LIMITING VISCOSITY OF FERROMAGNETIC SUSPENSIONS IN A STRONG MAGNETIC FIELD

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The effective viscosity of a suspension of ferromagnetic particles with anisotropy of the "easy-axis" type is calculated. The case considered is that in which the magnetic field in which the suspension is flowing greatly exceeds the internal anisotropy field, and the concept of the "frozen" magnetic moments of the particles is inapplicable. The relationship between the viscosity and the anisotropy field is established. The question as to the magnitude of the viscosity for an arbitrary ratio of the internal and external fields is discussed.

1. The concept of rotational viscosity developed in [1] enables us to explain the experimentally observed [2, 3] increase in the viscosity of suspensions of ferromagnetic particles under the influence of an external magnetic field. The reason for this effect is as follows. In the absence of a field the angular velocity of rotation of the particles suspended in the flow $\boldsymbol{\Omega} = {}^1/_2$ rot **v** is equal to the local angular velocity of rotation of the liquid $\boldsymbol{\Omega} = {}^1/_2$ rot **v**. The viscosity is in this case described by the Einstein equation

$$\eta = \eta_0\left(1 + \frac{5}{2}\varphi\right) \tag{1.1}$$

where η_0 is the viscosity of the carrier liquid, φ is the volumetric concentration of the solid phase. On placing the suspension in a uniform magnetic field H the latter exerts an orientation effect on the magnetic moments of the particles μ, preventing the free rotation of the particles in the vortical flow. The difference in angular velocities so arising $\Omega - \omega$ corresponds to the moment of the frictional forces $6\eta_0 V(\Omega - \omega)$, where V is the volume of one spherical particle. This additional internal friction manifests itself as an increase in the effective viscosity of the suspension: $\eta_e = \eta + \eta_r$, where η_r is the rotational viscosity.

The quantity η_r depends considerably on the magnetocrystalline-anisotropy energy of the ferromagnetic material, which determines the interaction between the magnetic moment μ and the rotational degrees of freedom of the particle. In the absence of such an interaction (model of "free" magnetic dipoles) the orientations of the vector μ in the direction of the field H does not interfere with the free rotation of the particle in the flow ($\boldsymbol{\omega} = \boldsymbol{\Omega}$), so that in this model $\eta_r = 0$. In the limiting case of strong interaction, when the magnetic moment is rigidly connected to the body of the particle (model of "frozen" dipoles), an applied field greatly impedes its rotation, and for a sufficiently strong field the rotation of the particle in the flow stops completely, being replaced by sliding ($\omega = 0$) along the corresponding shear plaen. The rotational viscosity η_r(H) then reaches its limiting value (saturation effect) equal to [4]

$$\eta_r(\infty) = {}^3/_2\eta_0\varphi$$

In real ferromagnetic crystals a type of relationship intermediate between the models fo free and frozen dipoles is established between the direction of the vector μ and the crystallographic axes of the particle: For a finite magnetic-anisotropy energy we may only speak of a partial freezing of the magnetic moment. However strong the external field, even if it maintains the orientation of the moments μ absolutely constant, it cannot completely prevent the rotation of the particles due to hydrodynamic forces.

The limiting value of the viscosity $\eta_r(\infty)$ of any real magnetic suspension should lie in the range

$$0 < \eta_r(\infty) < {}^3/_2\eta_0\varphi$$

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We shall now calculate $\eta_r(\infty)$ for a suspension of ferromagnetic particles with anisotropy of the "easy-axis" type.

2. In stable magnetic colloids ferromagnetic particles with linear dimensions $\mu \sim 10^{-5}$–10^{-6} cm are generally employed. For the dimensions indicated each particle forms an individual uniformly magnetized domain with a dipole moment $\mu = M_S V$, where M_S is the saturated magnetization of the particle material. The energy of such a particle in an external field is determined by the equation

$$U = -\mu H\,(\mathbf{e}\cdot\mathbf{h}) - KV\,(\mathbf{e}\cdot\mathbf{n})^2, \quad \mathbf{e} = \boldsymbol{\mu} / \mu, \quad \mathbf{h} = \mathbf{H} / H \tag{2.1}$$

where K is the energy density of magnetocrystalline anisotropy, **n** is the unit vector in the direction of the axis of easy magnetization.

Every deviation of the vector e from the equilibrium orientation defined by the direction of the effective field

$$\mathbf{H}_e = -\mu^{-1}\frac{\partial U}{\partial \mathbf{e}} = H\mathbf{h} + 2KM_S^{-1}\mathbf{n}\,(\mathbf{e}\cdot\mathbf{n}) \tag{2.2}$$

is accompanied by a Larmor precession of the magnetic moment μ around $\mathbf{H}_e$. It should be remembered that the Larmor-precession attenuation time ($\lesssim 10^{-9}$ sec) is small compared with any hydrodynamic times; hence at every instant of time the internal state of the particle may be regarded as equilibrium ($\mathbf{e}\|\mathbf{H}_e$). We see from (2.2) that the magnitude and direction of $\mathbf{H}_e$ are determined by the vector sum of the external field H and the anisotropy field $H_A = 2K/M_S$. Hence for $H_A \gg H$ it is permissible to assume a frozen-in state of the moment ($\mathbf{e}\| \mathbf{n}$), as in [1, 5]. Omitting the inessential constant in the expression for U we obtain $U = -\mu H(\mathbf{nh})$ from (2.1). The moment of the magnetic forces acting on the particle

$$\mathbf{m} = -(\mathbf{n} \times \partial U / \partial \mathbf{n}) \tag{2.3}$$

is then equal to $(\mu \times' \mathrm{H})$.

For $K = 0$ ($H_A = 0$) we have an "isotropic" magnetic crystals in which the relationship between the magnetic and the mechanical degrees of freedom is broken (model of free dipoles). In this case the rotating moment $\mathbf{m}=0$, since the magnetic energy $U = -(\mu \mathrm{H})$ does not depend on the orientation **n** of the easy axis of the particle.

In order to determine the limiting viscosity of a suspension of ferromagnetic particles with finite anisotropy in a strong external field we must consider the case in which

$$H \gg H_A, \quad H_A = 2K / M_S \tag{2.4}$$

It follows from the condition of equilibrium $\mathbf{e}\|\mathbf{H}_e$ and Eq. (2.2) for the effective field $\mathbf{H}_e$ that on satisfying inequality (2.4) the magnetic moment of the particle may be regarded as parallel to the applied field: $\mathbf{e}=\mathbf{h}$. For the magnetic energy of the particle (2.1) and the rotating moment (torque) (2.3) we than obtain

$$U = -KV\,(\mathbf{n}\cdot\mathbf{h})^2, \quad \mathbf{m} = 2\;KV\,(\mathbf{n}\cdot\mathbf{h})\,(\mathbf{n}\times\mathbf{h}) \tag{2.5}$$

Subsequently instead of the pseudovectors we shall frequently use their dual antisymmetric tensors; for example, for (2.5) we have

$$m_{ik} = \varepsilon_{ikl} m_l = 2KV\,(n_i h_k - n_k h_i)\, n_l h_l \tag{2.6}$$

3. In the hydrodynamic description of the suspension as a homogeneous continuous medium, in order to allow for the rotational degrees of freedom of the particles we have to introduce an additional macroscopic variable S, the volume density of the internal moment of momentum. The latter has the sense of the product I $\langle\omega\rangle$, where I is the sum of the intrinsic moments of inertia of the spherical particles in unit volume of the suspension, $\langle\omega\rangle$ is the mean angular velocity of their rotation. The stress tensor of a medium with internal rotation contains the antisymmetrical part [6]

$$\sigma_{ik}{}^a = \varepsilon_{ikl}\sigma_l, \quad \boldsymbol{\delta} = (2\tau_S)^{-1}\,(\mathrm{S} - I\Omega) \tag{3.1}$$

where $\tau_S = a^2\rho/15\eta_0$ is the relaxation time of the internal moment ρ is the density of the particle. The change in the density of the internal moment of mementum is described by the equation

$$\frac{d\mathrm{S}}{dt} = -\frac{1}{\tau_S}(\mathrm{S} - I\Omega) + \mathbf{M} \tag{3.2}$$

where M is the volume density of the moment of the lateral forces acting directly on the particles and maintaining the difference between the angular velocities of rotation of the particles $\langle\omega\rangle$ and the liquid Ω: For M = O the relaxation of S to the equilibrium value $I\Omega$, takes place in a very short time, of the order of τ_S, and the stress tensor in symmetrical ($\sigma = 0$).

In the case under consideration the expression for M should be obtained by averaging the "microscopic" torque (2.6) with respect to the orientations of the easy axes of the particles

$$M_{ik} = N \langle m_{ik}\rangle = N_{il}B_{kl} - N_{kl}B_{il} \quad (3.3)$$
$$N_{ik} = N \langle n_i n_k\rangle, \quad B_{ik} = 2KVh_ih_k$$

(N is the number of particles in unit volume of the suspension). Since the two directions of the axis of easy magnetization are equivalent, the degree of orientation of the suspension consisting of such "quadrupole" particles is characterized by a symmetrical "orientation tensor" N_{ik}; the intensity of the factor creating a preferred orientation is determined by the "anisotropy tensor" B_{ik}. We may indicate an analogy between (3.3) and the mechanical moment $M_{ik} = I_iH_k - I_kH_i$, acting on a suspension of "rigid" dipoles: In the present case N_{ik} and B_{ik} play the same role as the magnetization I_i and the external field H_i, respectively, in the model of frozen magnetic moments.

To the equation

$$\frac{d}{dt} S_{ik} = -\frac{1}{\tau_S}(S_{ik} - I\Omega_{ik}) + (N_{il}B_{kl} - N_{kl}B_{il}) \quad (3.4)$$

obtained by substituting (3.3) into (3.2) we must add the equation of motion of the tensor N_{ik}. In order to derive the missing equation we make use of the following considerations. In a system of reference rotating at an angular velocity $\langle\omega\rangle$ in which the average rate of rotation of the particles equals zero, any deviation of the density of the orientation N_{ik} from the equilibrium value N_{ik}° should vanish as time progresses. Let us assume that this vanishing takes place in accordance with a relaxation law

$$\frac{d'}{dt} N_{ik} = -\frac{1}{\tau}(N_{ik} - N_{ik}^\circ) \quad (3.5)$$

where d'/dt is the derivative in the rotating system of reference τ is the time of orientational relaxation. The rates of change of the symmetrical tensor of the second rank in the stationary and rotating systems of reference are related by the kinematic equation

$$\frac{d}{dt} N_{ik} = -(\langle\omega_{il}\rangle N_{kl} + \langle\omega_{kl}\rangle N_{il}) + \frac{d'}{dt} N_{ik} \quad (3.6)$$

Substituting $\langle\omega_{ik}\rangle = I^{-1}S_{ik}$ and (3.5) into (3.6), we have

$$\frac{d}{dt} N_{ik} = -\frac{1}{\tau}(N_{ik} - N_{ik}^0) - \frac{1}{I}(S_{il}N_{kl} + S_{kl}N_{il}) \quad (3.7)$$

The orientations of the easy axes in the direction of the applied field prevents the thermal (Brownian) motion of the particles. Under these conditions the relaxation time τ should be identified with the Brownian rotational-diffusion time, i.e., for quadrupole particles $\tau = r_0V/kT$, while the equilibrium orientational distribution coincides with the canonical distribution

$$W_0 \sim \exp(-U / kT)$$

(k is Boltzmann's constant, T is the absolute temperature). Using Eq. (2.5) for U, we obtain a normalized distribution function

$$W_0 = (4\pi F)^{-1} \exp[\lambda (\mathbf{h}\cdot\mathbf{n})] \quad (3.8)$$
$$\lambda = KV / kT, \quad F(\lambda) = \int_0^1 e^{\lambda x^2} dx$$

For the components of the equilibrium orientation tensor

$$N_{ik}^\circ = N \langle n_i n_k\rangle_0 = N \int n_i n_k W_0 d^3\mathbf{n}$$

calculation gives

$$N_{ik}^\circ = N(F_1\delta_{ik} + F_2h_ih_k) \quad (3.9)$$
$$F_1 = \frac{1}{2}\left(1 - \frac{F'}{F}\right), \quad F_2 = \frac{3}{2}\left(\frac{F'}{F} - \frac{1}{3}\right), \quad F' = \frac{dF}{d\lambda}$$

The dimensionless parameter λ, the ratio of the magnetic anisotropy energy of the particle to the energy of its thermal motion, appears here as an analog of the Langevin argument $\xi = \mu H/kT$ in the model of frozen dipoles [1, 5]. We remember that this model corresponds to the assumption $\lambda \gg \xi$, while in the present analysis we have taken the opposite limiting case ($\xi \gg \lambda$).

Let us give the asymptotic formulas for the statistical integral $F(\lambda)$. For small λ this may be expressed in the form of a series

$$F = \sum_{n=0}^{\infty} \frac{\lambda^n}{n!\,(2n+1)}, \qquad \lambda \ll 1 \tag{3.10}$$

In order to find the asymptotic for large λ we note that F satisfies the equation

$$F' = (2\lambda)^{-1}(e^{\lambda} - F)$$

Taking account of this we obtain the equation

$$F = \frac{e^{\lambda}}{2\lambda}\left[1 + \sum_{n=1}^{\infty} \frac{(2n-1)!!}{2^n \lambda^n}\right], \qquad \lambda \gg 1 \tag{3.11}$$

It follows from (3.9)-(3.11) that for $\lambda = 0$ the suspension is isotropic ($N_{ik}^{\circ} = {}^1/_3 N\delta_{ik}$) while for $\lambda \to \infty$ the anisotropies of all the particles are parallel to the applied field ($N_{ik}^{\circ} = Nh_i h_k$).

4. In the steady-state case ($d/dt = 0$) Eqs. (3.4) and (3.7) take the form

$$\begin{aligned} S_{ik} - I\Omega_{ik} &= \tau_S (N_{il} B_{kl} - N_{kl} B_{il}) \\ N_{ik} - N_{ik}^{\circ} &= -\tau I^{-1} (S_{il} N_{kl} + S_{kl} N_{il}) \end{aligned} \tag{4.1}$$

Equation (4.1) enables us to eliminate S_{ik} from Eq. (3.1) for the antisymmetric part of the stress tensor. We obtain:

$$\sigma_i = (KV/4)\, \varepsilon_{ikl} (N_{km} B_{lm} - N_{lm} B_{km}) \tag{4.2}$$

$$N_{ik} - N_{ik}^{\circ} = -\tau (\Omega_{il} N_{kl} + \Omega_{kl} N_{il}) (\lambda / 3N) [N_{lm} h_l (N_{im} h_k + N_{km} h_i) - 2N_{il} N_{km} h_l h_m] \tag{4.3}$$

Coming to the solution of Eq. (4.3) we note that owing to the smallness of τ (10^{-5}-10^{-6} sec) the condition $\Omega\tau \ll 1$ is satisfied for all values of $\mathbf{\Omega}$ of practical interest. In still liquid ($\mathbf{\Omega} = 0$) the solution to Eq. (4.3) is $N_{ik} = N_{ik}^{\circ}$. The orientation tensor of the particles in a moving liquid differs from N_{ik}° as defined by Eqs. (3.9) but the difference $N_{ik} - N_{ik}^{\circ} = n_{ik}$ is small owing to the smallness of $\Omega\tau$. Regarding n_{ik} and $\Omega\tau$ as quantities of the same order and using the linear approximation for (4.3) we obtain

$$n_{ik} = -\frac{N\tau F_2}{1 + {}^1/_3\lambda F_2} (\Omega_{il} h_k + \Omega_{kl} h_i) h_l \tag{4.4}$$

Substituting the resultant value of N_{ik} into (4.2) we obtain

$$\begin{aligned} \boldsymbol{\sigma} &= 2\eta_r [\mathbf{h} \times (\mathbf{h} \times \Omega)], \quad \eta_r = {}^3/_2\, \eta_0 \varphi f_1(\lambda) \\ f_1 &= \frac{\lambda}{2}\left(\frac{F'}{F} - \frac{1}{3}\right)\left[1 + \frac{\lambda}{2}\left(\frac{F'}{F} - \frac{1}{3}\right)\right]^{-1} \end{aligned} \tag{4.5}$$

The coefficient η_r is the rotational viscosity [1, 7, 8]. On incorporating this into the Einstein formula (1.1) we have, to a first order accuracy in the concentration,

$$\eta_e = \eta_0 \left\{1 + \varphi\left[\frac{5}{2} + \frac{3}{2} f_1(\lambda) \sin^2\alpha\right]\right\} \tag{4.6}$$

where α is the angle between the vectors **h** and $\mathbf{\Omega}$. For $\mathbf{h} \parallel \mathbf{\Omega}$ the rotational viscosity does not appear, since in this case the orientation of the particle anisotropy axis along **h** does not prevent the particle from rotating at a velocity $\mathbf{\Omega}$ around the same axis.

A curve of $f_1(\lambda)$ is presented in Fig. 1 (continuous line). The asymptotic of this function may be found from (3.10) and (3.11)

$$f_1(\lambda) = \begin{cases} {}^2/_{45}\lambda^2, & \lambda \ll 1 \\ 1 - 3/\lambda, & \lambda \gg 1 \end{cases} \tag{4.7}$$

5. As indicated in § 4, in order to determine the rotational viscosity of a suspension we must express the antisymmetric part of the stress tensor (4.2) in the form (4.5). This may be done by eliminating the orientation tensor of the moving suspension N_{ik} from (4.2), this having been found in § 3 and 4 phenomenologically with the help of Eq. (3.7).

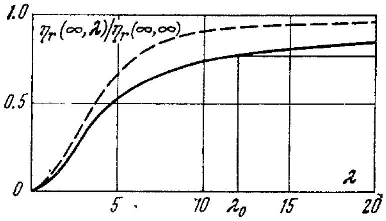

Fig. 1

In order to refine the dependence of the rotational viscosity on the anisotropy parameter λ let us calculate N_{ik} directly from the kinetic equation; for the Brownian particles of the suspension this means the Fokker–Planck equation. In considering the rotational diffusion this equation may be conveniently put in the form [9]

$$\frac{\partial W}{\partial t} + (\mathbf{L}\boldsymbol{\omega}) W = 0 \qquad \mathbf{L} \equiv \mathbf{n} \times \frac{\partial}{\partial \mathbf{n}} \tag{5.1}$$

where W(**n** t) is the probability density of the directions of the particle anisotropy axis, **L** is the operator of an infinitely small rotation, ω is the angular velocity of the particle. This latter is determined (neglecting inertia) from the condition that the sum of all the mechanical moments acting on the particle suspended in the liquid should vanish:

$$-\mathbf{L}U + 6\eta_0 V(\boldsymbol{\Omega} - \boldsymbol{\omega}) - kT\mathbf{L} \ln W = 0 \tag{5.2}$$

The first two terms in (5.2) are the moments of the regular forces, magnetic (2.5) and viscous, while the third term is the moment of the random forces. From (5.2) we obtain

$$\boldsymbol{\omega} = (6\tau)^{-1} [6\tau\boldsymbol{\Omega} + 2\lambda (\mathbf{nh}) (\mathbf{n} \times \mathbf{h}) - \mathbf{L} \ln W] \qquad (5.4)$$

Here we have used the notation introduced earlier: $\tau = \eta_0 V/kT$, $\lambda = KV/kT$. Substituting (5.3) into (5.10) we obtain the following equation for the steady-state distribution function

$$\mathbf{L} [\mathbf{L} - 2\lambda (\mathbf{nh}) (\mathbf{n} \times \mathbf{h}) - 6\tau\boldsymbol{\Omega}] W = 0 \qquad (5.4)$$

For a stationary suspension (Ω = 0) the Gibbs distribution (3.8) forms the solution W_0 to Eq. (5.4). Hence in the moving liquid (allowing for the smallness of $\Omega\tau$, we may conveniently seek the distribution function in the form

$$W = W_0(1 + \chi), \quad \langle\chi\rangle_0 = \int \chi W_0 d^3\mathbf{n} = 0 \tag{5.5}$$

where χ is a quantity of order $\Omega\tau$. We earlier mentioned the "two-sidedness" of the anisotropy axis: W is invariant with respect to the replacement of n by −**n**. It follows that the expansion of the scalar function χ in powers of the vector n may only contain even products of its components. In the first nonvanishing order

$$\chi = G_{ik} (\lambda, \ \mathbf{h}, \ \boldsymbol{\Omega}\tau) \, n_i n_k \tag{5.6}$$

Ω and **h** are pseudovectors. Taking account of this we may readily convince ourselves that the only true scalar of the form (5.6) linear with respect to $\Omega\tau$ is

$$\chi = g(\lambda) \, \tau \, (\mathbf{nh}) (\mathbf{n} \times \mathbf{h}) \cdot \boldsymbol{\Omega} \tag{5.7}$$

After substituting (5.5) and (5.7) into (5.4) the latter was multiplied by $n_i n_k$ and integrated. This method of determining the function g(λ) (analogous to the Chapman–Enskog method in the kinetic theory of gases) leads to the result

$$g = -2\lambda \left[1 + \frac{2\lambda}{3}\left(\frac{F'' - F'''}{F' - F''} - \frac{1}{2}\right)\right]^{-1} \tag{5.8}$$

Knowing the distribution function (5.5) we may calculate the correction n_{ik} to the tensor $N_{ik}{}^{\circ}$ due to the motion of the medium. Instead of (4.4) we find

$$n_{ik} = \langle n_i n_k \chi \rangle_0 = g(\lambda) (\Omega_{il} h_k + \Omega_{kl} h_i) h_l \tag{5.9}$$

with the g of (5.8). Making use of (5.9) we obtain the following for the rotational viscosity (see (4.5))

$$\eta_r = \frac{3}{2} \eta_0 \varphi f_2(\lambda), \quad f_2(\lambda) = \frac{\lambda}{2}\left(\frac{F'}{F} - \frac{1}{3}\right)\left[1 + \frac{2\lambda}{3}\left(\frac{F'' - F'''}{F' - F''} - \frac{1}{2}\right)\right]^{-1} \tag{5.10}$$

The asymptotics of the function f_2 are similar to (4.7)

$$f_2(\lambda) = \begin{cases} {}^2/_{45}\lambda^2, & \lambda \ll 1 \\ 1 - 1/2\lambda, & \lambda \gg 1 \end{cases} \tag{5.11}$$

The relationship $f_2(\lambda)$ is shown as the broken line in Fig. 1.

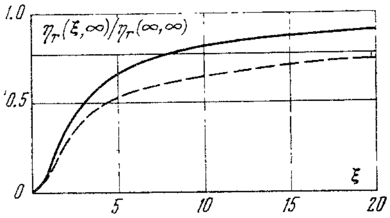

Fig. 2

For a suspension of particles with a cubic magnetocrystalline anisotropy an analogous calculation was presented in [10] for the case $\lambda \ll 1$. Instead of (5.11) the term $^8/_{175}\,\lambda^2$ was obtained. This is very close to the $^8/_{180}\,\lambda^2$ for the uniaxial crystal (5.11).

6. We may note the closeness of the functions $\eta_r(\lambda)$ determined by means of Eqs. (4.5) and (5.10) over the whole range of λ values. A similar situation held when calculating the magnetoviscosity of suspension of rigid dipoles ($\lambda \gg \xi$): the phenomenological result [1]

$$\eta_r(\xi) = \frac{3}{2}\eta_0\varphi\,\frac{\xi - \operatorname{th}\xi}{\xi + \operatorname{th}\xi}, \quad \xi = \mu H / kT \qquad (6.1)$$

was close to the kinetic expression [5].

For finite values of ξ and λ the rotational viscosity is a function of both these variables

$$\eta_r = \eta_r(\xi, \lambda)$$

The viscosity of the "magnetized" [in the sense of (2.4)] suspension calculated in this paper should be regarded as $\eta_r(\infty, \lambda)$, and $\eta_r(\xi)$ from (6.1) as $\eta_r(\xi, \infty)$. The curve of the latter is shown in Fig. 2 as a continuous line.

Every ferromagnetic suspension is characterized by a specific value of the parameter $\lambda = \lambda_0$, determined by the material and size of the particles. Also dependent on λ_0 (Fig. 1) is the limiting viscosity $\eta_r(\infty, \lambda_0)$, to which $\eta_r(\xi, \lambda_0)$ should tend asymptotically on increasing the external field. In the region $\xi \ll \lambda_0$ the functions $\eta_r(\xi, \lambda_0)$ and $\eta_r(\xi, \infty)$ should be close together. It may reasonably be expected that the broken curve in Fig. will give a qualitatively correct description of $\eta_r(\xi)$ for a fixed λ.

A comparison between experimental data relating to the magnetoviscosity of suspensions and the theoretical results may be used in order to determine such essentially "solid-state" parameters as the anisotropy constant and the saturated magnetization of a dispersed ferromagnetic material.

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