where $\varepsilon = O(\lambda^{-\nu})$. Further, we use for the denominators the integral representation

$$(x_i-E_i-i0)^{-1}=i\int_0^\infty \exp\{-i\tau_i(x_i-E_i-i0)\}\,d\tau_i.$$

The integral $J_0(\lambda, E_1, E_2)$ is then transformed into a fourfold integral with respect to the variables x_1, x_2 and τ_1, τ_2. The integrals with respect to the variables x_1 and x_2 can be calculated by means of the well-known formula for the Fourier transform of the exponential of a quadratic form of a symmetric matrix [9]. The remaining integral over the variables τ_1 and τ_2 can be reduced by an obvious change of variables to the integral $\Phi(\alpha, \beta, \Delta)$:

$$\Phi(\alpha,\beta,\Delta)=\int_\alpha^\infty dt\, e^{it^2}\int_{\beta+\Delta(t-\alpha)}^\infty d\tau e^{i\tau^2},$$

in terms of which the asymptotic behavior of the integral $J_0(\lambda, E_1, E_2)$ can also be expressed.

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DYNAMICS OF AN ENSEMBLE OF SINGLE-DOMAIN MAGNETIC PARTICLES

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The dynamics of an ensemble of noninteracting single-domain magnetic particles is investigated both on the basis of analytic solution of the Fokker–Planck equation and in the framework of the reduced-description method. It is shown that in the general case the shape of the resonance and relaxation curves is not Lorentzian. In the isotropic case, the deviations from Lorentzian form reach 7%. In the presence of anisotropy, the main source of broadening of a resonance is thermal spread of the precession frequencies of the magnetic moments. An exact expression is obtained for the integral time of longitudinal relaxation of magnetic particles with axial anisotropy; it is valid for any value of the potential barrier. It is shown that for isotropic particles the description based on one and two lowest moments of the distribution function is in good agreement with the obtained exact results. In the first approximation of the moment method a generalized equation of Landau–Lifshitz–Bloch type is obtained; it gives a reduced description of the dynamics of the ensemble of magnetic particles in the general nonlinear case.

1. Introduction

Among the various magnetic materials, magnetic composites — magnetic polymers and magnetic liquids — occupy a special position. In them, particles of a ferromagnet (iron,

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magnetite, etc.) which contain macroscopic numbers of atoms, behave at temperatures $T \ll T_c$ as single magnetic moments μ that interact both with the nonmagnetic matrix and with one another through a dipole–dipole interaction. The contact of a magnetic particle with the matrix, which plays the part of a thermal reservoir, leads to significant complications in its dynamics, which at nonzero temperatures cannot be formulated in the framework of the notion of motion of the magnetization vector of the particle. A similar situation arises in the case of a quantum spin that interacts with a thermal reservoir, for which one must use a density matrix. However, the behavior of the ensemble of single-domain magnetic particles, which can be ascribed an effective spin $S \gg 1$, can be described by the classical analog of the equation for the density matrix — the Fokker–Planck (FP) equation.

The standard arguments that lead to the FP equation for an ensemble of magnetic particles reduce to the following. The characteristic frequencies ω_{char} of the problem have the order of the precession frequency of the magnetic moments in the magnetic field: $\omega_0 = g\mu_B H/\hbar$ (the frequency does not depend on the number N of atoms in the particle), or the still smaller relaxation frequency Γ. Further, a need to introduce a distribution function of the particles with respect to the directions of the magnetic moment arises at temperatures $k_BT \gtrsim \mu H = g\mu_B S_0 NH$, where S_0 is the effective spin per atom. The thermal reservoir acts on a magnetic particle through random fields, the spectrum of which is truncated at frequencies of order $\omega_{max} \sim k_BT/\hbar$. It is readily seen that $\omega_{max}/\omega_{char} \sim S_0 N \gg 1$, i.e., for "heavy" particles, containing a macroscopically large number of magnetic atoms, conditions that enable us to treat the thermal reservoir as a source of delta-correlated noise hold. In its turn, the stochastic equation of motion of the magnetic particle with coefficients of white-noise type can be reduced to an FP equation for the distribution function. At the same time, since the white noise in the given problem arises as the limiting case of a process with finite correlation time, the stochastic equation of motion for the magnetic particle must be given the Stratonovich interpretation. We note in passing that allowance for the finiteness of the correlation time of the random fields that act on the magnetic particle leads, when the most natural models of the thermal reservoir are chosen, to noise correlation functions that are nonexponential. For example, if it is assumed that the required random field is proportional to the elastic deformation u of the lattice at the position $\mathbf{r}$ of the particle, then the corresponding correlation function $f(t) \propto \langle u(\mathbf{r}, 0) u(\mathbf{r}, t)\rangle$ will, in the harmonic approximation for $T \ll \theta_D$, have the form $f(t) \propto 1/t^2 - \gamma_T^2/\sinh^2(\gamma_T t)$, where $\gamma_T = \pi k_B T/\hbar$. It is easy to see that with respect to functions that vary weakly over the time $1/\gamma_T$ the correlation function f(t) plays the part of a delta function: $f(t) \propto T\delta(t)$.

The fact that the behavior of even one magnetic particle is, when allowance is made for its interaction with the thermal reservoir, described by a partial differential equation that cannot, in general, be reduced to an equation for the mean magnetization makes the status of the usually employed phenomenological equations for many-particle magnetic systems, formulated in terms of macroscopic variables, into a nontrival problem. In particular, this applies to the Landau–Lifshitz equation for an ordinary ferromagnet, the validity of which at higher temperatures ($T \lesssim T_c$) is not obvious.

Although the basic physical ideas about the behavior of an ensemble of feromagnetic particles without allowance for their interaction with each other were already formulated by Néel, [1], a systematic investigation of the dynamical processes in such systems on the basis of the solution of the corresponding FP equation has not yet been made. At the same time, detailed study of the simplest model — isotropic particles in a magnetic field $\mathbf{H}$ for different values of $\xi = g\mu_B S_0 NH/k_BT$ — would make it possible to clarify the status of the usually employed approximations of the type of the moment method (see, for example, [2]), to estimate the deviations of the resonance and relaxation curves from Lorentzian form, and, finally, to pose the problem of a reduced description of the dynamics of the ensemble of magnetic particles in the general nonlinear case.

The exact results for the problem of the linear susceptibilities χ of an ensemble of magnetic particles obtained in this paper show that the deviation of χ from Lorentzian behavior does not exceed 7%; the deviations are maximal for static field corresponding to $\xi \sim 1$ and are small in the limiting cases $\xi \ll 1$ and $\xi \gg 1$. For the analytic description of χ in the complete range of fields and frequencies, it is customary to use the method of modeling the distribution function — the method of moments. It is well known that in the simplest form (in which only the first moment is retained) the results for χ have Lorentzian form (see, for example, [3]). The following approximation of the method makes it possible to determine the deviation of the line shape $\chi(\omega)$ from a Lorentzian curve. As is shown in this paper, the second approximation of the moment method gives a fairly good description of the deviations for the longitudinal susceptibility at $\xi \sim 1$ and, moreover, has asymptotic behaviors that agree with those of the exact solution of the FP equation in the different limiting cases.

The analysis of the linear susceptibilities provides a basis for writing down a closed nonlinear equation for the magnetization of the ensemble of ferromagnetic particles. This equation, which may be called the Landau–Lifshitz–Bloch (LLB) equation, describes both the transverse and longitudinal relaxation of the magnetization; moreover, the relaxation frequencies Γ_1 and Γ_2 that occur in the equation depend on the magnetization itself. In the region of low and high temperatures, the LLB equation goes over into the Landau–Lifshitz and Bloch equations, respectively. We note that in the nonlinear case the treatment of the higher approximations of the moment method is inconvenient from the computational point of view.

More realistic models of a magnetic particle also contain an anisotropy energy, both crystallographic and magnetostatic, with dependence on the shape of the particle. In particular, if the ferromagnetic particle is elongated along a certain axis, the smallest value of the magnetostatic energy is attained for orientation of the vector $\boldsymbol{\mu}$ along this axis. In many cases, the contribution of the magnetostatic energy to the anisotropy is predominant. The main physical manifestations of the anisotropy are already evident in the simplest case of easy-axis ferromagnetic particles. The first effect is due to the fact that the precession frequency of the magnetic moment around the z axis depends on its projection μ_z. At nonzero temperatures, this leads to a smearing of the resonance in the transverse susceptibility that is appreciably greater than the broadening that is associated with the true damping and is described in the framework of the moment method in [2]. In the presence of anisotropy, the evolution of a certain smooth initial distribution with $\langle\mu_\perp\rangle = 0$ leads to reversible mixing and to a loss of the smoothness property by the distribution function, and this, in turn, leads to difficulties in numerical solution of the FP equation by grid methods. We note also that the mixing which arises because of the nonlinearity strongly increases the importance of the true damping, since the large gradients of the distribution function which arise as a result of the mixing lead to enhanced diffusion. As a result of these processes, the distribution function tends rapidly to a form that depends only on the energy, this corresponding in quantum language to a diagonal density matrix.

Specific effects can also arise if the magnetic particle has a barrier, associated with the anisotropy, between energy minima, its height satisfying $\Delta E \gg k_B T$. Then the relaxation time for transitions through the barrier is exponentially long. Great efforts have been expended on the solution of this problem, beginning with [4], in which the time of transition through the barrier was related to the smallest eigenvalue of the Fokker–Planck operator (approximation of a "long-lived" exponential). In the general case $\Delta E \sim k_B T$, such an approach gives an order of magnitude estimate for the relaxation time, but to determine the shape of the relaxation curve the complete FP equation must be solved. On the other hand, a more informative quantity (and, moreover, one that can be compared with experiment) is the integral relaxation time, which is proportional to the area under the curve that describes the relaxation of the magnetization after an abrupt change of the longitudinal magnetic field. In the linear case (for small jump of the magnetic field), it is possible to obtain a general expression for the integral relaxation time of a magnetic particle for all values of the parameter $\Delta E/k_B T$.

2. Fokker–Planck Equation for Ensemble of Ferromagnetic Particles

To simplify the expressions, we shall in what follows use a system of units in which $g\mu_B S_0 N = k_B = \hbar = 1$. Then the equation of motion of the unit vector $\boldsymbol{\mu}$ of the ferromagnetic particle can be written in the form

$$\frac{d\boldsymbol{\mu}}{dt} = [\boldsymbol{\mu}, (\mathbf{H}_{eff} + \mathbf{H}_{fl})] - \mathbf{R}, \tag{1}$$

where $\mathbf{H}_{eff} = -\partial\mathcal{H}/\partial\boldsymbol{\mu}$, $\mathcal{H}$ is the energy of the particle with allowance for the external field and the anisotropy, $\mathbf{H}_{fl}$ is the fluctuation field exerted by the thermal reservoir, and $\mathbf{R}$ is a relaxation term, the form of which will be determined below. To be specific, we restrict ourselves to considering magnetic particles with axial anisotropy: $\mathscr{H}=-\mathbf{H}\boldsymbol{\mu}-D\mu_z^2$, where $\mathbf{H}$ is the external magnetic field, and D is the anisotropy constant. In this case, the fluctuation field $\mathbf{H}_{fl}$ must have the form

$$\mathbf{H}_{fl}=\boldsymbol{\zeta}(t)+\hat{\varkappa}(t)\boldsymbol{\mu},$$

where $\zeta(t)$ and $\hat{\varkappa}(t)$ are a stochastic vector and a stochastic second-rank tensor, their correlation functions being given by

$$\langle\zeta_i(t)\zeta_j(t')\rangle=2\lambda_{ij}T\delta(t-t'),\quad \langle\varkappa_{ik}(t)\varkappa_{jl}(t')\rangle=2\lambda_{ik,\,jl}T\delta(t-t'),\quad \langle\zeta_i(t)\varkappa_{jk}(t')\rangle=2\lambda_{i,\,jk}T\delta(t-t'). \tag{2}$$

Using the standard methods [5], we can pass from the stochastic equation (1) to the FP equation for the probability density $f(\mathbf{M},t)=\langle\delta(\mathbf{M}-\boldsymbol{\mu}(t))\rangle$, which has the form

$$\frac{\partial f}{\partial t}=-\frac{\partial}{\partial \mathbf{M}}\left\{[\mathbf{M},\mathbf{H}_{eff}]-\mathbf{R}+T\left[\mathbf{M},\hat{G}\left[\mathbf{M},\frac{\partial}{\partial\mathbf{M}}\right]\right]\right\}f, \tag{3}$$

where $G_{ij}=\lambda_{ij}+(\lambda_{i,\,jk}+\lambda_{j,\,ik})M_k+\lambda_{ik,\,jl}M_kM_l$.

The relaxation term $\mathbf{R}$ is now uniquely determined from the condition for the existence of the equilibrium Gibbs distribution $f_0 \propto \exp(-\mathscr{H}/T)$ and is equal to $\mathbf{R}=[\mathbf{M},\hat{G}[\mathbf{M},\mathbf{H}_{eff}]]$. It is readily seen that for arbitrary form of the tensor function $\hat{G}$ the relaxation term $\mathbf{R}$ does not reduce to the standard expression proposed by Landau and Lifshitz and can have a more complicated form. This can be expected if the anisotropy energy is appreciable. However, for the sake of simplicity, we shall in this paper restrict ourselves to considering the case of isotropic fluctuations, when in the expression (2)

$$\lambda_{ij}=\lambda_1\delta_{ij},\quad \lambda_{i,\,kl}=0;\quad \lambda_{ik,\,jl}=\lambda_2\delta_{ij}\delta_{kl}.$$

In this case, $G_{ij} = (\lambda_1 + \lambda_2)\delta_{ij}$, and the FP equation takes the standard form

$$\frac{\partial f}{\partial t}=-\frac{\partial}{\partial \mathbf{M}}\left\{[\mathbf{M},\mathbf{H}_{eff}]-\lambda\left[\mathbf{M},\left[\mathbf{M},\left(\mathbf{H}_{eff}-T\frac{\partial}{\partial\mathbf{M}}\right)\right]\right]\right\}f, \tag{4}$$

where $\lambda = \lambda_1 + \lambda_2$.

In what follows, we require equations for the first two moments of the distribution function f:

$$\frac{\partial}{\partial t}\langle\boldsymbol{\mu}\rangle=\langle[\boldsymbol{\mu}\mathbf{H}_{eff}]\rangle-\lambda\{2T\langle\boldsymbol{\mu}\rangle+\langle[\boldsymbol{\mu}[\boldsymbol{\mu}\mathbf{H}_{eff}]]\rangle\}, \tag{5}$$

$$\frac{\partial}{\partial t}\langle\mu_i\mu_j\rangle=\langle[\boldsymbol{\mu},\mathbf{H}_{eff}]_i\mu_j\rangle-\lambda\{\langle[\boldsymbol{\mu}[\boldsymbol{\mu},\mathbf{H}_{eff}]]_i\mu_j\rangle+3T(\langle\mu_i\mu_j\rangle-\delta_{ij}/3)\}+i\leftrightarrow j. \tag{6}$$

Note that in the isotropic case ($\mathbf{H}_{eff}=\mathbf{H}$) and in the region of high temperatures ($T \gg H$), Eq. (5) becomes independent of the remaining moment equations. In the resulting equation for the magnetization $\mathbf{m}=\langle\boldsymbol{\mu}\rangle$, the relaxation term has isotropic Bloch form, which is a radical simplification and leads to Lorentzian expressions for the susceptibilities. However, in the presence of anisotropy, when $\mathbf{H}_{eff}=\mathbf{H}_{eff}(\boldsymbol{\mu})$, such simplifications arise only when the much stronger condition $\lambda T \gg H_{eff}$, which leads to vanishing of the transverse resonance, holds. At temperatures $H_{eff} \lesssim T \lesssim H_{eff}/\lambda$ there is no closed equation for the magnetization, and the shape of the transverse resonance line is determined by the mixing effect mentioned in the Introduction.

3. Linear Susceptibilities of the Ensemble of Magnetic Particles: Exact Results

We consider the case when the constant magnetic field is directed along the anisotropy axis, i.e., the energy of the magnetic particle has the form $\mathscr{H}=-H\mu_z-D\mu_z^2$. Then the mean static susceptibility of the particle is determined by the expression

$$m_z\equiv\langle\mu_z\rangle=\int_{-1}^{1}xf_0(x)\,dx=\frac{d}{d\xi}\ln Z=B(\xi,\alpha),$$

where

$$f_0=\frac{1}{Z}\exp(-\mathscr{H}/T),\quad Z=\int_{-1}^{1}\exp(-\mathscr{H}/T)\,dx$$

and we have introduced the notation $\xi=H/T$, $\alpha=D/T$, $x=M_z$.

In the presence of a small alternating longitudinal field, $h_z = h_{z0}\exp(-i\omega t)$, there is a small deviation of the distribution function from the equilibrium function,

$$\delta f=(h_z/T)f_0(x)q(x),$$

where the function q(x) satisfies an equation that follows from Eq. (4),

$$\left(\frac{d}{dx}+\xi+2\alpha x\right)(1-x^2)\frac{dq}{dx}+\frac{i}{\lambda}\tilde{\omega}q=(1-x^2)(\xi+2\alpha x)-2x, \tag{7}$$

where $\tilde{\omega} = \omega/T$. The ensemble longitudinal susceptibility $\chi_\parallel$ is determined from the function q(x) by the expression

$$\chi_\parallel=\frac{1}{T}\int_{-1}^{1} f_0(x)q(x)x\,dx.$$

Analytic solution of Eq. (7) is possible only in various limiting cases. The most interesting case is expansion of q(x) with respect to low frequencies, corresponding to a representation of the susceptibility in the form

$$\chi_\parallel=\chi_{0\parallel}(1+i\omega\tau+\ldots), \tag{8}$$

where $\chi_{0\parallel}=B'/T$ and $B'=\partial B(\xi,\alpha)/\partial\xi$. Using linear response theory, we can readily show that τ in (8) is none other than the integral relaxation time determined by the area under the curve that describes the relaxation of the magnetization after an abrupt change of the longitudinal magnetic field:

$$\tau=\int_0^\infty \delta m_z(t)\,dt/\delta m_z(0),$$

where $\delta m_z(t) = m_z(t) - m_z(\infty)$. The finding of the terms of the series (8) is facilitated by the lowering of the order of Eq. (7) when the expansion with respect to the low frequencies is made; the value of the integral relaxation time τ is given by

$$\tau=\frac{2}{\gamma B'}\int_{-1}^{1}\frac{dx}{1-x^2}\Phi^2(x)/f_0(x), \tag{9}$$

where $\gamma = 2\lambda T$ and $\Phi(x)=\int_{-1}^{x}(x'-B)f_0(x')\,dx'$.

The form of the function $\Phi(x)$ simplifies in two special cases. For zero external field ($\xi = 0$)

$$\Phi(x)=f_0(x)\{1-\exp[\alpha(1-x^2)]\}/(2\alpha).$$

In the isotropic case ($\alpha = 0$)

$$\Phi(x)=f_0(x)\{\operatorname{cth}\xi-x-\exp(-\xi x)/\operatorname{sh}\xi\}/\xi.$$

We give the most important asymptotic behaviors of the integral relaxation time τ:

$$\frac{1}{\tau}=\begin{cases}\frac{2}{\sqrt{\pi}}\gamma\alpha^{3/2}e^{-\alpha}, & \xi=0,\quad \alpha\gg1,\\ \gamma|\alpha|, & \xi=0,\quad \alpha<0,\quad |\alpha|\gg1,\\ \gamma\xi, & \alpha=0,\quad \xi\gg1,\\ \gamma\left(1-\frac{2}{5}\alpha+\frac{2}{35}\alpha^2+\frac{1}{9}\xi^2\right), & \xi,\ |\alpha|\ll1.\end{cases}$$

The first of these, corresponding to the case of a high-energy barrier, agrees with the smallest eigenvalue Λ_1 of the Fokker–Planck operator calculated by Brown [4] in the same limit. This is no surprise, since for a high barrier the condition $\Lambda_1 \ll \Lambda_n$ ($n \geq 2$) holds. In this case, the magnetization relaxes in accordance with a "pure" exponential with $\tau^{-1} = \Lambda_1$. The second and third asymptotic behaviors correspond to the absence of an energy barrier and to low temperatures. In these cases, τ^{-1} does not depend on the temperature and has the order of max(λH, λD). The last asymptotic behavior describes the corrections to the high-temperature Bloch behavior. We note the numerical proximity of this result to the high-temperature expansion of Λ_1 [4], in which coefficients α^2 and ξ^2 are equal to 48/875 and 1/10, respectively. For arbitrary values of the parameters of the problem, the use of the integral relaxation time τ is preferable to the use of Λ_1, since, on the one hand, there exists for τ the general analytic expression (9), which can be readily extended to arbitrary form of the axial anisotropy of the magnetic particle, and, on the other, τ is directly related to an experimentally observable quantity — the area under the relaxation curve.

The calculation of the following terms of the low-frequency expansion of the susceptibility $\chi_\parallel$ (Eq. (8)) would be a too laborious task. On the other hand, the high-frequency expansion of $\chi_\parallel$ is straightforward and can be carried through to high orders, giving information about the shape of the line of relaxation absorption. For example, for the case D = 0 we obtain

$$\chi_\parallel=\chi_{0\parallel}[\Gamma_{11}/-i\omega+(\Gamma_{12}/-i\omega)^2+(\Gamma_{13}/-i\omega)^3+\ldots], \tag{10}$$

where

$$\Gamma_{11}=\gamma B/\xi B',\quad \Gamma_{12}=\gamma[(\xi-2B)/\xi B']^{1/2},\quad \Gamma_{13}=\gamma[(B\xi^2+7B-2\xi)/\xi B']^{1/3},\quad B=\operatorname{cth}\xi-1/\xi. \tag{11}$$

The values of Γ_{1i} are close to each other and coincide in limiting cases: $\Gamma_{1i}\cong\gamma$ for $\xi\ll 1$ and $\Gamma_{1i}\cong\gamma\xi$ for $\xi\gg 1$. The relative deviations $\delta=|1-\Gamma_{1i}/\Gamma_{11}|$ reach 0.06 for $\xi\simeq 2.5$, indicating a nearly Lorentzian line shape.

We also give expressions for the longitudinal susceptibility $\chi_\parallel$ in the complete region of frequencies for D = 0 in the limits $H\ll T$ and $H\gg T$. In the first case ($\xi\ll 1$), the solution of Eq. (7) leads to the result

$$\chi_\parallel=\frac{1}{3T}\frac{\gamma}{\gamma-i\omega}\left[1-\frac{2}{15}\xi^2\frac{i\omega((5/2)\gamma-i\omega)}{(\gamma-i\omega)(3\gamma-i\omega)}\right]. \tag{12}$$

In the opposite limit ($\xi\gg 1$), the calculations give

$$\chi_\parallel=\chi_{0\parallel}\frac{\Gamma}{\Gamma-i\omega}\left[1+\frac{1}{\xi}\frac{i\omega}{\Gamma-i\omega}+\frac{2}{\xi^2}\frac{i\omega\Gamma(^3/_2\Gamma-i\omega)}{(\Gamma-i\omega)^2(2\Gamma-i\omega)}\right], \tag{13}$$

where $\Gamma=\gamma\xi$. Analysis of the expressions (8)–(13) makes it possible to elucidate the nature of the deviation, obtained in the following section, of the longitudinal susceptibility of the ensemble of single-domain magnetic particles from the Lorentz expression.

We now turn to investigation of the transverse susceptibility of an ensemble of magnetic particles. In the case of a circular alternating field, $h_x+ih_y=h_0\exp(-i\omega t)$, the correction to the equilibrium distribution function can be expressed in the form

$$\delta f=(h_0/T)f_0(x)[p_1(x)\sin(\varphi-\omega t)+p_2(x)\cos(\varphi-\omega t)],$$

where φ is the azimuthal angle of the vector μ. The linear combination $g(x)=p_1(x)-ip_2(x)$ satisfies the differential equation

$$\left(\frac{d}{dx}+\xi+2\alpha x\right)(1-x^2)\frac{dg}{dx}+\frac{i}{\lambda}(\xi+2\alpha x-\tilde\omega)g=-\lambda^{-1}\sqrt{1-x^2}[2\lambda+\lambda x(\xi+2\alpha x)-i(\xi+2\alpha x)], \tag{14}$$

which follows from the basic equation (4). The complex susceptibility $\chi_\perp$ is determined by the formula

$$\chi_\perp=\frac{1}{2T}\int_{-1}^{1}\sqrt{1-x^2}f_0(x)g(x)\,dx.$$

For a system of isotropic magnetic particles ($\alpha=0$), analytic solution of Eq. (14) is possible only in two limiting cases. Far from resonance, $\chi_\perp$ can be expressed in the form of the expansion

$$\chi_\perp=\chi_{0\perp}\frac{H}{H-\omega}\left\{1+i\frac{\Gamma_{21}}{H-\omega}\frac{\omega}{H}+\left(\frac{\Gamma_{22}}{H-\omega}\right)^2\frac{\omega}{H}+\ldots\right\}, \tag{15}$$

where $\chi_{0\perp}=B(\xi)/H$ and

$$\Gamma_{21}=(\gamma/2)(\xi/B-1),\quad \Gamma_{22}=(\gamma/2)\sqrt{\xi^2+4}. \tag{16}$$

The quantities Γ_{21} and Γ_{22} have equal limits ($\Gamma_{2i}\cong\gamma$ for $\xi\ll 1$ and $\Gamma_{2i}\cong\gamma\xi/2$ for $\xi\gg 1$), and their relative deviation $\delta=|1-\Gamma_{22}/\Gamma_{21}|$ reaches 0.04 when $\xi\approx 3.0$, indicating that

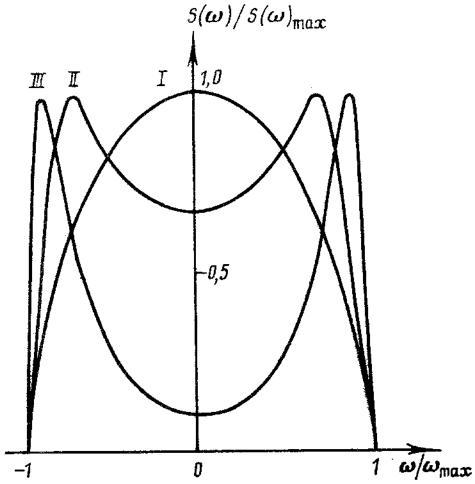

Fig. 1

the shape of the transverse resonance line is nearly Lorentzian.

We also give the high-temperature expansion of $\chi_\perp$:

$$\chi_\perp=\frac{1}{3T}\frac{H-i\gamma}{H-\omega-i\gamma}\left\{1-\frac{\xi^2}{15}\left[1+\frac{3}{2}\frac{\omega\gamma(H-\omega-5i\gamma/2)}{(H-i\gamma)(H-\omega-i\gamma)(H-\omega-3i\gamma)}\right]\right\}. \tag{17}$$

As noted in the Introduction, the presence of anisotropy gives rise to a new source of broadening of the transverse resonance line associated with the spread of the precession frequencies of the magnetic moments in the effective anisotropy field. Since this effect, which to a certain degree is analogous to inhomogeneous broadening, appreciably exceeds the true damping in real cases, to solve Eq. (14) we can set $\lambda = 0$, after which the result can be written down directly for the circular susceptibility:

$$\chi_\perp=\frac{1}{2T}\int_{-1}^{1}(1-x^2)f_0(x)\,\frac{\xi+2\alpha x}{\xi+2\alpha x-\tilde{\omega}-i0}dx.$$

The final expression for $\chi_\perp''$ has the form

$$\chi_\perp''=\frac{\pi}{2T}\frac{\tilde{\omega}}{(2|\alpha|)^3}\frac{1}{Z}\{(2\alpha)^2-(\tilde{\omega}-\xi)^2\}\exp\left(\frac{\tilde{\omega}^2-\xi^2}{4\alpha}\right).$$

The values of $\chi_\perp''$ are nonzero in the interval $(\tilde{\omega}-\xi)^2 \leqq (2\alpha)^2$; at low temperatures, the spectral density of the fluctuations $S(\omega) = \chi_\perp''/\omega$ degenerates into narrow peaks. For the easy-axis model ($\alpha > 0$), these peaks are at the boundaries of the interval and have different magnitudes when $\xi \neq 0$. In the case of easy-plane anisotropy ($\alpha < 0$), there is one peak (at $\xi = 0$ with Gaussian shape, centered at zero frequency). As the temperature is raised, the peaks spread, and in the limit $T \to \infty$ the spectral density of the fluctuations in both models is determined by the purely geometrical factor $(2\alpha)^2 - (\tilde{\omega} - \xi)^2$. The behavior of $S(\omega)$ for the easy-axis model with $\xi = 0$ is shown schematically in Fig. 1 for different temperatures. The two maxima in the curve of $S(\omega)$ merge into a single maximum for $\alpha = 1$.

4. Linear Susceptibilities of the Ensemble of Magnetic Particles: The Moment Method

Our investigation has shown that for a system of isotropic magnetic particles the deviations of the linear susceptibilities from Lorentzian form are not too large. This suggests the possibility of constructing approximate solutions of the Fokker–Planck equation in closed form for all parameter values of the problem. To this end, one can use the well-known moment method (see, for example, [2,3]), which consists of parametrizing the distribution function by means of a certain number of variables in such a way as to satisfy the same number of lowest moment equations of the type (5), (6). In contrast to the well-known Galerkin method, we shall find it more convenient to represent the distribution function f as the exponential of some polynomial in powers of the components of the vector **M** (see (3))

$$f(\mathbf{M},t)=Z^{-1}\exp(\xi(t)\mathbf{M}+\mathbf{M}\hat{\alpha}(t)\mathbf{M}+\ldots). \quad (18)$$

For isotropic particles and small deviations from equilibrium $\xi(t)=\xi_0+\delta\xi(t)$, where $\xi_0=H/T$ and $\delta\xi\ll\xi_0$ (in what follows, we shall generally omit the index in the symbol for ξ_0). The remaining parameters of the distribution function (18), including the tensor $\alpha(t)$, must also be small under the considered conditions compared with ξ_0.

In the first approximation of the moment method, we retain just the single parameter $\xi(t)$, whose dynamics is determined by Eq. (5). For the longitudinal and circular transverse susceptibilities, we obtain the expressions

$$\chi_\parallel=\chi_{0\parallel}\frac{\Gamma_1}{\Gamma_1-i\omega} \quad (19)$$

and

$$\chi_\perp=\chi_{0\perp}\frac{H-i\Gamma_2}{H-\omega-i\Gamma_2}, \quad (20)$$

where the longitudinal and transverse relaxation rates Γ_1 and Γ_2 are equal to the previously introduced Γ_{11} and Γ_{21} (see the expressions (11) and (16)). The expressions (19) and (20) are the Lorentzian expressions with which we must compare the exact results obtained in the previous section.

In the second approximation of the method of moments, we must retain in the expression (18) two parameters: ξ and $\hat{\alpha}$, the equations of motion for which follow from Eqs. (5) and (6). The result obtained for the longitudinal susceptibility has the form

$$\chi_\parallel=\chi_{0\parallel}\frac{\Gamma_1'\Gamma_1''-i\omega\Gamma_1}{(\Gamma_1'-i\omega)(\Gamma_1''-i\omega)}. \quad (21)$$

The corresponding calculations and expressions for Γ_1' and Γ_2'' are given in the Appendix. Since Γ_1' is numerically close to Γ_1, the expression (21) is close to a Lorentzian curve. It can be shown that the expansion of (21) with respect to high frequencies reproduces the first two terms of the exact expression (10). Moreover, the expansions of (21) also agree, at small ξ to accuracy ξ^2 and large ξ to accuracy ξ^{-2}, with the exact results (12) and (13). The expansion (21) at low frequencies analogous to the expansion (8) makes it possible to determine the integral relaxation time in the second approximation of the moment method:

$$\tau_2^{-1}=\frac{\Gamma_1'\Gamma_1''}{\Gamma_1'+\Gamma_1''-\Gamma_1}. \quad (22)$$

Comparing the expression (22) with the exact result (9), and also with $\tau_1^{-1}=\Gamma_1$ obtained in the first approximation of the moment method, we can estimate the convergence of the method for arbitrary values of the parameter $\xi = H/T$. Figure 2 shows the dependences of the relative deviations $\delta = \tau_i/\tau_1 - 1$ of the integral relaxation time from the Lorentzian result $\tau_1 = \Gamma_1^{-1}$ as a function of the parameter ξ. As can be seen from the graph, the largest deviations from the exact result (to which the upper curve corresponds) are attained when $\xi \simeq 3$. These deviations are equal to 0.07 and 0.007, respectively, for the first and second approximations of the moment method. Thus, the second approximation of the moment method almost completely describes the deviation of $\chi_\parallel(\omega)$ from the Lorentzian expression. The graph of the spectral density of the fluctuations $S(\omega)$ is everywhere above the Lorentzian curve, and the deviation is maximal at $\omega = 0$ and disappears as $\omega \to \infty$. With regard to the real part of the longitudinal susceptibility, for $\omega = 0$ all the approximations give the exact result $\chi_{0\parallel}$, while for $\omega \neq 0$ the exact solution lies above the Lorentzian result. This can be seen, in particular, from the expansion (10), in which $\Gamma_{12}>\Gamma_{11}$.

We could consider similarly the transverse susceptibility $\chi_\perp$ in higher approximations of the moment method. However, the calculations in this case are much more cumbersome compared with the longitudinal case because of the need to retain in the expression (18) various cost components of the tensor $\hat{\alpha}$.

The moment method cannot be used to describe the dynamics of a system of easy-axis

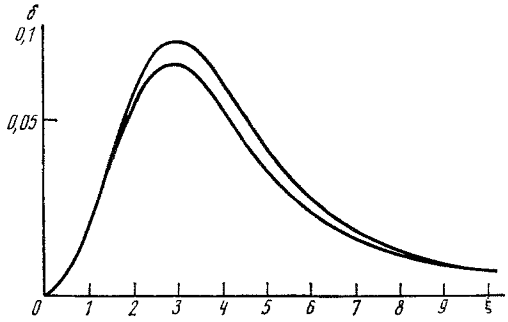

Fig. 2

magnetic particles if the energy barrier is high: $\Delta E \gg T$. The calculation shows that by means of the first few approximations of the moment method it is not possible to describe the exponentially long relaxation times characteristic of this case. The question of the calculation of the transverse susceptibility in the presence of anisotropy in the framework of the moment method does not even arise, since we have already obtained a solution that is exact in the limit of weak damping. This solution does not correspond to the one found in [3] by means of the moment method.

5. Nonlinear Dynamical Processes in a System of Ferromagnetic Particles. The Landau–Lifshitz–Bloch Equation

Our analysis in the previous sections of the linear susceptibilities of an ensemble of single-domain magnetic particles has shown that in the isotropic case the successive approximations of the moment method give rapid convergence to the exact results for all values of the parameter $\xi = H/T$. With regard to the more complicated nonlinear problems, it is meaningful, accepting a 5–7% accuracy, to restrict ourselves to the first approximation of the moment method. Besides the sharp increase in the number of variables, the retention in the expression (18) of the quantities $\alpha_{ij}(t)$, etc., leads to a further difficulty associated with the necessity for numerical calculations to find the various moments of the distribution function. This reduces to nothing the advantages of all the approximations of the moment method beginning with second as compared with numerical solution of the original Fokker–Planck equation (3).

Thus, restricting ourselves to the first approximation of the moment method, we retain in the expression (18) only terms that contain the vector parameter $\boldsymbol{\xi}$ ("thermodynamic field"), and we use the chosen distribution function to calculate the left- and right-hand sides of the equation of motion for the magnetization (5). As a result, we arrive at a differential equation for the parameter $\boldsymbol{\xi}(t)$:

$$\frac{d\boldsymbol{\xi}}{dt} = [\boldsymbol{\xi}\mathbf{H}] - \Gamma_1\{1-(\boldsymbol{\xi}\boldsymbol{\xi}_0)/\xi^2\}\boldsymbol{\xi} - \Gamma_2[\boldsymbol{\xi}[\boldsymbol{\xi}\boldsymbol{\xi}_0]]/\xi^2, \quad (23)$$

in which the longitudinal and transverse relaxation coefficients depend on the unknown variable ξ:

$$\Gamma_1 = 2\lambda TB(\xi)/\xi B'(\xi), \quad \Gamma_2 = \lambda T(\xi/B(\xi)-1).$$

We recall that $\boldsymbol{\xi}_0 = \mathbf{H}/T$. From the solution of Eq. (23) we can calculate the mean magnetization of a particle:

$$\mathbf{m} = B(\xi)\boldsymbol{\xi}/\xi. \quad (24)$$

Of course, Eq. (23) admits reformulation in terms of the magnetization itself, but the corresponding equation is inconvenient for practical calculations, since it contains a function that is the inverse of the Langevin function: $\xi(m)$.

One further form of expression of Eq. (23) is

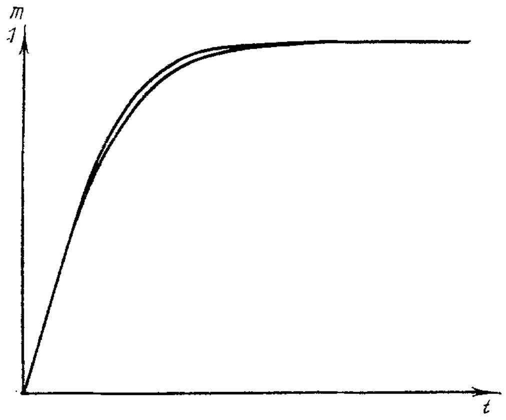

Fig. 3

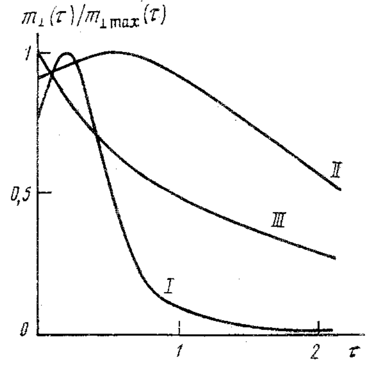

Fig. 4

$$\frac{d\boldsymbol{\xi}}{dt}=[\boldsymbol{\xi}\mathbf{H}]-\Gamma_1(\boldsymbol{\xi}-\boldsymbol{\xi}_0)-(\Gamma_2-\Gamma_1)[\boldsymbol{\xi}[\boldsymbol{\xi}\boldsymbol{\xi}_0]]/\xi^2. \quad (25)$$

It can be seen from this that at high temperatures, when ξ, $\xi_0 \ll 1$ and the coefficients Γ_1 and Γ_2 become equal ($\Gamma_1 \cong \Gamma_2 \cong \gamma = 2\lambda T$), Eq. (25) goes over into the Bloch equation. On the other hand, at low temperatures, when ξ, $\xi_0 \gg 1$, the dependence of the magnitude of the vector $\mathbf{m}$ on ξ disappears (saturation). At the same time, the term in Eq. (23) that describes the relaxation of the vector $\boldsymbol{\xi}$ in magnitude ceases to play a part, and we return to the Landau–Lifshitz equation. For parameter values ξ, $\xi_0 \sim 1$ Eq. (23) describes an intermediate situation and can be called the LLB equation.

The proposed method can be used to investigate, for example, a dynamical problem for a system of magnetic particles such as that of the relaxation from some initial state. We first demonstrate the possibility of such a description by means of the following example, which admits an exact analytic solution. Suppose that a system of magnetic particles at zero temperature is in a zero magnetic field, and that the initial distribution function is isotropic. After instantaneous switching on of a static magnetic field $\mathbf{H}$ the magnetization grows with the time in accordance with the law

$$m(t)=\operatorname{cth}\tilde{t}\,(1-2\tilde{t}/\operatorname{sh}2\tilde{t}),$$

where $\tilde{t} = \lambda Ht$. This same quantity can be calculated approximately by numerical solution of Eq. (23). As can be seen from Fig. 3, two curves run close to each other (the exact curve is the lower one), and the integral relaxation times corresponding to them are $1/\lambda H$ and $0.936/\lambda H$. As in the linear case, the difference between these quantities does not exceed 7%.

Having shown that Eq. (23) describes satisfactorily the nonlinear relaxation of the system of isotropic magnetic particles, we illustrate its use by the example of the problem of the switching of the external magnetic field through an angle θ at an arbitrary temperature. In this case, the LLB equation for the vector $\boldsymbol{\xi}$ can be conveniently expressed in a coordinate system that rotates with frequency $\omega = H$ around the axis $\mathbf{z}\|\mathbf{H}$. Its numerical solution permits determination of the time dependence of the magnetization of the system. For $\theta > \pi/2$, the behavior of the transverse component of the magnetization of the system, $m_\perp(t)$, is very different in the regions of high ($\xi \ll 1$) and low ($\xi \gg 1$) temperatures. In the first case, we have an exponential decrease of $m_\perp(t)$, which corresponds to the Bloch equation. For $\xi \gg 1$, we obtain a result that agrees with the Landau–Lifshitz equation, i.e., the vector $\mathbf{m}$ is rotated, and its projection $m_\perp$ passes through a maximum. With increasing temperature, this maximum gradually decreases and at $\xi \sim 1$ disappears. The behavior of $m_\perp(t)$ for different temperatures is shown in Fig. 4 (ξ = 50, 10, 0.1; curves I, II, III, respectively). Note that these results could have been predicted on the basis of qualitative considerations.

Thus, the proposed equation (23) can be used to describe, with a satisfactory degree of accuracy, various nonlinear dynamical processes in systems of noninteracting magnetic particles. Hitherto, these questions have been investigated only in various limiting cases (see, for example, [6]).

The LLB equation is also valid for description of the dynamics of a system of mutually interacting magnetic particles in situations when the static properties of the system can be satisfactorily described by mean-field theory. To take into account the interaction in the LLB equation, it is necessary to replace the external magnetic field by a self-consistent field, which depends on the magnetizations of the particles that are the neighbors of the considered particle. If the magnetization varies weakly over the mean interparticle separation, the LLB equation reduces to a differential equation with respect to the spatial variables. Such an equation admits a parallel with the well-known Landau-Lifshitz equation for an ordinary ferromagnet and indicates a way in which this last can be generalized to the region of higher temperatures $T \lesssim T_C$.

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Appendix

In the second approximation of the moment method, construction of the function $f(\mathbf{M})$ requires us to retain the parameters ξ and $\bar{\alpha}$. In the case of the linear longitudinal susceptibility, there are two time-dependent variables: $\delta\xi_z(t)=\xi e^{-i\omega t}$ and $\alpha_{zz}(t)=\alpha e^{-i\omega t}$, which are chosen in such a way as to satisfy the linearized equations for $\langle\mu_z\rangle$ and $\langle\mu_z^2\rangle$. As a result, for ξ and α we obtain a system of two algebraic equations:

$$\xi(\Gamma_1-i\omega)+A_1\alpha(\Gamma_3-i\omega)=h_0\Gamma_1/T, \quad \xi(\Gamma_3-i\omega)+A_2\alpha(\Gamma_4-i\omega)=h_0\Gamma_3/T,$$

where

$$\Gamma_3=\frac{\gamma(\xi_0-3B)}{B-\xi_0B'}, \quad \Gamma_4=\gamma\frac{4(3B-\xi_0)+B\xi_0^2}{\xi_0(1+B')-4B}, \quad A_1=\frac{6(B-\xi_0B')}{B'\xi_0^2}, \quad A_2=\frac{6(\xi_0(1+B')-4B)}{\xi_0(B-\xi_0B')}.$$

The longitudinal susceptibility $\chi_\parallel$ can be expressed in terms of the parameters ξ and α as follows:

$$\chi_\parallel=\frac{1}{T}\frac{1}{h_0}B'[\xi+A_1\alpha].$$

From this it is possible to obtain the expression (21), in which the relaxation parameters are determined by the expression

$$\Gamma_1^{\prime(\prime\prime)}=\frac{1}{2(1-R)}[\Gamma_1+\Gamma_4-2R\Gamma_3\mp\sqrt{(\Gamma_4-\Gamma_1)^2+4R(\Gamma_4-\Gamma_3)(\Gamma_1-\Gamma_3)}],$$

where $R = A_1/A_2$.

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