T \mu_h \frac{dH}{dt} dt. \quad (28)$$

Using notations (3) and $H = H_0 \cos \omega t$, we easily obtain

$$W = \frac{k_B \Theta}{\tau} w, \quad w = h_0 \omega \frac{\tau_{N0}}{T} \int_0^T \mu_h \sin \omega t dt, \quad (29)$$

$$h_0 = \mu_0 \frac{m}{k_B \Theta} H_0,$$

where w is the dimensionless energy dissipation intensity (heat generation intensity) in the particle.

Some calculation results w as a function of field frequency ω are shown in Fig. 5.

As can be seen, at relatively high frequencies w increases with field amplitude faster than according to the law $w \sim h_0^2$. As ω approaches infinity, w tends to certain saturation. This is a well-known result in magnetic hyperthermia theory.

Some results of the relationship w/h_0^2 to h_0 are shown in Fig. 6.

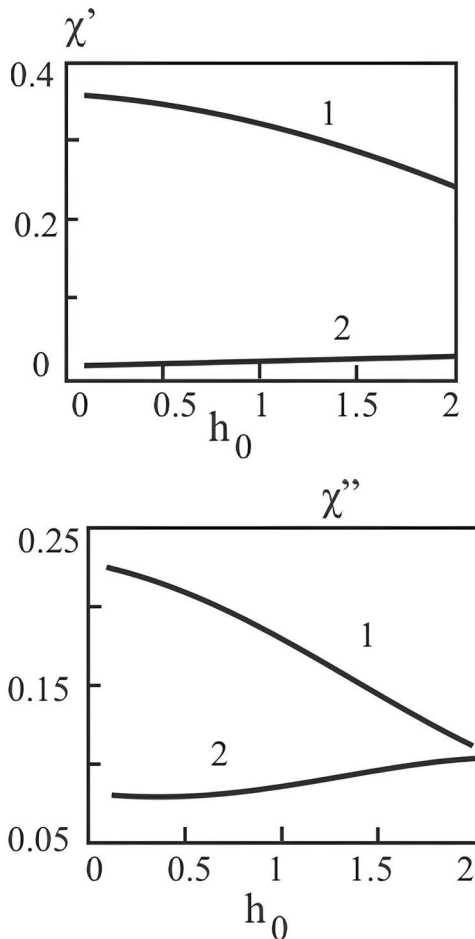


Fig. 4. Real and imaginary parts of effective susceptibility χ versus amplitude h_0 of oscillating field. Curves: 1 — $\omega = 0.1$; 2 — $\omega = \omega_0$. Parameter ω_0 is the same as in Fig. 3

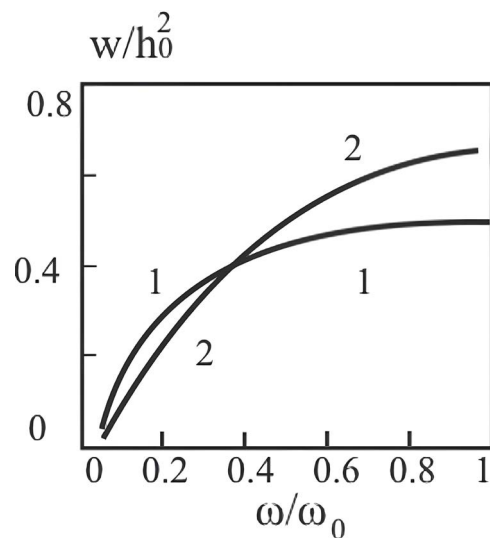


Fig. 5. Dimensionless heat generation intensity in particles versus angular field frequency. Curves: 1 — $h_0 = 0.5$; 2 — $h_0 = 2$. Parameter ω_0 is the same as in Figs. 3, 4

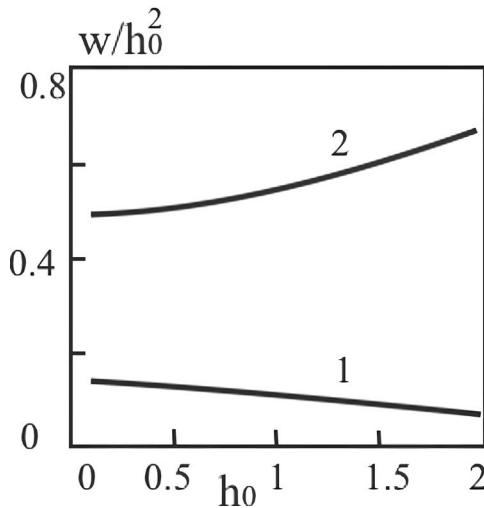


Fig. 6. Dependence of dimensionless energy dissipation intensity w on dimensionless amplitude h_0 of magnetic field. Curves 1 and 2 — $\omega = 0.1\omega_0$ and $\omega = \omega_0$. Parameter ω_0 is the same as in Fig. 3

This figure also shows a faster than H_0^2 dependence of W on H_0 , when the field frequency is sufficiently high. Note that the scaling dependence $W \sim H_0^\gamma$, $\gamma > 2$ in composites with immobilized magnetic particles was discovered in experiments [18]. Thus, the theoretical analysis shows that the increase in heat generation intensity with field amplitude faster than according to the quadratic law is physically possible and cannot be considered as a result of a methodological error in the experiment.

4. CONCLUSIONS

The magnetization reversal kinetics and heat generation in a completely immobile single-domain ferromagnetic particle under the influence of an oscillating magnetic field have been theoretically considered. High magnetic anisotropy of the particle was assumed, which is quite typical for many experiments and technological applications of nanodispersed magnetic composites.

Our calculations, based on the mathematically consistent Kramers theory of Brownian diffusion through a high potential barrier, showed that the magnetic energy dissipation rate W , as expected, depends on the field amplitude H_0 slower than according to the law $W \sim H_0^2$, when the field frequency is relatively low. When the frequency exceeds a certain threshold value, the function $W(H)$ changes faster than H_0^2 . This conclusion

explains, at least qualitatively, recent experimental results [18], which obtained the scaling relation

$$W \sim H_0^\gamma, \quad \gamma > 2.$$

We considered the case of parallel orientation of the magnetic field and the easy magnetization axis of the particle. Within the $\sigma \gg 1$, $\sigma \gg h$ approximation, it is not fundamentally difficult to consider the case of an arbitrary angle β between these vectors. For this, it is sufficient to replace the dimensionless field h with $h \cos \beta$ in the relations obtained here, neglecting the small magnetization component in the direction perpendicular to the particle's anisotropy axis.

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