Theory of dispersion of the magnetic susceptibility of fine ferromagnetic particles

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The motion of the magnetic moment is considered for a uniformly magnetized single-domain particle in a periodic field parallel or perpendicular to the axis of easy magnetization. It is shown that the character of the dispersion of the magnetic susceptibility depends substantially on the ratio $\sigma = KV/kT$ of the magnetic anisotropy energy to the thermal energy. With decrease of the parameter σ, there is an increase of the probability of thermal fluctuations in the direction of the magnetic moment of a particle (an analog of Brownian rotation), and this leads to broadening of the absorption line and decrease of the resonance frequency. A critical value $\sigma = \sigma_*$ is found, at which the characteristic frequency of precession of the magnetic moment in the anisotropy field vanishes. Near σ_* the characteristic frequency is $\omega_0[2(\sigma-\sigma_*)/5\sigma_*]^{1/2}$, where ω_0 is its asymptotic value as $\sigma\to\infty$. It is shown that the resonance (Lorentz) lines corresponding to large σ are continuously transformed, with decrease of this parameter, to purely relaxational (Debye) lines.

1. INTRODUCTION

In the classical paper of Landau and Lifshitz[1], which initiated the modern theory of ferromagnetic resonance, one topic considered in particular was the motion of the magnetic moment μ of a uniformly magnetized domain in the internal anisotropy field and an external radiofrequency field. If $\mu = MV$, where M is the magnetic-moment density and is equal to the saturation magnetization and where V is the domain volume, the Landau-Lifshitz equation can be written in the form

$$\dot{\boldsymbol{\mu}} = -\gamma[\boldsymbol{\mu}\mathbf{H}_e] - \alpha\gamma\mu^{-1}[\boldsymbol{\mu}[\boldsymbol{\mu}\mathbf{H}_e]]. \qquad (1)$$

Here γ is the gyromagnetic ratio for electrons; α is a dimensionless damping constant; H_e is the effective field, equal to $-\partial U/\partial\mu$; and U is the magnetic energy of the domain. For a uniaxial ferromagnetic crystal

$$U = -\mu H\mathbf{eh} - KV(\mathbf{en})^2, \quad \mathbf{h}=\mathbf{H}/H, \quad \mathbf{e}=\boldsymbol{\mu}/\mu, \qquad (2)$$

where K is the constant of effective magnetic anisotropy, n is a unit vector in the direction of the axis of easiest magnetization, and H is the applied radiofrequency field.

For H = 0, Eq. (1) describes the free precession of the vector μ in the anisotropy field $H_A = 2K/M$ with characteristic frequency $\omega_0 = \gamma H_A$ and relaxation time

$$\tau_0 = (\alpha\omega_0)^{-1} = M/2\alpha\gamma K. \qquad (3)$$

In a periodic field H perpendicular to H_A, the motion of the magnetic moment has a typically resonant character; the real and imaginary parts of the magnetic susceptibility have a so-called Lorentz form; the dimensionless (measured in fractions of ω_0) width of these lines along the frequency scale is of order α.

The goal of the present paper is to solve the following problem: how does the dimension of the particle affect the character of the motion of its magnetic moment? The dimensional effects discussed in the article should show up in very fine (~100 Å) particles, the interactions between which are here neglected. An adequate physical example is a dilute colloidal suspension of ferromagnetic particles in any nonmagnetic matrix (whether solid or liquid is immaterial, since the characteristic frequencies are so high that motion of the magnetic moment with respect to the body of the particle cannot excite any appreciable motion of the particle with respect to the liquid[2]).

In the range of dimensions in which the magnetic-anisotropy energy KV is comparable with the thermal energy kT, a fluctuational mechanism of reorientation of the vector μ becomes important. The motion of the magnetic moment of a single-domain particle under the influence of thermal fluctuations (first pointed out by Néel[3]) is analogous to the Brownian rotation of a particle in a liquid and can be described by a Fokker-Planck equation[4]. Here the Landau-Lifshitz equation (1) plays the role of a dynamic equation describing the regular change of the vector μ. The coefficient $\alpha\gamma/\mu$ before the relaxational term in (1) has the meaning of a rotational mobility of the magnetic moment, so that for the rotational diffusion coefficient in Einstein's formula one obtains $D = \alpha\gamma kT/\mu$. On comparing the characteristic time of orientational diffusion of the magnetic moment.

$$\tau = (2D)^{-1} = MV/2\alpha\gamma kT \qquad (4)$$

with the time of rotation of a Brownian particle in a viscous liquid $\tau_B = 3\eta V/kT$, we conclude that the role of the viscosity η in the mechanism of magnetic diffusion is played by the quantity $M/6\alpha\gamma$. We note that between τ of (4) and τ_0 of (3) there is the simple relation

$$\tau = \sigma\tau_0, \quad \sigma = KV/kT. \qquad (5)$$

We point out also the analogy between the precession of the magnetic moment in the anisotropy field and the cyclotron rotation of charged particles of a plasma in a magnetic field. In order that it may be possible to speak of precession at all, it is obviously necessary that its period be small in comparison with the rotational diffusion time (the latter here plays the same role as does the free-passage time of the particles of a plasma). In other words, over a time τ the vector μ "forgets" about its precession produced by the magnetic torque. By (3) and (5) the condition for existence of precession $\omega_0\tau \gg 1$ reduces to $\sigma \gg \alpha$; that is, it is satisfied only for sufficiently coarse particles, $V \gg \alpha kT/K$.

In Sec. 2, the Fokker-Planck equation for the probability W of orientations of the particle's magnetic moment is derived, and its spectral properties are analyzed. The eigenvalues Λ of this equation determine the frequencies ImΛ and damping decrements ReΛ of the normal modes, by superposition of which one can describe an arbitrary deviation of W from the equilibrium distribution W_0. For small σ there are no characteristic frequencies (all Λ's are real), so that in the absence of an alternating field the vector μ relaxes monotonically into the direction of H_A. Complex Λ's occur at values of σ larger than a certain critical σ_*, near which $\mathrm{Im}\Lambda$ is proportional to the square root of the difference $\sigma-\sigma_*$.

Results are presented of numerical (Galerkin's method, carried out by computer) and analytic (the method of moments) investigation of the function $\Lambda(\sigma)$. Values of

$$\mathrm{Im}\,\Lambda=\omega_0,\quad \mathrm{Re}\,\Lambda=\alpha\omega_0, \tag{6}$$

determined by the Landau-Lifshitz equation, are obtained for the corresponding levels of the spectrum of Λ in the low-temperature limit $\sigma\to\infty$, when the probability of thermal fluctuations in the directions of the magnetic moment μ approaches zero.

In Sec. 3 the magnetic susceptibility χ is found, in a periodic field parallel ($\chi_\parallel$) or perpendicular ($\chi_\perp$) to the axis of easy magnetization. It is shown how the resonance (lorentz) lines of $\chi_\perp$[1], obtained at $\sigma\gg 1$, become deformed with diminution of σ and in continuous fashion are transformed at $\sigma\ll 1$ to purely relaxational (Debye) lines. The dependence of the resonance frequency and of the width of the absorption line on σ is determined.

2. SPECTRAL PROPERTIES OF THE FOKKER-PLANCK EQUATION

The probability $W(\mathbf{e}, t)$ of various orientations of the magnetic moment $\boldsymbol{\mu}=\mu\mathbf{e}$ of the particle must satisfy the conservation law

$$\frac{\partial W}{\partial t}+\mathrm{div}(\mathbf{v}W)=0, \tag{7}$$

where $\mathbf{v}$ is the velocity of motion of the tip of the vector $\mathbf{e}$ along the surface of a sphere of unit radius. This velocity is defined as

$$\mathbf{v}=\frac{d\mathbf{e}}{dt}=\frac{1}{\mu}\frac{d\boldsymbol{\mu}}{dt}$$

and is made up of two parts—a regular part $\mathbf{v}_r$ and a stochastic (Brownian) part $\mathbf{v}_s$. The first of these, according to (1) and (2), is

$$\begin{gathered}\mathbf{v}_r=-\gamma[\mathbf{e}\mathbf{H}_e]-\alpha\gamma[\mathbf{e}[\mathbf{e}\mathbf{H}_e]],\\ \mathbf{H}_e=-\partial U/\partial\boldsymbol{\mu}=H\mathbf{h}+2KV\mu^{-1}(\mathbf{en})\mathbf{n}.\end{gathered} \tag{8}$$

An expression for the random velocity of wandering $\mathbf{v}_s$ can also be obtained[4] from the Landau-Lifshitz equation, by replacing the regular field H_e in (8) by the stochastic field

$$\mathbf{H}_s=-kT\partial\ln W/\partial\boldsymbol{\mu},$$

this gives

$$\mathbf{v}_s=\gamma\frac{kT}{\mu}[\mathbf{e}\nabla]\ln W+\alpha\gamma\frac{kT}{\mu}[\mathbf{e}[\mathbf{e}\nabla]]\ln W. \tag{9}$$

On substituting $\mathbf{v}=\mathbf{v}_r+\mathbf{v}_s$ in (7), we obtain, after simple transformations, the Fokker-Planck equation

$$\begin{gathered}2\tau\dot W=-i\hat{\mathbf{L}}\{\alpha^{-1}\xi\mathbf{h}+2\alpha^{-1}\sigma(\mathbf{en})\mathbf{n}+\xi[\mathbf{eh}]+2\sigma(\mathbf{en})[\mathbf{en}]-i\hat{\mathbf{L}}\}W,\\ \xi=\mu H/kT,\quad \hat{\mathbf{L}}=-i[\mathbf{e}\nabla].\end{gathered} \tag{10}$$

In the absence of an external field ($\xi=0$), Eq. (10) in spherical coordinates, with the polar axis along $\mathbf{n}$, takes the form

$$2\tau\frac{\partial W}{\partial t}=\frac{\partial^2W}{\partial\vartheta^2}+\mathrm{ctg}\,\vartheta\frac{\partial W}{\partial\vartheta}+\frac{1}{\sin^2\vartheta}\frac{\partial^2W}{\partial\varphi^2}$$

$$-2\frac{\sigma}{\alpha}\cos\vartheta\frac{\partial W}{\partial\varphi}+2\sigma\sin\vartheta\cos\vartheta\frac{\partial W}{\partial\vartheta}+2\sigma(3\cos^2\vartheta-1)W. \tag{11}$$

The stationary normalized solution of this equation is the Gibbs distribution

$$W_0=(4\pi F)^{-1}e^{\sigma x^2},\quad F(\sigma)=\int_0^1 e^{\sigma x^2}dx,\quad x=\mathbf{en}=\cos\vartheta. \tag{12}$$

An arbitrary deviation $W-W_0$ of the distribution function from its equilibrium value can be expanded in normal modes,

$$W_{lm}(x,\varphi,t)=\Psi_{lm}(x)\exp\{\sigma x^2+im\varphi-\Lambda_{lm}t\}. \tag{13}$$

On substituting (13) in (11), we obtain for the amplitudes of the normal modes Ψ_{lm} and for the dimensionless decrements $\lambda_{lm}=2\Lambda_{lm}\tau$ the equation

$$\begin{gathered}-\lambda\Psi=\hat A\Psi,\\ \hat A=\frac{d}{dx}\left[(1-x^2)\frac{d}{dx}\right]-\frac{m^2}{1-x^2}+2\sigma x(1-x^2)\frac{d}{dx}-2im\frac{\sigma}{\alpha}x.\end{gathered} \tag{14}$$

The solutions $\Psi(x)$ of (14) are orthogonal to the solutions of the conjugate problem

$$-\lambda^*\Phi=\hat A^+\Phi, \tag{15}$$

where $\hat A^+$ is the Hermitian conjugate operator to $\hat A$. The operator $\hat A^+$ looks especially simple if one requires orthogonality with weights:

$$\int_{-1}^{+1}\Psi_{km}(x)\Phi_{lm}(x)e^{\sigma x^2}dx=N\delta_{kl} \tag{16}$$

(N = const is a normalizing integral). To obtain the conjugate problem, we multiply the equation complex-conjugate to (14) by $\Phi(x)\exp(\sigma x^2)$ and integrate with respect to x:

$$\int_{-1}^{+1}\Phi e^{\sigma x^2}(\hat A^*\Psi^*+\lambda^*\Psi^*)dx=0.$$

By use of (14), this expression is easily transformed to the form

$$\int_{-1}^{+1}\Psi^*e^{\sigma x^2}(\hat A^*\Phi+\lambda^*\Phi)dx=0,$$

whence follows the equation for $\Phi(x)$

$$-\lambda^*\Phi=\hat A^*\Phi.$$

On comparing this with equation (15), we conclude that $\hat A^+=\hat A^*$; that is, the operator $\hat A^+$ differs from $\hat A$ only with respect to the sign of the last term in (14). This fact enables us to relate their eigenfunctions in a simple manner: $\Phi^*=\Psi$. Thus the orthogonality condition (16) takes the form

$$\int_{-1}^{+1}\Psi_{km}(x)\Psi_{lm}(x)e^{\sigma x^2}dx=N\delta_{kl}. \tag{17}$$

The nonhermicity of the operator $\hat A$ indicates that its eigenvalues λ may be complex.

Investigation of the spectrum of $\lambda(\sigma)$ is conveniently begun by consideration of the case of small σ. For $\sigma=0$, Eq. (14) is satisfied by the Legendre associated polynomials

$$\Psi_{lm}^{(0)}(x)=i^l\left\{\frac{2l+1}{2}\frac{(l-m)!}{(l+m)!}\right\}^{1/2}P_l^m(x),\quad \lambda_{lm}(0)=l(l+1). \tag{18}$$

Here the normalization is so chosen that the integral $N=+1$ for even l and $N=-1$ for odd.

For nonzero but sufficiently small σ, the solution of (14) can be constructed from series in powers of σ.

We shall not give these simple calculations. We point out only that the decrements λ in an arbitrary order of the perturbation theory remain real, whereas the functions Ψ at $\sigma \neq 0$ cease to be even and can be expressed as the sum of a real even part (index g) and an imaginary odd part (index u):

$$\Psi=\Psi_g+i\Psi_u \tag{19}$$

(for example, an expansion originating from an even level contains, at small σ, a small imaginary part, odd in x). It is nevertheless possible even when $\sigma \neq 0$ to speak as before of "even" and "odd" solutions, depending on which levels of the spectrum they approach at $\sigma \to 0$. We remark that the normalizing integrals (17)

$$N=\int \Psi^2 e^{\sigma x^2}\,dx=\int_{-1}^{+1} (\Psi_g^2-\Psi_u^2)e^{\sigma x^2}\,dx$$

of even functions are positive (since for $\sigma \to 0$ Ψ_u vanishes in them), whereas those of odd functions are negative.

Thus as long as the series in powers of σ converge, the perturbations (13) of the distribution function described by them decrease monotonically with time. This can also be seen directly. If one multiplies Eq. (14) by $\Psi^*\exp(\sigma x^2)$, integrates with respect to x, and subtracts from the resulting relation its complex conjugate, one obtains

$$\operatorname{Im}\lambda=2m\frac{\sigma}{\alpha}\frac{\int x|\Psi|^2\exp(\sigma x^2)\,dx}{\int |\Psi|^2\exp(\sigma x^2)\,dx}.$$

The integral in the numerator of this formula, on substitution of Ψ from (19), vanishes identically (by the parity properties).

Thus the occurrence of oscillatory perturbations is possible only for finite values of σ, larger than a certain σ_*. The latter also determines the radius of convergence of the series mentioned above. On multiplying (14) by $\Psi\exp(\sigma x^2)$ and operating further as in the derivation of the preceding relation, we obtain with allowance for (19)

$$(\lambda^*-\lambda)\int (\Psi_g^2-\Psi_u^2)e^{\sigma x^2}\,dx=0.$$

Hence it is evident that the occurrence of oscillatory ($\lambda \neq \lambda^*$) perturbations for $\sigma > \sigma_*$ is preceded by the vanishing, at the point σ_*, of the normalizing integral N. This type of singularity in the spectra of nonhermitian operators is well known[5]; they occur upon intersection of two levels to which, in the real range ($\sigma < \sigma_*$), correspond functions with normalizing integrals of opposite signs[6].

For investigation of intersections in the spectrum of decrements, a convenient method is that applied by Landau and Lifshitz[7] in the theory of electronic terms of molecules. For some $\sigma = \sigma_0$ let two neighboring decrements λ_1 and λ_2 be real and close together ("quasidegenerate"). Because of the alternation of even and odd levels, the normalizing integrals of the amplitudes Ψ_1 and Ψ_2 have opposite signs (let us say that $N_1 = +1$ and $N_2 = -1$). The amplitude at a neighboring point $\sigma_0 + \epsilon$ can be expressed in the form

$$\Psi=c_1\Psi_1+c_2\Psi_2.$$

For determination of the expansion coefficients c_1 and c_2, one obtains in the usual way, from (14), a homogeneous system of two linear equations. On setting the determinant of this system equal to zero, we find the decrements near the point σ_0:

$$\lambda_\pm=\frac{1}{2}[\lambda_1+\lambda_2-(B_{11}-B_{22})\varepsilon]\pm\{{}^1/_4[\lambda_1-\lambda_2-(B_{11}+B_{22})\varepsilon]^2-B_{12}B_{21}\varepsilon^2\}^{1/2}, \tag{20}$$

where

$$B_{ik}=\int_{-1}^{+1}\Psi_i\frac{\partial\hat{A}}{\partial\sigma}\Psi_k\exp(\sigma x^2)\,dx.$$

Proceeding to the analysis of formula (20), we note first of all that in the range of reality of the decrements, where the functions Ψ_i are expressible in the form (19), all the matrix elements B_{ik} are real. The decrements λ_+ and λ_- intersect at the point $\sigma_* = \sigma_0 + \epsilon$, where ϵ is found from the condition that the radicand in (20) must vanish. Satisfaction of this condition is possible only when $B_{12}B_{21} \geq 0$, since in the case $B_{12}B_{21} < 0$ the radicand is essentially positive for all ϵ. When $B_{12}B_{21} > 0$, there occurs at the point σ_* a "confluence" of two real levels; the radicand changes sign at this point, and for $\sigma > \sigma_*$ the decrements λ_+ and λ_- form a complex-conjugate pair. A "simple" intersection, for which the decrements would remain real on both sides of σ_*, is impossible, since it requires that the product $B_{12}B_{21}$ vanish identically with respect to σ_0, and this is known not to take place.

Thus neighboring real decrements either undergo confluence at some point σ_*, forming a complex-conjugate pair, or do not intersect at all.

A calculation of the decrement spectrum was done by Galerkin's method. The $\Psi_{lm}^{(0)}$ of (18), the solutions of equation (14) for $\sigma = 0$, were taken as the system of basis functions. Calculation of the function $\lambda(\sigma)$ over a sufficiently wide range of values of the parameter σ requires a large number of basis functions. In the interval investigated, $0 < \sigma < 20$, the approximation used was

$$\Psi=\sum_{l=1}^{n} c_{lm}\Psi_{lm}^{(0)} \tag{21}$$

with n = 20, and check calculations were made with n = 30. Diagonalization of the characteristic determinant was accomplished on an electronic computer.

Figure 1 shows the bottom levels of the spectrum for m = 1 and $\alpha = 0.1$. Their form corresponds completely to the general ideas about the structure of the spectrum of eigenvalues of equation (14). Evident on the figure is the confluence of the real levels λ_{11} and λ_{21}, with production of oscillatory modes. The dashed line shows the real part of the complex-conjugate decrements. Their imaginary part is also shown in the figure. The coordinate of the confluence point is $\sigma_* = 0.24$. This value determines the critical volume of the ferromagnetic particle, $V_* = \sigma_* kT/K$, at which the characteristic fre-

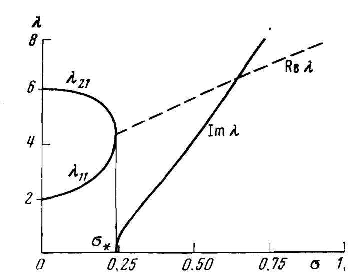

FIG. 1

quency of precession of its magnetic moment vanishes. For the critical diameter of a spherical particle at room temperature and with $K = 10^6$ to 10^4 erg/cm^3, we get $d_* = 30$ to 120 Å. We note that at such linear dimensions, ferromagnetism as a cooperative phenomenon is still fully retained: its destruction, according to an estimate of Vonsovskiĭ[8], occurs at $d \approx 10$ Å.

For $\sigma \gtrsim 15$, the decrements come out on the asymptote

$$\operatorname{Re}\lambda = 2\sigma, \quad \operatorname{Im}\lambda = \pm 2\sigma/\alpha. \tag{22}$$

Hence for the dimensional decrements $\Lambda = \lambda/2\tau$ one obtains, by (3) and (5), the asymptotic expressions (6).

3. DISPERSION OF THE MAGNETIC SUSCEPTIBILITY

The degree of orientation of the fluctuating magnetic moment $\boldsymbol{\mu} = \mu\mathbf{e}$ can be determined by averaging of the components of the vector e_i and of their products with the distribution function satisfying equation (10). By taking into account the hermicity of the operator $\hat{\mathbf{L}}$, it is easy to write an equation for an arbitrary moment of the distribution function. One obtains, as usual, an infinite system of coupled equations.

In decoupling this system, it is necessary to keep at least two equations—for the first and second moments of the distribution function:

$$\tau\frac{d}{dt}\langle e_i\rangle = -\langle e_i\rangle + \sigma n_i n_k\langle e_k\rangle - \frac{\sigma}{\alpha}e_{ikl}n_l n_m\langle e_k e_m\rangle + \frac{\xi}{2}\left(h_i + \frac{1}{\alpha}e_{ikl}h_k\langle e_l\rangle - h_k\langle e_i e_k\rangle\right), \tag{23}$$

$$\begin{aligned}\tau\frac{d}{dt}\langle e_i e_k\rangle = {}& \delta_{ik} - 3\langle e_i e_k\rangle + \sigma[n_i n_l\langle e_k e_l\rangle + n_k n_l\langle e_i e_l\rangle] + \frac{\sigma}{\alpha}n_n n_l[e_{klm}\langle e_i e_m e_n\rangle + e_{ilm}\langle e_k e_m e_n\rangle] \\ & - 2\sigma n_m n_l\langle e_i e_k e_l e_m\rangle + \frac{\xi}{2}[h_i\langle e_k\rangle + h_k\langle e_i\rangle + \frac{1}{\alpha}h_l(e_{klm}\langle e_i e_m\rangle + e_{ilm}\langle e_k e_m\rangle) - 2h_l\langle e_i e_k e_l\rangle].\end{aligned} \tag{24}$$

By means of the single equation (23) alone, no satisfactory description of the motion of the magnetic moment can be obtained: for any method of closure, this equation does not contain the characteristic frequency of precession of the vector $\langle\mathbf{e}\rangle$ in the anisotropy field. Only "entanglement" of the dipole (e_i) and quadrupole ($e_i e_k$) branches leads, in full agreement with the results of the preceding section, to the occurrence at finite σ of a characteristic frequency of oscillation and consequently makes possible ferromagnetic resonance in a periodic external field.

In accordance with the chosen (two moment) approximation, we shall seek a distribution function in the form

$$W = W_0(1 + a_i e_i + b_{ik}e_i e_k), \tag{25}$$

where W_0 is the equilibrium function determined by formula (12), and where a_i and b_{ik} are independent of the components of the vector $\mathbf{e}$ and are small quantities of the same order as the amplitude of the radiofrequency field ξ. On carrying out in (23) and (24) an averaging with the function (25) and on requiring only linear accuracy with respect to the quantities mentioned, we obtain for a_i and b_{ik} the equations

$$\begin{aligned}\tau X_{ik}\dot{a}_k = {}& -\{X_{ik} + \sigma[n_l n_m X_{ilmk} - n_i n_l X_{lk}]\}a_k \\ & - (\sigma/\alpha)e_{ikp}n_n n_p[X_{knlm} - X_{kn}X_{lm}]b_{lm} + (\xi/2)[h_i - h_k X_{ik}],\end{aligned} \tag{26}$$

$$\begin{aligned}\tau[X_{iklm} - X_{ik}X_{lm}]\dot{b}_{lm} = {}& -\{3[X_{iklm} - X_{ik}X_{lm}] - \sigma n_i n_n[X_{knlm} - X_{kn}X_{lm}] - \sigma n_k n_n[X_{inlm} - X_{in}X_{lm}] \\ & + 2\sigma n_n n_p[X_{iknplm} - X_{iknp}X_{lm}]\}b_{lm} + (\sigma/\alpha)n_l n_n[e_{knp}X_{iplm} + e_{inp}X_{kplm}]a_m \\ & + (\xi/2\alpha)h_l[e_{klm}X_{im} + e_{ilm}X_{km}],\end{aligned} \tag{27}$$

where we use the notation

$$X_{ik\ldots s} \equiv \langle e_i e_k \ldots e_s\rangle_0 = (4\pi F)^{-1}\int e_i e_k \ldots e_s \exp\{\sigma(\mathbf{en})^2\}d^3\mathbf{e}$$

for the moments of the function W_0. These quantities can be expressed in terms of derivatives of $F(\sigma)$ by means of (12), and their tensorial structure in terms of δ symbols and components of the vector $\mathbf{n}$. For example,

$$X_{ik} = \frac{1}{2}\left(1 - \frac{F'}{F}\right)\delta_{ik} + \frac{3}{2}\left(\frac{F'}{F} - \frac{1}{3}\right)n_i n_k.$$

We note also that the first moment of the distribution function (25) is connected with a_k by the relation

$$\langle e_i\rangle = X_{ik}a_k. \tag{28}$$

We first determine the longitudinal susceptibility $\chi_{\parallel}$, supposing that the external field is parallel to the anisotropy field: $\mathbf{h} = \mathbf{n} = (0, 0, 1)$. In this case one obtains for a_z, from (26), the closed equation

$$\tau X_{zz}\dot{a}_z = -[(1-\sigma)X_{zz} + \sigma X_{zzzz}]a_z + \frac{\xi}{2}(1 - X_{zz}).$$

By substituting $X_{zz} = F'/F$, $X_{zzzz} = F''/F$ and using (28), we write this equation in the form

$$\langle\dot{e}_z\rangle = -\tau_{\parallel}^{-1}\langle e_z\rangle + \alpha\gamma H(1 - F'/F), \qquad \tau_{\parallel} = \tau[1 - \sigma + \sigma F''/F']^{-1}. \tag{29}$$

In the absence of an external field, Eq. (29) describes a relaxation of the projection of the magnetic moment $\boldsymbol{\mu}$ on the direction of the axis of easy magnetization. Thermal fluctuations cause transitions between the states with $\boldsymbol{\mu} = \pm\mu\mathbf{n}$. The transition probability depends on the ratio of the height of the potential barrier KV between these states to the thermal energy kT; that is, it is determined by the parameter σ. If at the initial instant the particle was magnetized along the z axis, then the projection of the magnetic moment on this axis will decrease with time according to the expression $\exp(-t/\tau_{\parallel})$—"superparamagnetism"[8].

A graph of $\tau_{\parallel}/\tau$, constructed by formula (29, is shown in Fig. 2 by the dashed line. The solid line shows the function $2\lambda_{10}^{-1}(\sigma)$ calculated by equation (14) by Galerkin's method (by use of the approximation (21) with m = 0, n = 20). For $\sigma \gtrsim 2$ the solid curve is described well by the asymptotic formula of Brown[4]

$$(\tau_{\parallel}/\tau)_B = \sigma^{-1/2}e^{\sigma},$$

whereas the dotted approaches the asymptote 2σ. By the Appendix and formula (5), we obtain from (29)

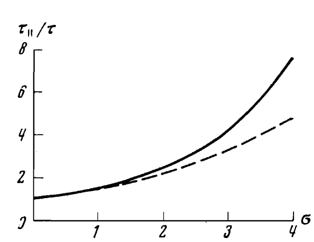

FIG. 2

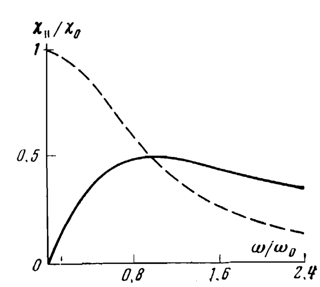

FIG. 3

$$\tau_{\parallel}=\tau_0\begin{cases}\sigma & (\sigma\ll 1)\\ 2\sigma^2 & (\sigma\gg 1)\end{cases}. \tag{30}$$

In a periodic field $H = H_0e^{i\omega t}$ parallel to the anisotropy field H_A, we find from (29) the magnetic susceptibility of unit volume

$$\chi_{\parallel}=(M/H_0)\langle e_z\rangle=\chi_0(1+i\omega\tau_{\parallel})^{-1},$$
$$\chi_0=\frac{M}{H_A}\frac{\sigma(1-F'/F)}{[1-\sigma(1-F''/F')]} \tag{31}$$

The frequency dependence of $\chi_{\parallel}$ is typical of relaxational systems: $\chi'_{\parallel}$ decreases monotonically with increase of ω, $\chi''_{\parallel}$ has a diffuse maximum at frequency $\omega_r = \tau_{\parallel}^{-1}$. In the limit of small σ, the "resonance" frequency $\omega_r = \omega_0\alpha/\sigma$ becomes infinite, whereas the static susceptibility is, to the second order of accuracy,

$$\chi_0=\frac{M}{H_A}\cdot\frac{2}{3}\sigma\left(1+\frac{4}{15}\sigma\right)=\frac{\mu M}{3kT}\left(1+\frac{4KV}{15kT}\right). \tag{32}$$

Thus the magnetic susceptibility of a system of fine ferromagnetic particles ($\sigma \ll 1$) is described by the Langevin formula characteristic of paramagnetic gases. The real and imaginary parts of $\chi_{\parallel}$ for $\sigma = \alpha = 0.1$ are given in Fig. 3.

For $\sigma \gg 1$, the fluctuational mechanism of reorientation of the magnetic moment is "frozen" (according to (30) $\tau_{\parallel}$ increases as σ^2). Under these conditions a weak alternating field ($\xi \ll \sigma$, $\omega\tau_{\parallel} \gg 1$) is incapable of insuring any appreciable probability of a transition between the states $\mu = \pm\mu n$, and for $\sigma\to\infty$ the longitudinal susceptibility (31) approaches zero ($\chi'_{\parallel}\sim\sigma^{-3}$, $\chi''_{\parallel}\sim\sigma^{-1}$).

We now consider the magnetic properties of subdomain particles in a transverse external field. Directing it along the x axis (the z axis, as before, is oriented along the anisotropy field), we obtain from (26) and (27) the following equations for the components $a_x = a$ and $b_{yz} = b$:

$$2\tau\dot{a}=-\lambda_a a-4\frac{\sigma}{\alpha}\frac{F'-F''}{F-F'}b+\xi\frac{F+F'}{F-F'},$$
$$2\tau\dot{b}=-\lambda_b b+\frac{\sigma}{\alpha}a-\frac{\sigma}{\alpha}\xi. \tag{33}$$

Here

$$\lambda_a=2[1+\sigma(F'-F'')/(F-F')],$$
$$\lambda_b=2[3-\sigma+2\sigma(F''-F''')/(F'-F'')]. \tag{34}$$

The homogeneous system of equations from (33) for $\xi = 0$ has the damped solutions

$$a=a_0e^{-\lambda t/2\tau},\quad b=b_0e^{-\lambda t/2\tau}$$

with a decrement $\lambda(\sigma)$ determined from the condition for compatibility of the system

$$(\lambda-\lambda_a)(\lambda-\lambda_b)+(2\sigma/\alpha)^2(F'-F'')/(F-F')=0. \tag{35}$$

The roots of the quadratic equation (35) describe well the behavior of the curves in Fig. 1. For $\sigma = 0$ we have $\lambda_1 = \lambda_a(0) = 2$ and $\lambda_2 = \lambda_b(0) = 6$, which agrees with the corresponding eigenvalues (18) of the Fokker-Planck equation. We note further that for all ferromagnets, $\alpha \ll 1$. Therefore for nonvanishing but small values of σ it is sufficient to retain this parameter only in the combination σ/α in (35). In this approximation, the discriminant of Eq. (35) changes sign at the point $\sigma_* = \alpha\sqrt{5}$. For $\alpha = 0.1$ we thus have $\sigma_* \approx 0.22$, which is close to the value $\sigma_* \approx 0.24$ given in Section 2. Finally, at $\sigma \gg 1$, Eq. (35 has the roots (cf. (22))

$$\lambda_{1,2}=2\sigma(1\pm i\alpha^{-1}). \tag{36}$$

The dimensional decrements corresponding to the values (36)

$$\Lambda=\lambda/2\tau=\alpha\omega_0\pm i\omega_0$$

coincide with the eigenvalues (6) of the Landau-Lifshitz equation (1).

We shall calculate the magnetic susceptibility $\chi_{\perp}$ in a periodic field ξ. On setting

$$(a,b,\xi)=(a_0,b_0,\xi_0)e^{i\omega t}$$

in (33) and solving the inhomogeneous system of equations for the amplitudes a_0 and b_0, we find

$$a_0=\Delta_a/\Delta,\quad \Delta=(\lambda_1+2i\omega\tau)(\lambda_2+2i\omega\tau),$$
$$\Delta_a=\xi_0(F-F')^{-1}[(F+F')(\lambda_b+2i\omega\tau)+(2\sigma/\alpha)^2(F'-F'')].$$

In the determinant Δ of the system, λ_1 and λ_2 denote the roots of (35). By expressing $\langle e_x\rangle$ in terms of a_x by formula (28), we obtain for the magnetic susceptibility

$$\chi_{\perp}=\frac{M}{H_0}\frac{(F-F')\Delta_a}{2F\Delta}.$$

After simple transformations that use (34) and (35), the last formula can be put into the form

$$\chi_{\perp}=\frac{M}{H_A}\frac{\omega_0(\omega_0R_3+i\omega R_4)}{\omega_0^2R_1-\omega^2+2i\omega\omega_0R_2}. \tag{37}$$

Here the coefficients R_i are functions of σ:

$$R_1=\frac{F'-F''}{F-F'}+\left(\frac{\alpha}{\sigma}\right)^2\left[1+\sigma\frac{F'-F''}{F-F'}\right]\left[3-\sigma+2\sigma\frac{F''-F'''}{F'-F''}\right],$$
$$R_2=\frac{\alpha}{2\sigma}\left[4-\sigma+\sigma\frac{F'-F''}{F-F'}+2\sigma\frac{F''-F'''}{F'-F''}\right],$$
$$R_3=\sigma\frac{F-F'}{F}+\frac{\alpha^2(F+F')}{2\sigma F}\left[3-\sigma+2\sigma\frac{F''-F'''}{F'-F''}\right],$$
$$R_4=\alpha(F+F')/2F. \tag{38}$$

Hence, by use of formulas given in the Appendix, we obtain in the case of small σ

$$R_1\approx 3\left(\frac{\alpha}{\sigma}\right)^2\left(1+\frac{16}{105}\sigma\right),\quad R_2\approx 2\frac{\alpha}{\sigma}\left(1+\frac{1}{70}\sigma\right),$$
$$R_3\approx 2\frac{\alpha^2}{\sigma}\left(1+\frac{2}{105}\sigma\right),\quad R_4\approx\frac{2}{3}\alpha\left(1+\frac{1}{15}\sigma\right) \tag{39}$$

the limiting values for $\sigma\to\infty$ are

$$R_1=R_3=1+\alpha^2,\quad R_2=R_4=\alpha. \tag{40}$$

In the last case, as should be true for $KV \gg kT$, (37) and (40) lead to the result of Landau and Lifshitz[1]

$$\chi_{\perp}=\gamma M\frac{(1+\alpha^2)\omega_0+i\alpha\omega}{(1+\alpha^2)\omega_0^2-\omega^2+2i\alpha\omega\omega_0}, \tag{41}$$

while for $KV \ll kT$, substitution of (39) in (37) gives

$$\chi_{\perp}=\chi_0(1+i\omega\tau)^{-1}, \tag{42}$$
$$\chi_0=\frac{\mu M}{3kT}\left(1-\frac{2KV}{15kT}\right), \tag{43}$$

where τ is defined by formula (4). As $KV/kT\to 0$, the longitudinal and transverse static susceptibilities (32) and (43) coincide. The same is true also of the complete susceptibility: on comparing $\chi_{\perp}$ of (42) with the expression for $\chi_{\parallel}$ obtained in the limit of small σ, we see that they are identical. This result is entirely natural, since in this limit the particle becomes isotropic (the magnetic anisotropy constant K drops out of all the formulas).

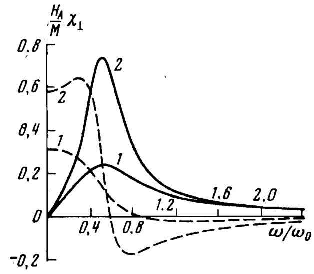

FIG. 4

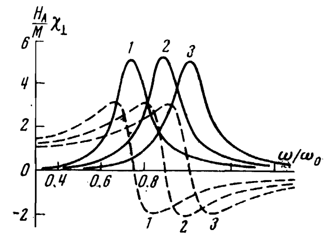

FIG. 5

FIG. 4. Dashed lines, $\chi_\perp'$; solid lines, $\chi_\perp''$. 1, $\sigma = 0.5$; 2, $\sigma = 1.0$.

FIG. 5. Dashed lines, $\chi_\perp'$; solid lines, $\chi_\perp''$. 1, $\sigma = 5$; 2, $\sigma = 10$; 3, $\sigma = \infty$ (the Landau-Lifshitz susceptibility).

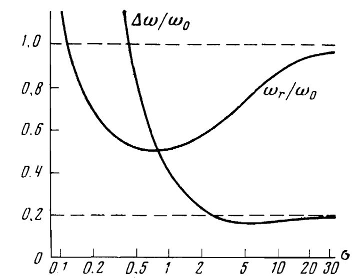

FIG. 6

At $\sigma = 0.1$, the graphs of the functions $\chi_\perp(\omega)$ calculated by formulas (37) and (38) practically coincide with the graphs of $\chi_{\parallel}(\omega)$ shown in Fig. 3. The $\chi_\perp$ lines shown in Fig. 4 are intermediate between the relaxation curves of Fig. 3 and the resonance curves of Fig. 5. At $\sigma = 0.5$, the dispersion of $\chi_\perp$ still has a relaxational character, although on the $\chi_\perp'$ curve there is already a note at a finite value of ω, typical of resonance curves. The dispersion at $\sigma = 1$ must be considered rather of resonance type.

Figure 6 shows, as functions of the parameter σ, the resonance frequency ω_r, determined by the position of the maximum on the $\chi_\perp''(\omega)$ curve, and the width $\Delta\omega$ of this curve at its half-height. The function $\omega_r(\sigma)$ has a minimum at $\sigma_0 = 0.73$. The value σ_0 may be considered a nominal boundary separating the regions of resonance ($\sigma > \sigma_0$) and relaxational ($\sigma < \sigma_0$) dispersion of the magnetic susceptibility. Excess of σ over σ_0 is accompanied by increase of the resonance frequency and narrowing of the absorption line; for $\sigma \to \infty$, the limiting values $\omega_r \approx \omega_0$ and $\Delta\omega \approx 2\alpha\omega_0$ are reached (these approximate equalities are more accurate, the smaller α).

We note that the value σ_0 is three times as large as the critical value σ_* at which the characteristic frequency of precession vanishes. This is explained by the fact that in the interval $\sigma_0 < \sigma < \sigma_*$ the characteristic frequency, though nonzero, is small, and therefore, because of the strong damping of the precession, the character of the dispersion of the susceptibility in this range of values of the parameter is the same (relaxational) as for $\sigma < \sigma_*$. But even in the relaxational region, that is for $\sigma < \sigma_0$, there is a maximum on the $\chi_\perp''$ curves ("resonance"; see Fig. 3 for $\chi_{\parallel} = \chi_\perp$). With decrease of σ this maximum becomes smoothed out and is shifted in the direction of larger frequencies. In the case $\sigma \ll 1$, when formula (42) is valid, the "resonance" frequency ω_r is equal to $\tau^{-1} = \alpha\omega_0/\sigma$, so that ω_r and $\Delta\omega$ become infinite for $\sigma \to 0$.

We have been concerned above with the magnetic properties of an individual particle. The magnetic susceptibility of a system of noninteracting particles, whose anisotropy axes are oriented in a random fashion, is

$$\chi = \frac{c}{3}(\chi_{\parallel} + 2\chi_\perp), \tag{44}$$

where c = nV is the volume concentration of the magnetic phase and n is the number density of the particles. In the case $\sigma \ll 1$, we get for the static susceptibility of such a system, by (32), (43), and (44),

$$\chi_0 = \frac{n\mu^2}{3kT}.$$

APPENDIX

Directly from the definition (12) of the function

$$F(\sigma) = \int_0^1 e^{\sigma x^2} dx \tag{A.1}$$

follows the formula for its derivatives at zero,

$$\left(\frac{d^n F}{d\sigma^n}\right)_{\sigma=0} = \frac{1}{2n+1}.$$

Thus in the case $\sigma \ll 1$ we have

$$F \approx 1 + {}^1/_3\sigma,\ F' \approx {}^1/_3 + {}^1/_5\sigma,$$
$$F'' \approx {}^1/_5 + {}^1/_7\sigma,\ F''' \approx {}^1/_7 + {}^1/_9\sigma.$$

In order to find an asymptotic expansion of F at large values of σ, we use the equation for this function

$$F' = (e^\sigma - F)/2\sigma, \tag{A.2}$$

which is obtained from (A.1) by differentiating with respect to σ and subsequently integrating the right side with respect to x. On substitution in (A.2) of

$$F = e^\sigma f/2\sigma \tag{A.3}$$

one obtains for the function $f(\sigma)$ the equation

$$f' + (1 - 1/2\sigma) f = 1$$

or, on transforming to the variable $\rho = \sigma^{-1}$,

$$-\rho^2 \frac{df}{d\rho} + \left(1 - \frac{\rho}{2}\right) f = 1. \tag{A.4}$$

We seek a solution of the last equation in the form of a series

$$f = \sum_n a_n \rho^n. \tag{A.5}$$

For the coefficients a_n, we obtain from (A.4) the recurrence formula

$$a_n = (n - {}^1/_2) a_{n-1},\quad a_0 = 1. \tag{A.6}$$

On substituting (A.5) and (A.6) in (A.3), we have

$$F = \frac{e^\sigma}{2\sigma}\left(1 + \frac{1}{2\sigma} + \frac{3}{4\sigma^2} + \ldots\right).$$

By differentiating this formula, we find asymptotic ($\sigma \gg 1$) expressions for the derivatives.

$$F' = \frac{e^\sigma}{2\sigma}\left(1 - \frac{1}{2\sigma} - \frac{1}{4\sigma^2} + \ldots\right),$$
$$F'' = \frac{e^\sigma}{2\sigma}\left(1 - \frac{3}{2\sigma} + \frac{3}{4\sigma^2} + \ldots\right),$$
$$F''' = \frac{e^\sigma}{2\sigma}\left(1 - \frac{5}{2\sigma} + \frac{15}{4\sigma^2} + \ldots\right).$$

*$[\mu H_e] \equiv \mu \times H_e$.

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Translated by W. F. Brown, Jr.
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