

the first term will be the most important.

By the methods described it is possible to investigate experimentally whether there are locally-biaxial fluctuations, i.e., whether the constant c_1 is nonzero. In order to eliminate the effect of complicating terms such as the additional terms in (5.4), it is appropriate to make $\mathbf{k} - \mathbf{k}'$ parallel to \mathbf{n}^0 . These terms then disappear.

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Magnetic resonance of an isotropic superparamagnet¹⁾

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Magnetic resonance induced by a circularly or linearly polarized high-frequency field \mathbf{h} in an isotropic superparamagnet located in a stationary field \mathbf{H} is investigated. It is shown that orientational diffusion of the magnetic moment, leading to superparamagnetism of small particles of the ferromagnet, deforms the absorption line. The line shape is defined by the Langevin parameter $\sigma = MVH/kT$ (M is the magnetization, V the particle volume). The resonance line at $\sigma \gg 1$ changes into a purely relaxation line at $\sigma \ll 1$. Thermal fluctuations are taken into account by replacing the dynamic description of motion of the magnetic moment (Landau-Lifshitz equation) by a statistical description (Fokker-Planck equation). Two methods of solving the problem are presented. One is based on a direct solution of the Fokker-Planck equation for a circularly polarized field \mathbf{h} . This method involves a search for a rotating coordinate system whose axis of rotation coincides with the direction of precession of the mean magnetization. The other method of calculating the dynamic susceptibility is based on the use of the equation of motion of the magnetization of the superparamagnet. The equation is derived from the Fokker-Planck equation by the method of moments.

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INTRODUCTION

A superparamagnet is an ensemble of non-interacting ferromagnetic particles, the dimensions of which are so small that the formation of a domain structure in them is energywise unfavored; thus, each particle is magnetized to saturation even in the absence of an external magnetic field, although the total magnetic moment of the ensemble is equal to zero. Under the influence of a magnetic field \mathbf{H} , a system of such particles becomes magnetized like a classical Langevin paramagnet

$$\langle m_z \rangle = \langle M_z / M \rangle = \text{cth } \sigma - \sigma^{-1} = L(\sigma),$$

where $\sigma = MVH/kT$, M is the magnetization per unit volume at $T=0$, and V is the volume of the particle.

At sufficiently small V and not too large H , the characteristic paramagnetic-disorder temperatures can lie much lower than the Curie temperature T_c ; greatest interest attaches then to the temperature region $T \ll T_c$, where the temperature dependence of M can be neglected.

The theoretical study of the dynamics of superparamagnets was initiated by Brown,^[1] who wrote down the Fokker-Planck equation for such a system and investigated the processes of relaxation in anisotropic particles. The magnetic resonance of an anisotropic superparamagnet under the influence of a weak high-frequency field at $H=0$ (resonance in the effective field of uniaxial magnetic anisotropy) was investigated in^[2]. Dynamic magnetic hysteresis of an isotropic superparamagnet under the influence of an alternating magnetic field of arbitrary amplitude was investigated in^[3].

The present paper is devoted to the dynamic susceptibility of an isotropic superparamagnet placed in a constant field \mathbf{H} and in a weak high-frequency field \mathbf{h} . In this situation, size effects typical of ultrasmall particles of a ferromagnet should appear. We note that a particle with diameter on the order of 100 Å contains only $10^4 - 10^5$ atoms. This number is not large enough to be able to neglect the thermal fluctuations of the field. The amplitude of the fluctuating field is of the order of kT/MV , so that for microcrystals with volume $V \lesssim 10^{-18} \text{ cm}^3$ at a

magnetization of the material $M \approx 10^3$ G and at room temperature amounts to 10^3 Oe. This quantity is commensurate with the intensity of the constant field ($H \sim 10^3$ Oe), in which ferromagnetic resonance (FMR) is usually observed. The presence of an appreciable random field causes stochastic reorientation of the magnetic moments of the particles and this, as will be shown below, greatly influences the FMR line shape.

In Secs. 1 and 2 we consider magnetic resonance in a field h of circular polarization. The method of solving the Fokker-Planck equation is based here on a search for a rotating coordinate system whose rotation axis coincides with the direction of macroscopic magnetization of the ensemble of particles. Approximate analytic expressions are obtained for the temperature dependence of the absorption line width and for the position of the resonant maximum; these expressions are valid in a wide temperature interval.

In Sec. 3, the Fokker-Planck equation is used to derive the equation of motion of the macroscopic magnetization of a superparamagnet. The equation for the first moment of the distribution function is made closed under the assumption of a weak disequilibrium. The obtained equation of motion of the magnetization is used to calculate the dynamic susceptibility in a linearly-polarized high-frequency field. It is shown that the absorption line becomes deformed with decreasing Langevin argument σ : it changes from a resonance shape at $\sigma \gg 1$ to a relaxation shape at $\sigma \ll 1$; in this limit, the relative line width $\Delta H/H$ increases like σ^{-1} .

1. THE FOKKER-PLANCK EQUATION IN A SPHERICAL COORDINATE SYSTEM

We consider an ensemble of small spherical magnetically-isotropic ferromagnetic particles. Let a constant magnetic field H be directed along the z axis and a high-frequency field h rotate with frequency ω in the xy plane. The equation of motion of the magnetization M in each particle is the Landau-Lifshitz equation

$$\dot{M} = -\gamma[M \times \mathcal{H}] - \frac{\xi\gamma}{M}[M \times [M \times \mathcal{H}]], \quad (1.1)$$

where the wave field $\mathcal{H} = H + h$ consists of a constant and an alternating component. In the case of circular polarization of the high-frequency field h , the vector \mathcal{H} has components

$$\mathcal{H} = \{h \cos \omega t, h \sin \omega t, H\}. \quad (1.2)$$

The Fokker-Planck kinetic equation for the distribution function $W(\theta_1, \varphi_1, t)$ can be obtained by the method used in^[1-3]:

$$\begin{aligned} \frac{1}{\gamma} \frac{\partial W}{\partial t} = & \frac{\xi H}{\sigma} \left[\frac{\partial^2 W}{\partial \theta^2} + \csc^2 \theta \frac{\partial^2 W}{\partial \varphi^2} + \operatorname{ctg} \theta \frac{\partial W}{\partial \theta} \right] + [\xi H \sin \theta \\ & - \xi h \cos \theta \cos(\varphi - \omega t) - h \sin(\varphi - \omega t)] \frac{\partial W}{\partial \theta} + [-H + h \operatorname{ctg} \theta \cos(\varphi - \omega t) \\ & + \xi h \csc \theta \sin(\varphi - \omega t)] \frac{\partial W}{\partial \varphi} + 2\xi [H \cos \theta + h \sin \theta \cos(\varphi - \omega t)] W. \end{aligned} \quad (1.3)$$

Let us consider some limiting cases.

In the absence of the alternating field h , the system that has been taken out of equilibrium relaxes to the ground state. If furthermore the temperature is also equal to zero, then the solution (1.3) takes the form of the product of δ functions:

$$W = \delta \left(x - \frac{x^0 + \operatorname{th} \xi \gamma H t}{1 + x^0 \operatorname{th} \xi \gamma H t} \right) \delta(\varphi - \varphi^0 - \gamma H t), \quad (1.4)$$

where $x = \cos \theta$, and θ^0 and φ^0 are the initial angles. It is seen that the distribution function differs from zero only along the trajectory of motion of the magnetic moment determined by the Landau-Lifshitz equations:

$$d\theta/dt = -\xi \gamma H \sin \theta, \quad d\varphi/dt = \gamma H. \quad (1.5)$$

For sufficiently large t we obtain

$$W = \delta(x-1) \delta(\varphi - \varphi^0 - \gamma H t). \quad (1.6)$$

In the random-phase approximation, this function goes over into the limit of the equilibrium Gibbs distribution as $T \rightarrow 0$:

$$W = \delta(x-1) = \lim_{\sigma \rightarrow 0} e^{\sigma x} / \int_{-1}^1 e^{\sigma x} dx. \quad (1.7)$$

If $h \neq 0$ but is small enough ($h/H \ll 1$) then, at any temperature in the steady state, the average magnetization vector $\langle M \rangle$ of the particle ensemble will precess about the z axis with a frequency ω , i. e., it will remain immobile relative to the rotating field h . It is therefore reasonable to change over into a rotating coordinate system; in this system Eq. (1.3) for the steady-state motion takes the following form (we retain the same symbol for the azimuthal angle φ in the rotating system):

$$\begin{aligned} \frac{\xi H}{\sigma} \left(\frac{\partial^2 W}{\partial \theta^2} + \csc^2 \theta \frac{\partial^2 W}{\partial \varphi^2} + \operatorname{ctg} \theta \frac{\partial W}{\partial \theta} \right) + (\xi H \sin \theta \\ - \xi h \cos \theta \cos \varphi - h \sin \varphi) \frac{\partial W}{\partial \theta} + \left(-H + \frac{\omega}{\gamma} + h \operatorname{ctg} \theta \cos \varphi \right. \\ \left. + \xi h \csc \theta \sin \varphi \right) \frac{\partial W}{\partial \varphi} + 2\xi (H \cos \theta + h \sin \theta \cos \varphi) W = 0. \end{aligned} \quad (1.8)$$

The time does not enter explicitly in this equation—the function W for the steady state does not depend on t in the rotating coordinate system. Let us examine qualitatively the form that the distribution function assumes in this system. If $h=0$ and $\omega=0$, the equation is satisfied by a function that does not depend on the azimuthal angle φ :

$$W = \sigma e^{\sigma \cos \theta} / 4\pi \operatorname{sh} \sigma. \quad (1.9)$$

This function is shown by the solid line in Fig. 1. If $h \neq 0$ but $\omega=0$, the function W tilts away from the z axis without changing its shape (Fig. 1, dashed line). In the xyz system the function W now depends both on θ and on φ . However, in the $x'y'z'$ system, which is rotated through an angle θ_0 such that the z' axis coincides with the direction of the resultant field $H+h$, the function W is again independent of φ and takes the form (1.9). (Actually it is necessary here to make in (1.9), besides the substitution $\theta \rightarrow \theta'$, the substitution $H \rightarrow (H^2 + h^2)^{1/2}$, but we shall neglect throughout the corrections of order higher than the first power of h .)

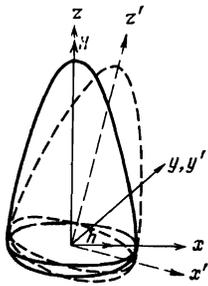


FIG. 1. Equilibrium distribution functions: the solid line corresponds to the case when the weak perpendicular magnetic field $h=0$, and the dashed line corresponds to $h \neq 0$.

Let now $\omega \neq 0$. Then, in addition to the inclination of the function W through a certain angle away from the z axis (which now no longer coincides with the direction of the combined magnetic field), the function will also rotate through a certain angle φ_0 that depends on the frequency ω (the angle by which the average magnetic moment lags the rotating field); in this case, as will be shown later on, a certain deformation of the figure itself will take place—its symmetry becomes lower (Fig. 2). Naturally, the function W in the xyz system has in this case an extremely complicated form.

We rotate the coordinate system $x'y'z'$ through arbitrary angles θ_0 and φ_0 relative to the xyz system. As will be shown later on, $\theta_0 \sim h/H \ll 1$, so that we confine ourselves from the outset to its first power; no limitations whatever are imposed on the value of φ_0 ($\varphi_0 = \pi/2$ at the resonance point):

$$\begin{aligned} \sin \theta \cos \varphi &= \cos \varphi_0 \sin \theta' \cos \varphi' - \sin \varphi_0 \sin \theta' \sin \varphi' + \theta_0 \cos \varphi_0 \cos \theta', \\ \sin \theta \sin \varphi &= \cos \varphi_0 \sin \theta' \sin \varphi' + \sin \varphi_0 \sin \theta' \cos \varphi' \\ &\quad + \theta_0 \sin \varphi_0 \cos \theta', \\ \cos \theta &= \cos \theta' - \theta_0 \sin \theta' \cos \varphi'. \end{aligned} \quad (1.10)$$

In the primed spherical system, the Fokker-Planck equation takes the form

$$\begin{aligned} &\frac{\xi H}{\sigma} \left(\frac{\partial^2 W}{\partial \theta'^2} + \csc^2 \theta' \frac{\partial^2 W}{\partial \varphi'^2} + \text{ctg } \theta' \frac{\partial W}{\partial \theta'} \right) + \left[- \left(H - \frac{\omega}{\gamma} \right) \theta_0 \sin \varphi' \right. \\ &+ \xi H (\sin \theta' + \theta_0 \cos \theta' \cos \varphi') + h \sin (\varphi' + \varphi_0) - \xi h \cos \theta' \cos (\varphi' + \varphi_0) \left. \right] \frac{\partial W}{\partial \theta'} \\ &+ \left[- \left(H - \frac{\omega}{\gamma} \right) (1 + \theta_0 \text{ctg } \theta') - \xi H \theta_0 \sin \varphi' \csc \theta' + h \text{ctg } \theta' \cos (\varphi' + \varphi_0) \right. \\ &+ \xi h \csc \theta' \sin (\varphi' + \varphi_0) \left. \right] \frac{\partial W}{\partial \varphi'} + 2\xi [H (\cos \theta' - \theta_0 \sin \theta' \cos \varphi') \\ &+ h \sin \theta' \cos (\varphi' + \varphi_0)] W = 0. \end{aligned} \quad (1.11)$$

What considerations should govern the choice of θ_0 and φ_0 ? It seems natural to require that the z' axis of the rotated system pass through the maximum of the distribution function W . Indeed, in this case one can expect the function W in the primed system to have the simplest form. It will be more convenient for us, however, to rotate the coordinate system in a different manner. To solve the magnetic-resonance problem it suffices to know the mathematical expectation values of the projections of the magnetization in the initial (rotating) system xyz . We there rotate the coordinate system in such a way that the z' axis passes through the direction of the expectation value of the orientation of the magnetization of the ensemble (in view of the asymmetry of the function W , the direction of $\langle \mathbf{m} \rangle$ does not coincide

with the maximum of the function W). Indeed, in this case we have in the primed coordinate system

$$\langle m_x' \rangle = 0, \quad \langle m_y' \rangle = 0. \quad (1.12)$$

The sought mathematical expectations of the projections of the magnetization in the xyz system are then expressed, as can be seen from (1.10), in the following manner:

$$\langle m_x \rangle = \theta_0 \cos \varphi_0 \langle m_x' \rangle, \quad \langle m_y \rangle = \theta_0 \sin \varphi_0 \langle m_x' \rangle, \quad \langle m_z \rangle = \langle m_z' \rangle. \quad (1.13)$$

Thus, to find all the quantities of interest to us it suffices to know only the angles of rotation and $\langle m_x' \rangle$.

We consider Eq. (1.11), assuming that θ_0 and φ_0 have already been chosen such that the z' axis passes through the direction of the average magnetization. The dependence of the function W on φ' should then be weak and should vanish as $h \rightarrow 0$, i. e., $\partial W / \partial \varphi' \sim h$ and $\partial^2 W / \partial \varphi'^2 \sim h$. Neglecting in (1.11) the products of the small quantities $h \partial W / \partial \varphi'$ and $\theta_0 \partial W / \partial \varphi'$, we see that its solution can be represented in the form

$$W(\theta', \varphi') = C \exp \{ \sigma [\cos \theta' + u(\theta') \cos \varphi' + v(\theta') \sin \varphi'] \}, \quad (1.14)$$

where C is the normalization constant, while u and v are unknown functions of the polar angle θ' proportional to h . Substitution of (1.14) in (1.11) leads to the following differential equations for the functions u and v :

$$\begin{aligned} \frac{d^2 u}{d\theta'^2} + (\text{ctg } \theta' - \sigma \sin \theta') \frac{du}{d\theta'} - \csc^2 \theta' u - \frac{\sigma}{\xi} \left(H - \frac{\omega}{\gamma} \right) v &= F_u, \\ \frac{d^2 v}{d\theta'^2} + (\text{ctg } \theta' - \sigma \sin \theta') \frac{dv}{d\theta'} - \csc^2 \theta' v + \frac{\sigma}{\xi} \left(H - \frac{\omega}{\gamma} \right) u &= F_v, \end{aligned} \quad (1.15)$$

where the right-hand sides are determined by the expressions

$$\begin{aligned} F_u &= h \sigma \sin \theta' \left[\cos \theta' \left(\frac{H \theta_0}{h} - \cos \varphi_0 \right) + \frac{1}{\xi} \sin \varphi_0 + \frac{2 \theta_0 H}{\sigma h} - \frac{2}{\sigma} \cos \varphi_0 \right], \\ F_v &= h \sigma \sin \theta' \left[\cos \theta' \sin \varphi_0 - \frac{H - \omega/\gamma}{\xi h} \theta_0 + \frac{1}{\xi} \cos \varphi_0 + \frac{2}{\sigma} \sin \varphi_0 \right]. \end{aligned} \quad (1.16)$$

To determine the unknown angles θ_0 and φ_0 of rotation of the coordinate system it is most convenient to change over from Eq. (1.11) to the equations for the moments in the primed system. Using (1.11) for the distribution factor W , we have for the first moments of $\langle m_x' \rangle$, $\langle m_y' \rangle$, and $\langle m_z' \rangle$, respectively,

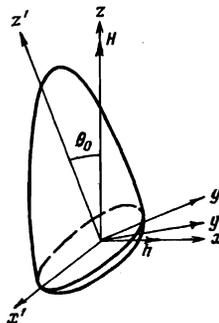


FIG. 2. Stationary distribution function in rotating coordinate system.

$$\begin{aligned}
(H-\omega/\gamma)\langle m_y' \rangle + \xi H[\langle m_x' m_z' \rangle + (1-\langle m_x'^2 \rangle)\theta_0] + h \sin \varphi_0 \langle m_x' \rangle \\
- \xi h[\langle m_x' m_y' \rangle \sin \varphi_0 + (1-\langle m_x'^2 \rangle)\cos \varphi_0] - \frac{2\xi H}{\sigma} \langle m_x' \rangle = 0, \\
(H-\omega/\gamma)(\langle m_x' \rangle + \theta_0 \langle m_z' \rangle) - \xi H(\langle m_y' m_z' \rangle - \theta_0 \langle m_x' m_y' \rangle) \\
- h \cos \varphi_0 \langle m_x' \rangle - \xi h[\langle m_x' m_y' \rangle \cos \varphi_0 + (1-\langle m_y'^2 \rangle)\sin \varphi_0] + \frac{2\xi H}{\sigma} \langle m_y' \rangle = 0, \\
\left(H - \frac{\omega}{\gamma}\right)\theta_0 \langle m_y' \rangle - \xi H(1-\langle m_x'^2 \rangle + \theta_0 \langle m_x' m_z' \rangle) - \frac{2\xi H}{\sigma} \langle m_x' \rangle \\
+ \xi h(\langle m_x' m_z' \rangle \cos \varphi_0 - \langle m_y' m_z' \rangle \sin \varphi_0) - h(\langle m_x' \rangle \sin \varphi_0 + \langle m_y' \rangle \cos \varphi_0) = 0.
\end{aligned} \quad (1.17)$$

We impose the conditions (1.12) on this system and calculate the paired moments and $\langle m_x' \rangle$, using expression (1.14) for the distribution function. All the integrals with respect to φ are then expressed in terms of the Bessel functions I_n of argument $\sigma(u^2 + v^2)^{1/2}$ (see, e.g., [4], formula 3.937). When they are subsequently expanded up to the first powers of σu and σv , we obtain the same results that can be obtained also in simpler fashion—by directly representing the distribution function (1.14) in the form of an expansion in these quantities:

$$W(\theta', \varphi') \approx C \exp(\sigma \cos \theta') (1 + \sigma u \cos \varphi' + \sigma v \sin \varphi'). \quad (1.18)$$

In this approximation we have

$$\begin{aligned}
\langle m_x' \rangle = L, \quad \langle m_x' m_y' \rangle = 0, \\
\langle m_x'^2 \rangle = \langle m_y'^2 \rangle = L/\sigma, \quad \langle m_z'^2 \rangle = 1 - 2L/\sigma, \\
\langle m_x' m_z' \rangle = \frac{\sigma^2}{4 \operatorname{sh} \sigma} \int_0^\pi e^{\sigma \cos \theta} \sin^2 \theta \cos \theta u(\theta) d\theta, \\
\langle m_y' m_z' \rangle = \frac{\sigma^2}{4 \operatorname{sh} \sigma} \int_0^\pi e^{\sigma \cos \theta} \sin^2 \theta \cos \theta v(\theta) d\theta.
\end{aligned} \quad (1.19)$$

Substituting the obtained expressions in the system (1.17), we find that the third equation is satisfied identically, and the first two take on the form

$$\begin{aligned}
\xi H(1-L/\sigma)\theta_0 + Lh \sin \varphi_0 - (1-L/\sigma)\xi h \cos \varphi_0 = -\xi H \langle m_x' m_z' \rangle, \\
-L(H-\omega/\gamma)\theta_0 + Lh \cos \varphi_0 + (1-L/\sigma)\xi h \sin \varphi_0 = -\xi H \langle m_y' m_z' \rangle
\end{aligned} \quad (1.20)$$

and determine the values of the angles θ_0 and φ_0 through which it is necessary to rotate the coordinate system in order to satisfy the conditions (1.12). We note that the same equations can be obtained also in a different manner, namely by multiplying each equation of the system (1.15) by $e^{\sigma \cos \theta'} \sin^2 \theta'$ and integrating with respect to $d\theta'$.

2. MAGNETIC RESONANCE IN A ROTATING HIGH-FREQUENCY FIELD

The exact solution of the problem of linear magnetic resonance of an isotropic superparamagnet is given by expressions (1.13), in which it is necessary to substitute $\langle m_x' \rangle = L$ in accordance with (1.19), and the expressions obtained for θ_0 and φ_0 from (1.20). For an exact solution of the system (1.20), however, it is necessary to know the solutions of the system of differential equations (1.15), $u(\theta')$ and $v(\theta')$, which enter in the right-hand side of (1.20).

Multiplying the second equation of the system (1.15) by i and adding to the first, we obtain one equation for the complex function $z = u + iv$:

$$\frac{d^2 z}{d\theta'^2} + (\operatorname{ctg} \theta' - \sigma \sin \theta') \frac{dz}{d\theta'} - \left(\operatorname{csc}^2 \theta' - i \frac{H-\omega/\gamma}{\xi} \sigma \right) z = F_u + iF_v. \quad (2.1)$$

At $|\sigma \sin \theta'| \ll |\operatorname{cot} \theta'|$, this equation is transformed into the equation for spherical functions; in the general case, however, Eq. (2.1), to our knowledge, does not belong to any class of investigated differential equations. We shall therefore not deal with it; to obtain approximate solutions we shall use the equations for moments of higher degree.

A. Zeroth-approximation solution

The zeroth-approximation solution can be obtained by setting the cross correlators $\langle m_x' m_z' \rangle$ and $\langle m_y' m_z' \rangle$ equal to zero. We then determine θ_0 and φ_0 from (1.20) and, after substituting them in (1.13), we obtain for the mean deviations of the magnetization the resonant expressions:

$$\langle m_x \rangle = \frac{h}{H} \frac{L\omega_0(\omega_0 - \omega)}{(\omega_0 - \omega)^2 + (\eta\omega)^2}, \quad \langle m_y \rangle = -\frac{h}{H} \frac{L\eta\omega_0\omega}{(\omega_0 - \omega)^2 + (\eta\omega)^2}, \quad (2.2)$$

where ω_0 and the effective relaxation parameter η are functions of the temperature:

$$\omega_0 = \gamma H(1 + \eta^2), \quad \eta = \xi(L^{-1} - \sigma^{-1}). \quad (2.3)$$

In addition, the temperature dependence enters in (2.2) via the Langevin function L , which determines the temperature dependence (m_x). The average power of the high-frequency field, absorbed by a unit value of the superparamagnetic particle, is given by

$$P = \frac{h^2 M}{H} \frac{L\eta\omega_0\omega^2}{(\omega_0 - \omega)^2 + (\eta\omega)^2} = \frac{L\eta\omega^2 \gamma h^2 M}{(\gamma H - \omega)^2 + (\eta\gamma H)^2}. \quad (2.4)$$

The absorbed power has a maximum at the point $\omega = \omega_0$; the value of P at the maximum and the width of the resonance line at half its height are respectively given by

$$P_0 = \gamma h^2 M L(1 + \eta^2)/\eta, \quad \Delta\omega = 2\eta\omega. \quad (2.5)$$

B. First-approximation solution

To obtain a more exact solution we write down from (1.11) the equations for the second moments in the primed coordinate system:

$$\begin{aligned}
(H-\omega/\gamma)\langle m_y' m_z' \rangle + \xi H[2\langle m_x' m_z'^2 \rangle + \sin \theta_0(\langle m_x' \rangle - 2\langle m_x'^2 m_z' \rangle)] \\
- \xi h \cos \varphi_0(\langle m_x' \rangle - 2\langle m_x'^2 m_z' \rangle) + h \sin \varphi_0(\langle m_x'^2 \rangle - \langle m_x'^2 \rangle) + 6\xi \langle m_x' m_z' \rangle / \sigma = 0,
\end{aligned} \quad (2.6)$$

$$\begin{aligned}
(H-\omega/\gamma)[\langle m_x' m_z' \rangle + \sin \theta_0(\langle m_x'^2 \rangle - \langle m_y'^2 \rangle)] - 2\xi H \langle m_y' m_z' \rangle \\
+ \xi h \sin \varphi_0(\langle m_x' \rangle - 2\langle m_y'^2 m_z' \rangle) + h \cos \varphi_0(\langle m_x'^2 \rangle - \langle m_y'^2 \rangle) - 6\xi \langle m_y' m_z' \rangle / \sigma = 0.
\end{aligned}$$

In addition to the quantities determined by (1.19), these equations contain triple moments. Using the distribution function (1.18), we obtain

$$\langle m_x' m_y' m_z' \rangle = 0, \quad \langle m_x'^2 m_z' \rangle = \langle m_y'^2 m_z' \rangle = (1 - 3L/\sigma) / \sigma \quad (2.7)$$

(the first equation has already been substituted in (2.6)). The triple moments $\langle m_x' m_z'^2 \rangle$ and $\langle m_y' m_z'^2 \rangle$ are expressed in terms of integrals of the functions $u(\theta')$ and $v(\theta')$; neglecting them, we obtain from (2.6) expressions for

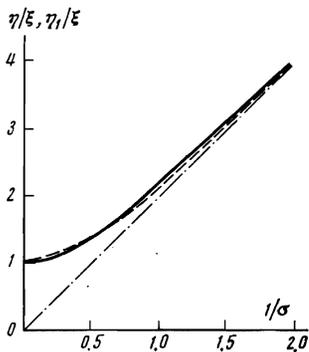


FIG. 3. Dependence of the effective relaxation parameters η (solid line) and η_1 (dashed line) on the temperature.

$\langle m_x' m_x' \rangle$ and $\langle m_y' m_y' \rangle$ and substitute them in (1.20). We can now obtain from (1.20) more accurate values of the angles θ_0 and φ_0 through which it is necessary to rotate the z' axis to coincide with the direction of the average magnetic moment of the ensemble. Substituting them in (1.13), we obtain for $\langle m_x \rangle$, $\langle m_y \rangle$, and P the expressions

$$\langle m_x \rangle = Lh \frac{bd+ac}{a^2+d^2}, \quad \langle m_y \rangle = Lh \frac{cd-ab}{a^2+d^2}, \quad (2.8)$$

$$P = -hM\omega \langle m_y \rangle,$$

where the quantities a , b , c , and d are determined by the expressions

$$a = H \left\{ \eta - \frac{\xi}{LD} \left[\left(H - \frac{\omega}{\gamma} \right)^2 \left(1 - \frac{3L}{\sigma} \right) + \frac{6(\xi H)^2 S}{\sigma} \right] \right\},$$

$$b = 1 + \frac{\xi^2 H}{LD} \left[\left(H - \frac{\omega}{\gamma} \right) S - \frac{6H}{\sigma} \left(1 - \frac{3L}{\sigma} \right) \right],$$

$$c = \eta - \frac{\xi H}{LD} \left[\left(H - \frac{\omega}{\gamma} \right) \left(1 - \frac{3L}{\sigma} \right) + \frac{6\xi^2 H}{\sigma} S \right], \quad (2.9)$$

$$d = \left(H - \frac{\omega}{\gamma} \right) \left\{ 1 - \frac{(\xi H)^2}{LD} \left[S - \frac{6}{\sigma} \left(1 - \frac{3L}{\sigma} \right) \right] \right\},$$

$$D = (H - \omega/\gamma)^2 + (6\xi H/\sigma)^2, \quad S = L - 2/\sigma + 6L/\sigma^2.$$

An investigation of the function $P(\omega)$ shows that the resonant frequency ω_0 and the line width $\Delta\omega$ are determined in the first approximation by the expressions

$$\omega_0 = \gamma H (1 + \eta_1^2), \quad \Delta\omega = 2\omega\eta_1, \quad (2.10)$$

where the effective relaxation parameter η_1 takes the form

$$\eta_1 = \xi \frac{8 - \sigma L - 12L/\sigma}{9L - \sigma}. \quad (2.11)$$

This expression differs significantly in form from the effective zeroth-approximation relaxation parameter. Actually, however, the difference between them is small in the entire temperature interval. Figure 3 shows plots of the temperature dependence of η and η_1 . The curves hardly differ from each other at all values of T . At high temperatures, the effective relaxation parameters tend to a common asymptote

$$\frac{\eta}{\xi} = 2 \frac{kT}{MHV} + \frac{1}{5} \frac{MHV}{kT} + \dots, \quad \frac{\eta_1}{\xi} = 2 \frac{kT}{MHV} + \frac{1}{6} \frac{MHV}{kT} + \dots \quad (2.12)$$

At low temperatures the expansion takes the form

$$\frac{\eta}{\xi} = 1 + \left(\frac{kT}{MHV} \right)^2 + \dots, \quad \frac{\eta_1}{\xi} = 1 + 3 \left(\frac{kT}{MHV} \right)^2 + \dots \quad (2.13)$$

The difference in the numerical coefficient preceding T^2 , as seen from Fig. 3, does not cause an appreciable difference between η_1 and η . Thus, in the employed method even the zeroth approximation yields a satisfactory analytic expression.

The successive approximation process can be easily continued. To find the second-approximation solution we write down the equations for the third moments in the primed system, etc.

For a linearly-polarized high-frequency field $P_{1,p}$, the absorbed power is given by the expression

$$P_{1,p} = \frac{1}{2} [P(\omega) + P(-\omega)], \quad (2.14)$$

where $P(\omega)$ corresponds to (2.4) in the linear approximation and to (2.8) in the first approximation.

3. EQUATION OF MOTION OF THE MAGNETIZATION. SUSCEPTIBILITY IN A LINEARLY POLARIZED FIELD

The Fokker-Planck equation (1.3) can be rewritten in vector form

$$2\tau \hat{W} = -i\hat{J} \{ -i\hat{J} + \xi^{-1}\sigma + [\mathbf{m} \times \sigma] \} W, \quad (3.1)$$

where $\hat{J} = -i[\mathbf{m} \times \partial/\partial \mathbf{m}]$ is an infinitesimally small rotation operator, $\mathbf{m} = \mathbf{M}/M$ is a unit vector of the magnetic moment, $\sigma = (MV/kT)\mathcal{H}$ is the dimensionless external field, and

$$\tau = MV/2\xi\gamma kT \quad (3.2)$$

is the characteristic time of the orientational diffusion of the magnetic moment of the particle (see^[2]).

In a constant field $\mathcal{H} = H$, a stationary normalized solution of (3.1) is the Gibbs distribution (1.9):

$$W_0 = \sigma e^{m\sigma}/4\pi \text{sh } \sigma. \quad (3.3)$$

Averaging the vector \mathbf{m} with the function W_0 leads to the known expression for the equilibrium magnetic moments of the particle:

$$\langle \mathbf{m} \rangle_0 = \int \mathbf{m} W_0 d^3\mathbf{m} = L(\sigma)\mathbf{n}, \quad \mathbf{n} = \mathbf{H}/H.$$

In the non-equilibrium situation, the macroscopic magnetic moment of the superparamagnet $\langle \mathbf{m} \rangle$ is determined by the average of the "microscopic" variable \mathbf{m} with a distribution function W satisfying Eq. (3.1). From (3.1) we can obtain for $\langle \mathbf{m} \rangle$ the equation

$$2\tau \frac{d\langle \mathbf{m} \rangle}{dt} = -\frac{1}{\xi} [\langle \mathbf{m} \rangle \times \sigma] - 2\langle \mathbf{m} \rangle - \langle [\mathbf{m} \times [\mathbf{m} \times \sigma]] \rangle, \quad (3.4)$$

which, however, is not closed and is only the first link of an infinite chain of coupled equations for the higher moments of the distribution function.

We confine ourselves below to the single-moment approximation, which is permissible in the case of small

deviations from equilibrium. In this approximation, to close the equation (3.4) we must put

$$W = W_0(1 + \alpha \mathbf{m}), \quad (3.5)$$

where W_0 is the equilibrium distribution function defined by (3.3), the vector α is independent of \mathbf{m} , and $(\alpha \cdot \mathbf{m}) \ll 1$. Averaging \mathbf{m} with the function W from (3.5) we have

$$\langle m_i \rangle = \langle m_i \rangle_0 + \alpha_k (\langle m_i m_k \rangle_0 - \langle m_i \rangle_0 \langle m_k \rangle_0). \quad (3.6)$$

The angle brackets with the subscript 0 will henceforth denote mean values calculated with the equilibrium function W_0 .

Under FMR conditions, the non-equilibrium part of the magnetization of the particle $\mu = M[\langle \mathbf{m} \rangle - \langle \mathbf{m} \rangle_0]$ (see (3.6)) is the result of the radio-frequency field with amplitude $h \ll H$ (in dimensionless form, $\sigma_1 \ll \sigma$, where $\sigma_1 = MVh/kT$). Therefore the vectors α and σ_1 should be regarded as quantities of the same order. Taking this into account, we obtain, upon substitution of (3.5) in (3.4), the following inhomogeneous equation for the parameter α :

$$2\tau[\langle m_i m_k \rangle_0 - \langle m_i \rangle_0 \langle m_k \rangle_0] \frac{d\alpha_k}{dt} = -2[\langle m_i m_k \rangle_0 - \langle m_i \rangle_0 \langle m_k \rangle_0] \alpha_k - (\sigma/\xi) e_{ik} n_i [\langle m_k m_m \rangle_0 - \langle m_k \rangle_0 \langle m_m \rangle_0] \alpha_m - \sigma n_k [\langle m_i m_k m_l \rangle_0 - \langle m_i m_k \rangle_0 \langle m_l \rangle_0] \alpha_l + \sigma_{ii} - \langle m_i m_k \rangle_0 \sigma_{ik} - \xi^{-1} e_{ik} \langle m_k \rangle_0 \sigma_{ii}.$$

From this we easily obtain with the aid of (3.6) an equation for the nonequilibrium part of the magnetization

$$\dot{\mu} = -\gamma[\mu \times \mathbf{H}] - \frac{1}{H^2 \tau_{\parallel}} \mathbf{H}(\mu \mathbf{H}) - \frac{1}{H^2 \tau_{\perp}} [\mathbf{H} \times [\mu \times \mathbf{H}]] + \frac{M_0}{H} \left\{ \gamma[\mathbf{h} \times \mathbf{H}] + \frac{1}{H^2 \tau} \mathbf{H}(\mathbf{h} \mathbf{H}) + \frac{1}{H^2 \tau_{\perp}} [\mathbf{H} \times [\mathbf{h} \times \mathbf{H}]] \right\}, \quad (3.7)$$

where $M_0 = ML(\sigma)$. Equation (3.7) takes the form of a Bloch equation linearized in μ and \mathbf{h} , with two relaxation times²⁾ for the magnetization components parallel and perpendicular to the constant field

$$\tau_{\parallel} = \frac{d \ln L}{d \ln \sigma} \tau, \quad \tau_{\perp} = \frac{2L}{\sigma - L} \tau. \quad (3.8)$$

At $\sigma \ll 1$ it follows from (3.8) that

$$\tau_{\parallel} \approx (1 - 2/\sigma^2) \tau, \quad \tau_{\perp} \approx (1 - 1/\sigma^2) \tau,$$

and at $\sigma \gg 1$

$$\tau_{\parallel} = 1/2 \tau_{\perp}, \quad \tau_{\perp} = 2\tau/\sigma = (\xi \omega_H)^{-1}, \quad \omega_H = \gamma H.$$

Thus, in weak fields ($H \ll kT/MV$) the magnetization relaxation time depends only on the volume and the temperature of the particles: $\tau_{\parallel} \approx \tau_{\perp} \approx \tau \sim V/T$. To the contrary, when the inverse inequality is satisfied ($H \gg kT/MV$), the time of damping of the free precession of the magnetic moment of the particles in the field H is determined only by the field intensity $\tau_{\perp} \sim \tau_{\parallel} \sim H^{-1}$. It follows therefore that with decreasing ratio V/T (at fixed H), the magnetization relaxation time also decreases,

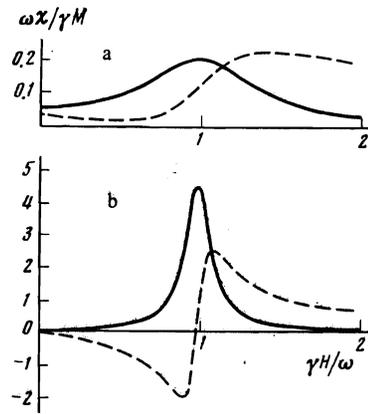


FIG. 4. Dynamic susceptibility of a superparamagnet in a linearly polarized field; dashed lines— χ' , solid— χ'' ; a) $\beta = 0.5$, b) $\beta = 10$. In the calculations it was assumed that $\xi = 0.1$.

and it is this which leads to the broadening of the absorption line.

Let us calculate the dynamic susceptibility in a linearly polarized field $h \sim e^{i\omega t}$. Putting in (3.7) $\mathbf{h} \perp \mathbf{H}$ and $\mu_{\parallel} = \chi_{\parallel}(\omega) h_{\parallel}$, we obtain for the magnetic susceptibility in the direction of the radio-frequency field the expression

$$\chi = \frac{ML}{H} \frac{\omega_H^2(1+\eta^2) + i\eta\omega\omega_H}{\omega_H^2(1+\eta^2) - \omega^2 + 2i\eta\omega\omega_H} \quad (3.9)$$

where η is determined by formula (2.3): in the derivation of (3.9) we use the relation $\tau_{\perp} = (\eta\omega_H)^{-1}$, which results from (2.3), (3.2), and (3.8).

If $\sigma \gg 1$ (strong field fields or massive particles), then $l \approx 1$, $\eta \approx \xi$, and formula (3.9) coincides with the known expression for the dynamic susceptibility, which is obtained directly from the Landau-Lifshitz equation. In the case $\sigma \ll 1$ (weak fields or minute particles), the susceptibility loses its resonant character; substituting in (3.9) the value $\eta = 2\xi/\sigma$ obtained from (2.3) in the limit of small σ , we get

$$\chi = \frac{M^2 V}{3kT} \frac{1}{1 + i\omega\tau} \quad (3.10)$$

with τ from (3.2). Thus, with decreasing parameter $\sigma = MVH/kT$, the high-frequency field energy absorption line is deformed and changes from a typically resonant (Lorentz) shape at $\sigma \gg 1$ into a pure relaxation (Debye) shape at $\sigma \ll 1$.

In FMR observations, the experimental situation is such that the operating frequency ω of the instrument is fixed, and the controlled parameters are the magnetizing field H and the temperature T . Figure 4 shows, as functions of the dimensionless field intensity $\gamma H/\omega$, the real and imaginary parts χ' and χ'' of the complex susceptibility, calculated from formula (3.9) for two values of the dimensionless reciprocal temperature $\beta = \omega MV/\gamma kT$; the value $\beta = 1$ is obtained at room temperatures, $M = 10^3$ G, and $\omega = 10^{10}$ sec⁻¹ for particles of diameter 50 Å. The non-Lorentzian character of the suscepti-

bility at small β is particularly clearly seen from the χ'' lines—at $\beta < 0.65$ these curves have no nodes at all (see Fig. 4a).

Calculation of the susceptibility lines shows that the resonant field H_r , corresponding to the maximum value of the imaginary part of the susceptibility

$$\chi_r''(H_r) = \max\{\chi''(H)\},$$

depends little on the parameter β . Much more sensitive to changes of this parameter are the quantity χ_r'' and the absorption line width ΔH at the height $\chi_r''/2$.

In the limit as $\beta \rightarrow \infty$ ($T/V \rightarrow 0$) the line shape is determined by the Landau-Lifshitz equation, and therefore

$$\chi_r'' = \gamma M / 2\xi\omega, \quad \Delta H / H_r = 2\xi.$$

With decreasing volume of the particle (with increasing ratio T/V), the height of the absorption peak decreases monotonically, and the relative line width increases without limit.

It appears that the arguments advanced above can explain the results of Bagguley's experiment.^[6] In an investigation of dilute colloidal suspensions of spherical ferromagnetic-metal particles (with linear dimensions on the order of 100 Å), he observed exceedingly broad (compared with those obtained with bulky samples) FMR lines: in a field $H \approx 3000$ Oe the width ΔH was 1400 Oe

for iron and 1000 Oe for nickel, exceeding by one order of magnitude the anisotropic fields of the indicated metals. In both cases, the gyromagnetic ratio determined from the position of the resonance peak did not differ (with experimental accuracy) from that measured on a bulky sample.

¹*Editor's note:* This article is a consolidation, made at our request, of two almost simultaneously submitted papers.

²Formulas perfectly similar to (3.8) are obtained also for the relaxation times of the magnetization of a suspension of rigid dipoles.^[5] The "superparamagnetism" of such a suspension is ensured by the rotational diffusion of the particles in a liquid with viscosity η ; therefore in place of τ from (3.2) the formulas of^[5] contain the Brownian time $\tau_B = 3\eta N/kT$. As seen from a comparison of τ_B with τ , in the case considered in the present article the role of viscosity is played by the quantity $M/6\xi\gamma$ (magnetic viscosity).

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Temperature dependence of the density distribution of the superfluid component of helium II in pores

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An oscillating-disk method is proposed for the direct determination of the density of the superfluid component in pores. The validity of the $\langle\rho_s\rangle/\rho_{sb}$ distribution derived by Kiknadze and Mamaladze is demonstrated. It is established that near T_0 the density of the superfluid component of helium II in pores varies linearly with temperature ($\langle\rho_s\rangle \sim T_0 - T$), whereas far from T_λ it is described by the law $\langle\rho_s\rangle \sim (T_\lambda - T)^{2/3}$.

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The density distribution predicted by the Ginzburg-Pitaevskii-Mamaladze (GPM) phenomenological theory^[1,2] for the superfluid component of helium II in pores has been experimentally studied by quite a large number of foreign investigators,^[3-8] who used, in the main, fourth-sound and gyroscopic techniques and studied the law of variation with temperature of the mean superfluid density $\langle\rho_s\rangle$ in pores. In^[9,10], Kiknadze and Mamaladze derived an analytic expression for $\langle\rho_s\rangle/\rho_{sb}$ (ρ_{sb} is the bulk superfluid-component density) as a function of temperature and the geometry of the vessel.

The present paper is devoted to the study of these problems in pores of cylindrical geometry by an oscillating-disk method. The proposed method enables us to obviate a number of difficulties encountered in fourth-sound experiments. We have in mind, for example, the correction for the scattering of sound by the grains of packed powder, the necessity for normalization of the temperature-dependence curves for the fourth-sound velocity, the application of a low-frequency signal for an independent determination of the transition temperature.