

# Theory of time-dependent magnetic phenomena for a fine-particle ensemble

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Theory of time-dependent magnetic phenomena for an ensemble of small ferromagnetic particles with uniaxial anisotropy – Stoner–Wohlfarth (SW) particles – is presented. Magnetic viscosity of such a system is calculated using the energy barrier distribution density  $\rho(E)$  for an SW ensemble. It is shown that, due to the nonmonotonic dependence of the energy barrier versus easy axis orientation angle,  $\rho(E)$  has essential singularities at least for one energy value. These singularities result in nonlinear temperature behavior of the maximum magnetic viscosity  $S_{\max}$  and in complex dependence of  $S_{\max}$  versus the applied field. It is also shown that the interparticle interactions and the particle size distribution eliminate the singularities in energy barrier density, so that  $S_{\max}$  increases linearly with the temperature. The dependence of  $S_{\max}$  on the particles volume concentration, anisotropy constant and width of the particle size distribution is studied.

## 1. Introduction

The importance of non-equilibrium kinetic remagnetization processes for both fundamental physical research and various practical applications of magnetic systems causes growing interest to these phenomena [1–6]. It is well known that the relaxation of the system with single-barrier energy  $E$  obeys the exponential law  $M(t) \sim \exp(-\gamma t)$ , where the relaxation speed  $\gamma$  is determined by the barrier energy as  $\gamma = \gamma_0 \exp(-E/T)$  with the constant  $\gamma_0 \sim 10^9 \text{ s}^{-1}$  for usual magnetic systems.

Real systems always have some distribution of the energy barriers, and do not demonstrate this exponential behavior. It was shown [6] that such distribution can lead to the so-called linear loga-

arithmic time dependence of the magnetization in the form

$$M(t) = C - S \ln t, \quad (1)$$

where  $S (= \text{Const})$  is called the magnetic viscosity. In spite that the equation (1) was derived by Street and Wooley for the very special energy barrier distribution density  $\rho(E)$  (in ref. [6]  $\rho(E) = \text{Const} > 0$  for  $E_{\min} < E < E_{\max}$  and  $\rho(E) = 0$  otherwise) and for limited time region, it works very well in many different experimental situations [3,5–8].

Later it was shown [3] that the constant magnetic viscosity approximation (1) is valid, if the energy barrier density  $\rho(E)$  does not change rapidly in the energy interval  $T \ln \tau_{\text{beg}} < E < T \ln \tau_{\text{end}}$ , where  $\tau = \gamma_0 t$  and  $t_{\text{beg(end)}}$  corresponds to the time of the measurement's beginning (end) after the external parameter (usually the applied field) was changed. Due to the logarithmic dependence on the measurement time this energy interval is relatively narrow even for very large time region (i.e., for  $t_{\text{beg}} = 10 \text{ s}$  and  $t_{\text{end}} = 10^4 \text{ s}$

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we have  $25T < E < 32T$ ) so that the condition for  $\rho(E)$  mentioned above is often satisfied very well.

Further attempts to connect the magnetic viscosity coefficient  $S$  with the physical properties of the system studied have led to the famous expression

$$S = T\rho(E_c), \quad (2)$$

where the critical energy  $E_c = T \ln \tau$  separates (in the first approximation) magnetization domains, which have already reversed their magnetization (with the energy barrier  $E < E_c$ ) during the time  $\tau$  from those which are still in the initial state. The expression (2) corresponds to the so called 'critical energy approach' [1,3,5] and is valid (similar to eq. (1)) for the barrier density, varying slowly in the corresponding energy interval.

Next approximation can be obtained expanding  $\rho(E)$  near the energy  $E_0 = T \ln \tau_{\text{beg}}$ . Using the relation  $E - E_0 = T \ln(\tau/\tau_{\text{beg}})$ , the logarithmic time series can be obtained for  $S$  in the form [9]

$$S = T\rho(E_0) + \sum_{k=1}^{\infty} A_k (\ln \tau/\tau_0)^k. \quad (3)$$

Expressions (1)–(3) can be used in practical calculations only if the energy barrier density  $\rho(E)$  for the system studied is known. It is usually not the case, so that one should assume a certain form of  $\rho(E)$  using general information about the system (i.e., the particle size distribution) and some more or less reasonable assumptions like quadratic dependence of the energy barrier on the applied field [1,2,4].

For this reason it would be very useful to develop a theory of the time-dependent magnetization processes for the magnetic system for which the energy barrier distribution density can be calculated exactly. This requires a good understanding of the hysteresis phenomena in this system to calculate the energy barrier dependence on the applied field and other magnetic parameters. Presently the most suitable system for this purpose seems to be an ensemble of fine single-domain magnetic particles with the uniaxial (shape or crystalline) magnetic anisotropy, which

is known as Stoner–Wohlfarth (SW) ensemble [10].

For the particles described above the exact dependence of the switching field on the easy-axis orientation angle is known, which enables to obtain an analytical expression for the energy barrier dependence versus external field and particle orientation angle [11]. Hence the density of energy barriers can be found for any given distribution of particles' easy axes, allowing to calculate the magnetic viscosity for this system from first principles. The interaction field distribution density for the SW ensemble is known as well [12], which enables to take into account interparticle interactions and particle size distribution also without any uncontrolled approximations. The following paper fulfills the described program.

First of all, we will introduce the generalized definition of the magnetic viscosity. Let us consider the system of magnetic moments, which was previously saturated in one (positive) direction using the strong magnetic field and then placed in the opposite field  $\mathbf{H}_0 = -H_0 \mathbf{e}_z$ . Those magnetic moments, which have in this field two equilibrium states, separated by an energy barrier  $E > 0$ , will be responsible for the time-dependent magnetic phenomena. If we denote the  $z$ -projection of the moment with the energy barrier  $E$  before and after reversing as  $M_{z\uparrow}$  and  $M_{z\downarrow}$ , then the change in the magnetization due to the reversing of this moment will be  $\delta M(E) = M_{z\uparrow} - M_{z\downarrow}$  and the time-dependent magnetization of the system with the energy barrier distribution can be written as (we omit the index  $z$ )

$$m(\tau) = \int_{E_{\min}}^{E_{\max}} \rho(E) \delta m(E) e^{-\Gamma(E)\tau} dE + m^\infty, \quad (4)$$

where we have introduced the reduced magnetization  $m = M/M_s$  ( $M_s$  denotes the saturation magnetization), time  $\tau = t\gamma_0$  and relaxation speed  $\Gamma = \exp(-E/T)$ . The quantity  $m^\infty$  represents the magnetization of the system after the infinite measurement time and is equal to the magnetization of the same system saturated in a large negative field and then placed in a field  $\mathbf{H}_0$  of the same direction.

The system will obey the linear logarithmic time dependence law only for sufficiently slow varying energy barrier density  $\rho(E)$  (see above). Nevertheless, it is always possible to define the generalized magnetic viscosity. Namely, the time derivative of the magnetization, according to eq. (1), can be written in the form

$$dm/d \ln t = -S,$$

which can be used for the definition of magnetic viscosity:

$$S = -dm/d \ln \tau = -\tau dm/d\tau,$$

and after substitution of (4) we obtain

$$S = \tau \int_{E_{\min}}^{E_{\max}} \rho(E) \delta m(E) \Gamma(E) e^{-\Gamma(E)\tau} dE. \quad (5)$$

The magnetic viscosity defined in this way can (and, in general, will) be time dependent. The advantage of this definition is, that it enables to define the magnetic viscosity for any system and leads to the time-independent viscosity (i.e., eq. to (1) with  $S = \text{Const}$ ) for the system with sufficiently wide and slow varying energy barrier distribution.

## 2. Energy barrier density for an ideal (noninteracting) Stoner–Wohlfarth ensemble

To calculate the magnetic viscosity using the general definition (5) first of all the energy barrier density  $\rho(E)$  should be found. For simplicity we consider the ensemble of SW particles with the random distribution of easy axes, but all our results can be obtained in the similar way for arbitrary orientation distribution, except for the case of aligned or almost aligned ensemble (see below.)

for the particle with orientation  $0 < \theta_0 < \pi/2$  (this is the angle between the easy axes and the external field) the energy barrier  $E$  in the applied field  $\mathbf{h}_a = -h_a \mathbf{e}_z$  is [11]

$$E = 2\left(\frac{2}{3}\right)^{3/2} KV \left(1 - \frac{h_a}{h_s(\theta_0)}\right)^{3/2} \sin 2(\theta_s - \theta_0) \quad (6)$$

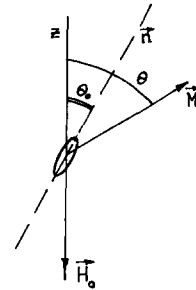


Fig. 1. The magnetic particle with the anisotropy axis  $n$  and magnetization  $M$  in the external field  $H_0$ .

(see fig. 1), where  $K$  denotes the uniaxial anisotropy constant,  $V$  the particle volume and the dependence of the switching field  $h_s$  and the switching angle  $\theta_s$  on the particle orientation  $\theta_0$  is given by [11,12]

$$\tan^3(\theta_s - \theta_0) = \tan \theta_0, \quad (7)$$

$$h_s(\theta_0)$$

$$= \frac{[1 - \tan^{2/3}(\theta_s - \theta_0) + \tan^{4/3}(\theta_s - \theta_0)]^{1/2}}{1 + \tan^{2/3}(\theta_s - \theta_0)} \quad (8)$$

(the reduced field is  $h = H/H_K$  with the anisotropy field  $H_K = 2K/M_s$ ). The switching field has a minimum  $h_s = \frac{1}{2}$  for  $\theta_0 = \pi/4$  and two maxima  $h_s = 1$  for  $\theta_0 = 0$  and  $\theta_0 = \pi/2$ . Near the minimum  $h_s$  is given by [12]

$$h_s(\theta_0) = \frac{1}{2} + \frac{1}{3}(\theta_0 - \pi/4)^2. \quad (8a)$$

The expression (6) is valid for all angles  $\theta_0$ , except for  $\theta_0 \ll 1$  [11], which corresponds to the particles with the axes almost aligned in the field direction. The fraction of such particles is usually small ( $\sim \sin \theta_0 \sim \theta_0 \ll 1$ ), except for the case of aligned or almost aligned ensemble, which requires separate consideration. It should also be mentioned, that expression (6) is formally derived for  $\Delta h/h_s = (h_s - h_a)/h_s \ll 1$ , but a numerical study has shown that it works with very high accuracy up to  $\Delta h/h_s \sim 0.5$ , which covers the whole field region of practical interest [11].

For the known easy-axes distribution  $f(\theta_0)$  (for the random orientation  $f(\theta_0) = \sin \theta_0$ ) the en-

ergy barrier density can be found from eqs. (6)–(8) in the usual way, i.e.

$$\rho(E) = f(\theta_0(E)) \left| \frac{d\theta_0(E)}{dE} \right|, \quad (9)$$

where  $\theta_0(E)$  is the angle dependence on the energy barrier obtained from eq. (6). Due to the very complex dependence (6) the function  $\theta_0(E)$  and the corresponding derivative  $d\theta_0(E)/dE$  can be calculated exactly only numerically. But basic features of the energy barrier density, such as singularities and limit points, can be derived analytically.

The analysis shows that for various external fields  $0 < h_a < 1$  there are five different types of the dependencies  $E(\theta_0)$ , which results in five types of the energy barrier density (fig. 2).

(i) For small fields  $0 < h_a < \frac{1}{11}$  (fig. 2a)  $E(\theta_0)$  has a single extremum (maximum) for  $\theta_0 = \pi/4$ .

$E(\theta_0)$  near this maximum can be found using eq. (8a) and has the form

$$E(\Delta\theta_0) = E_{s1}(1 + b_1\Delta\theta_0^2), \quad (10)$$

where  $\Delta\theta_0 = \theta_0 - \pi/4$ ,  $E_{s1} = U(1 - 2h_a)^{3/2}$ ,  $b_1 = 2(11h_a - 1)/9(1 - 2h_a)$  (here  $b_1 < 0$  and  $U = 2(\frac{2}{3})^{3/2} KV$ ).

This extremum causes a singularity in the energy barrier density  $\rho(E)$ , because in the extremum point  $dE/d\theta_0 = 0$  and hence  $\rho(E) \sim d\theta_0/dE \sim (dE/d\theta_0)^{-1} \rightarrow \infty$ . The quadratic extremum (10) will result in the inverse square root singularity

$$\rho(E) = \frac{1}{2^{3/2}} \frac{1}{\sqrt{b_1 E_{s1}}} \frac{1}{\sqrt{E - E_{s1}}}. \quad (11)$$

(ii) For  $h_a = \frac{1}{11}$  (not shown in fig. 2) the qualitative behavior of  $E(\theta_0)$  is similar to the previous case, but the maximum of  $E(\theta_0)$  for  $\theta_0 = \pi/4$  is

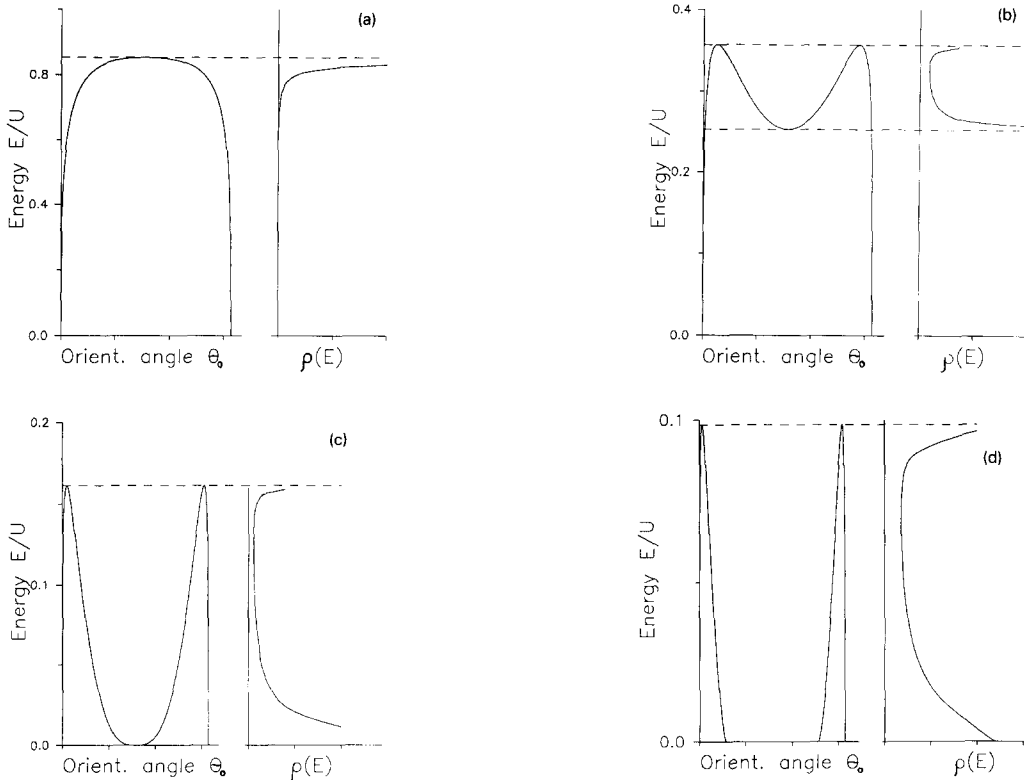


Fig. 2. Energy barrier density for the randomly oriented ideal Stoner–Wohlfarth ensemble for different applied fields: (a)  $0 < h_a < \frac{1}{11}$ , (b)  $\frac{1}{11} < h_a < \frac{1}{2}$ , (c)  $h_a = \frac{1}{2}$ , (d)  $\frac{1}{2} < h_a < 1$ .

quartic, i.e.,  $E(\Delta\theta_0) = E_{s1}(1 - \frac{4}{81}\Delta\theta_0^4)$ , leading to a singularity of the form

$$\rho(E) = \frac{1}{4\sqrt{2}} \frac{1}{\frac{4}{81}E_{s1}^{1/4}} \frac{1}{(E - E_{s1})^{3/4}}.$$

(iii) In the most interesting (from both theoretical and practical points of view) region  $\frac{1}{11} < h_a < \frac{1}{2}$  the function  $E(\theta_0)$  has one quadratic minimum for  $\theta_0 = \pi/4$  with parameters, as in eq. (10) (but now  $b_1 > 0$ ) and two symmetrical maxima (quadratic as well), for  $\theta_0 = \pi/4 \pm \Delta\theta_{\max}$  with the parameters  $E_{s2}$  and  $b_2$ , which should be calculated numerically. Hence the density of energy barriers has two singularities for  $E = E_{s1}$  and  $E = E_{s2}$  given by the expressions similar to eq. (11) (fig. 2b).

(iv) In the applied field  $h_a = \frac{1}{2}$  the minimum in the  $E(\theta_0)$  dependence has the form

$$E(\Delta\theta_0) = \frac{2}{3}U |\Delta\theta_0|^3,$$

resulting in a  $\rho(E)$  singularity for  $E \rightarrow 0$ :

$$\rho(E) = \frac{1}{\sqrt{2}} \frac{1}{(2U)^{1/3}} \frac{1}{E^{2/3}}.$$

The singularity for  $E_{s2} > 0$  has the same form, as in the previous case (fig. 2c).

(v) For the last region of interest  $\frac{1}{2} < h_a < 1$  only the particles with  $h_s(\theta_0) > h_a$  will contribute to the time-dependent magnetization. The  $E(\theta_0)$  dependence is shown in fig. 2d with the corresponding density of energy barriers, which has now only one singularity arising from the quadratic maxima in  $E(\theta_0)$ .

### 3. Magnetic viscosity for the ideal Stoner–Wohlfarth ensemble

Exact calculations of the magnetic viscosity for arbitrary system parameters can be done only numerically. But due to the fact that the major contribution to the energy barrier density arises from the singularities, basic results can be obtained analytically.

Expression (5) for the magnetic viscosity can be rewritten in the form

$$S = \tau \int_{E_{\min}}^{E_{\max}} \rho(E) \delta m(E) K(E, \tau) dE, \quad (12)$$

where the kernel of this integral,

$$K(E, \tau) = \exp(-E/T - \tau e^{-E/T}), \quad (13)$$

has as a function of  $E$  a sharp (due to the large value of  $\tau \sim 10^9 - 10^{13}$  s) peak of the width  $\Delta E \sim T$  around the critical energy  $E_c = T \ln \tau$ , where  $K(E, \tau)$  has its maximum value  $K_{\max}(E, \tau) = 1/(e\tau)$ .

Due to this feature of the kernel  $K(E, \tau)$  and to the singularities in the energy barrier density  $\rho(E)$ , the integral (12) and hence the magnetic viscosity can be large only if at least one singular point  $E_{s1}$  or  $E_{s2}$  of  $\rho(E)$  is in the vicinity of the critical energy  $E_c$ :  $|E_{s1(2)} - E_c| \leq T$ . It can be shown, that the singularity arising from the  $E(\theta_0)$  extremum near  $\theta_0 = \pi/4$  gives the largest contribution in the density of energy barriers and hence the maximum magnetic viscosity should be observed for parameters (external field, measurement time, particle characteristics, etc.) corresponding to the case  $|E_{s1} - E_c| \leq T$ . For this energy region we can neglect the non-singular part of  $\rho(E)$  and write the magnetic viscosity as

$$S = \frac{\tau \delta m(E_c)}{2\sqrt{2}b_1 E_{s1}} \int_{E_{s1}}^{+\infty} \frac{1}{\sqrt{E - E_{s1}}} \times \exp(-E/T - \tau e^{-E/T}) dE. \quad (14)$$

Here we have expanded the upper integral limit to  $+\infty$  using the rapid decrease of  $K(E, \tau)$  for  $E > E_c$  and replaced the slowly varying function  $\delta m(E)$  by its value  $\delta m(E_c)$  at the critical point. In this case ( $E \approx E_{s1}$ )  $\delta m(E_c)$  corresponds to the reversing of particles with the orientation  $\theta_0 \approx \pi/4$  and hence  $\delta m(E_c) \approx 1$  in the applied field  $h_a \sim 0.5$  (for  $h_a = 0.5$  the exact solution of the equilibrium equations gives  $\delta m(\theta_0 = \pi/4, h_a = 0.5) = ((7 + 4\sqrt{2})^{1/2} + (7 - 4\sqrt{2})^{1/2})/4 \approx 1.18$ ).

Changing variables to  $x = (E - E_{s1})/T$  and introducing the notation  $\alpha = \exp(-E_{s1}/T)$ , we obtain

$$S = \frac{\delta m(E_c) \sqrt{T}}{2\sqrt{2b_1 E_{s1}}} \alpha \tau \int_0^{+\infty} \frac{1}{\sqrt{x}} \times \exp(-x - \alpha \tau e^{-x}) dx. \quad (14a)$$

The value of the magnetic viscosity (14a) is determined by the product  $\alpha \tau = \exp(-\delta)$ , where  $\delta = (E_{s1} - E_c)/T$ . It is easy to show that for  $|\delta| \gg 1$   $S$  is at least exponentially small for both  $\delta > 0$  ( $S \sim e^{-\delta}$ ) and  $\delta < 0$  ( $S \sim 1/\mu^{\mu-1}$ , where  $\mu = e^{|\delta|} \gg 1$ ). Hence, as it was mentioned above,  $S$  should have its maximum value for  $|\delta| \leq 1$ . Expanding  $\alpha \tau = e^{-\delta}$  up to the second order in  $\delta$ , we obtain the desired expression for the magnetic viscosity in the form

$$S = \frac{\delta m(E_c) \sqrt{T}}{2\sqrt{2b_1 E_{s1}}} \left( I_0 + \delta I_1 - \frac{\delta^2}{2} I_2 \right), \quad (15)$$

where  $I_i$  are some definite integrals, which can be easily evaluated numerically:  $I_0 \approx 0.836$ ,  $I_1 \approx -0.374$  and  $I_2 \approx 0.355$ . As a function of  $\delta$ , this expression achieves its maximum value

$$S = \frac{\delta m(E_c) \sqrt{T}}{2\sqrt{2b_1 E_{s1}}} \left( I_0 + \frac{I_1^2}{2I_2} \right) \approx 1.02 \frac{\delta m(E_c) \sqrt{T}}{2\sqrt{2b E_{s1}}} \quad (16)$$

for  $\delta = I_1/I_2 \approx -1.05$ . As it is usually the case for the quadratic Taylor expansion with the coefficients of the same order of magnitude, the value of the maximum is determined with higher accuracy than its position.

Substituting  $b_1$  and  $E_{s1}$  from eq. (10), we obtain the final expression for the maximum magnetic viscosity:

$$S \approx \frac{0.72 \delta m(E_c)}{(11h_a - 1)^{1/2} (1 - 2h_a)^{1/4}} \sqrt{\frac{T}{KV}}. \quad (17)$$

The square-root temperature dependence of this maximum is the direct consequence of the singularity in the energy barrier density  $\rho(E)$  (for analytical  $\rho(E)$  this dependence is always linear).

This expression is not valid for  $h_a = \frac{1}{11}$  and  $h_a = \frac{1}{2}$  and in the vicinity of these fields, where the singularities in the energy barrier density are different from the inverse square root form used by the derivation of eq. (17).

The field region, where the magnetic viscosity is large (for other parameters being unchanged) can be found from the condition  $\delta \sim 1$ :

$$\Delta h_a \sim (T/KV)^{2/3} \ln(\tau)^{-1/3}$$

( $\Delta h_a \ll 1$ , because  $T \ll KV$  and  $\tau \gg 1$ ). The same condition enables to calculate the measurement time, for which the maximum of the magnetic viscosity should be observed:

$$\tau^{\max} \approx \exp\left(\frac{U}{T} (1 - 2h_a)^{3/2}\right). \quad (18)$$

To obtain the exact field dependence of the magnetic viscosity for the whole field range numerical computations are required. But the qualitative behavior can again be traced using the same main features of  $\rho(E)$  and the kernel  $K(E, \tau)$ .

For the most interesting case of the measurement parameters corresponding to the critical energy  $E_c < E_{s1}$  ( $h_a = 1/11$ ) (in other words, for  $T \ln \tau < 0.8KV$ ) this behavior can be explained using fig. 3. In this figure the kernel  $K(E, \tau)$  (dashed line) is shown together with the energy

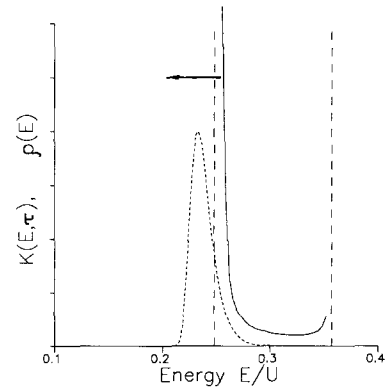


Fig. 3. To the explanation of the field dependence of the magnetic viscosity: kernel  $K(E, \tau)$  (13) (dotted line) and the energy barrier density  $\rho(E)$  (solid line) from the integral (12). Vertical dashed lines represent the asymptotics of  $\rho(E)$ . The arrow indicates the shift direction of  $\rho(E)$  by increasing the external field.

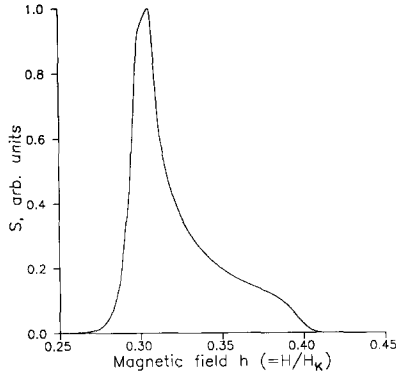


Fig. 4. Field dependence of the magnetic viscosity  $S$  ( $T/U = 0.01$ ).

barrier density  $\rho(E)$  (solid line). Magnetic viscosity, as is defined by eq. (12), is proportional to the integral from the product of these functions.

For small external fields, when both  $E_{s1} > E_c$  and  $E_{s2} > E_c$ , magnetic viscosity is very small. It increases with the field, when the energy barrier density shifts towards the lower energy (in the arrow direction in fig. 3) and  $E_{s1}$  decreases tending to  $E_c$ . The viscosity achieves its maximum, when  $E_{s1} \approx E_c$  for the field  $h_{a1} \approx 0.5(1 - ((T/U) \times \ln \tau)^{2/3})$ .

Then, with the further increase of the field, magnetic viscosity decreases because the critical energy corresponds now to the minimum in the energy barrier density between two singularities. After this minimum is passed,  $S$  could in principle increase again due to the second singularity in  $\rho(E)$ , but the change in the magnetization  $\delta m(E)$  corresponding to this singularity is much less than for  $E_{s1}$ . For this reason, whether the second singularity will cause the second maximum or not,  $S$  depends on the easy axes orientation density. Numerical calculations show, that for the random particle orientation there is no second maximum in the  $S(h)$  dependence (fig. 4).

#### 4. Interparticle interaction effects

For real fine-particle ensembles the effects of magnetic interparticle interaction should be taken into account. It was shown, that for the low

volume concentration of the ferromagnetic phase  $\eta \ll 1$  and for strong anisotropy  $\beta = 2K/I_s^2 \gg 1$  these effects can be described adequately by the mean field theory [12].

In this theory the interaction field, applied to the given particle by all other particles of the ensemble, is treated as a random field, not correlated either with the position of the particle, or with its moment direction. Hence any quantity  $G$  in this approximation can be calculated as

$$G(\mathbf{h}_0) = \int G^{(0)}(\mathbf{h})f(\mathbf{h} - \mathbf{h}_0) d\mathbf{h}, \quad (19)$$

where  $G^{(0)}$  denotes the corresponding quantity for the ideal ensemble and  $f(\mathbf{h})$  the three-dimensional distribution density of the interaction field. It was also shown [12] that  $f(\mathbf{h})$  for the dipole-interacting particles is not Gaussian, as it was suggested by many authors, but can be approximated with high accuracy by the restricted Lorentzian distribution.

To describe the influence of the interparticle interaction on the magnetic viscosity, we cannot start with the final expression (17), because it was obtained using the assumption  $E_{s1} \approx E_c$ , i.e. for  $h_0 \approx 0.5(1 - ((T/U) \ln \tau)^{2/3})$ , in other words, for a definite value of the magnetic field. To take the distribution of these fields into account, we should begin with the energy barrier distribution density for the interacting ensemble. For the semiquantitative analysis the one-dimensional version of 3D-expression (19) can be used, which gives for the energy barrier density

$$\rho(E, h_0) = \int \rho^{(0)}(E, h_z)f(h_z - h_0) dh_z, \quad (20)$$

where  $\rho^{(0)}$  describes the energy barrier density for the noninteracting case, discussed in section 2. The distribution density of the  $z$ -component of the random field can be written as [12]

$$f(h_z) = \frac{C_N}{1 + (h_z/\Delta)^2}, |h_z| \leq a/\beta \quad (21)$$

( $f(h_z) \equiv 0$  for  $|h_z| > a/\beta$ ). Here  $\beta = 2K/I_s^2 (\gg 1)$  is the reduced anisotropy constant, the width of the distribution  $\Delta$  is given by  $\Delta = A\eta/\beta$  ( $A \approx 4.54$ ), the normalizing constant  $C_N^{-1} = 2\Delta$

$\arctg(1/\Delta\beta) \approx \pi\Delta$  for  $\eta \ll 1$  and the restriction constant  $a$  can be found from the condition, that the dispersion of the random field is [12]

$$\overline{h_z^2} = \frac{4}{3} \frac{2\pi^2\eta}{15\beta^2}. \quad (22)$$

The energy barrier density defined as in eq. (20) is an analytical function. For this reason, to find the maximum magnetic viscosity  $S_{\max}$ , we have to determine the maximum value of the energy barrier density  $\rho_{\max}$ , after which  $S_{\max}$  will be given by  $S_{\max} = T \rho_{\max}$  (see eq. (2)).

For small particle concentration  $\eta \ll 1$  the distribution width  $\Delta \sim \eta/\beta \ll 1$  and hence the maximum of  $\rho(E)$  will be located in the vicinity of the singular point  $E_{s1}$ . For this energy region we can write the energy barrier density in the external field  $h_0$  (similar to eq. (14)) as

$$\rho(E) = \frac{C_N}{2\sqrt{2}} \int_{h_{\min}}^{h_0+a/\beta} \frac{1}{\sqrt{b_1(h)E_{s1}(h)}} \times \frac{1}{\sqrt{E-E_{s1}(h)}} \frac{1}{1+(h-h_0)^2/\Delta^2} dh, \quad (23)$$

where we have omitted index  $z$  by the interaction field  $h$ . The lower integral limit, found from the condition  $E \geq E_{s1}(h)$ , is given by  $h_{\min} = 0.5(1 - (E/U)^{2/3})$ .

Because the interaction field distribution is very narrow, slowly varying functions  $b_1(h)$  and  $E_{s1}(h)$  under the first square root can be replaced by the corresponding values in the  $h$ -distribution center  $h_0$ . In the second square root  $E_{s1}(h) = U(1-2h)^{3/2}$  can be expanded up to the first order of the small quantity  $\Delta h = h - h_0$ . Finally, changing variables to  $x = (h - h_{\min})/\Delta$ , we obtain ( $\delta = h_{\min}/\Delta$ )

$$\rho(E) = \frac{\sqrt{3}}{4U\pi\sqrt{\Delta}} \frac{1}{\sqrt{(11h_0-1)(1-2h_0)}} \times \int_0^{+\infty} \frac{1}{\sqrt{x(1+(x+\delta)^2)}} dx, \quad (24)$$

where we have used definitions of  $E_{s1}(h)$  and

$b_1(h)$  from eq. (10), replaced the normalized constant  $C_N$  by its limiting value  $C_N \approx (\pi\Delta)^{-1}$  for  $\eta \ll 1$  and expanded the upper integral limit  $x_{\max} \sim 1/\eta \gg 1$  to  $+\infty$  using the rapid decrease of the integrand for  $x \gg 1$ .

The similar procedure, as used to obtain eq. (15) from (14a), i.e. the expansion of the integrand up to the second order of  $\delta$ , results in an energy barrier density as function of  $\delta$  of

$$\rho(E) = \frac{\sqrt{3}}{4U\pi\sqrt{\Delta}} \frac{1}{\sqrt{(11h_0-1)(1-2h_0)}} \times [I_0 + \delta I_1 - \frac{1}{2}\delta^2 I_2].$$

The numerical values of the definite integrals  $I_i$  here are  $I_0 = \pi/\sqrt{2}$ ,  $I_1 = -(\pi/4)/\sin(\pi/4)$ ,  $I_2 = -I_1/2$ . The maximum value of the expression in the square brackets, achieved for  $\delta = I_1/I_2 = -2$  is equal to  $I_{\max} = (\pi/\sqrt{2})(1 + 8/\pi^2)$ . Using the value  $\Delta = A\eta/\beta$  ( $A \approx 4.54$ ) for the interaction field distribution width, the definition of  $U$  from eq. (10) and the formula (2) for the magnetic viscosity, we obtain the final expression for the maximum value of  $S$ :

$$S_{\max} \approx \frac{0.112}{\sqrt{(11h_0-1)(1-2h_0)}} \sqrt{\frac{\beta}{\eta}} \frac{T}{KV}. \quad (25)$$

As eq. (17), this expression is not valid near  $h_0 = \frac{1}{2}$  and  $h_0 = \frac{1}{11}$ , where other asymptotic behavior of  $\rho^{(0)}$  applies. It should also be mentioned, that the value of  $\delta = -2$ , where the maximum of  $\rho$  is achieved, shows that the determination of the integral's maximum value in eq. (24) using the quadratic Taylor expansion is not very accurate. But this error is of the same order of magnitude, as the error introduced by the use of 1D version (20) instead of the exact 3D form (19).

Here we also underline, that due to the singularity in the energy barrier density for the ideal ensemble  $\rho^{(0)}$ , the use of the incorrect Gaussian distribution of the interaction field instead of eq. (21) would result not only in the wrong numerical value of  $S_{\max}$ , but also in the wrong functional dependence of the maximum magnetic viscosity on the anisotropy constant  $\beta$  ( $S_{\max} \sim \beta$  instead of  $S_{\max} \sim \sqrt{\beta}$  in eq. (25)).



**5. The interacting Stoner–Wohlfarth ensemble with the particle size distribution**

To find the energy barrier density, taking into account both the interparticle interaction and the particle size distribution, we can apply the procedure similarly to those used in section 4 and write  $\rho(E)$  near its maximum (which will be again located in the vicinity of  $E_{s1}$ ) as

$$\rho(E) = \frac{1}{2\sqrt{2}} \iint_{E_{s1}(h, V) < E} dh dE \times \frac{1}{\sqrt{b_1(h) E_{s1}(h, V)}} \frac{1}{\sqrt{E - E_{s1}(h, V)}} \times f_h(h) f_V(V), \tag{25}$$

where  $f_h(h)$  and  $f_V(V)$  denote the distribution densities of the interaction field and particle volume.

As in eq. (23), for the narrow  $h$  and  $V$  distributions slowly varying functions  $b_1(h)$  and  $E_{s1}(h, V)$  under the first square root can be replaced by their values in the distribution centers  $h_0$  and  $V_0$ . Expanding  $E_{s1}(h, V)$  up to the first order of small quantities  $v = (V_0 - V)/V_0 = \Delta V/V_0$  and  $u = 3(h - h_0)/(2h_0 - 1) = 3\Delta h/(2h_0 - 1)$ , we obtain

$$\rho(E) = \frac{1}{2\sqrt{2}b_1(h_0) E_{s1}(h_0, V_0)} \iint_{u+v < \delta\epsilon} du dv \times \frac{1}{\sqrt{\Delta\epsilon - (u+v)}} f_u(u) f_v(v), \tag{26}$$

where  $\Delta\epsilon = E/E_{s1}(h_0, V_0) - 1$ .

Further analytical calculations can be done only if the distribution densities of both quantities  $u$  and  $v$  are Gaussian. The distribution density of the particle volumes  $V$  (and of  $v = V/V_0$ ) is usually log normal and indeed tends to Gaussian in the small distribution width limit. But the distribution density of the interaction field  $\Delta h$ , and hence of the reduced quantity  $u$  is Lorentzian and for this reason one should use numerical methods to obtain exact results for each concrete case.

Nevertheless, to estimate qualitatively the joint influence of the interparticle interaction and particle size distribution, we can use for the interaction field the Gaussian distribution density with the correct dispersion (22). The error resulting from this substitution will be much less than in the previous case (see the end of section 4), because the energy barrier density for the noninteracting ensemble with the particle size distribution is already an analytical function. For this reason further averaging of this energy barrier density over the Gaussian field distribution (instead of Lorentzian) with the correct dispersion should provide satisfactory results.

In this approximation the sum in eq. (26)  $x = u + v$  of two independent random quantities  $u$  and  $v$ , having each Gaussian distributions with dispersions  $\sigma_u^2$  and  $\sigma_v^2$ , has itself the Gaussian distribution with the dispersion  $\sigma_x^2 = \sigma_u^2 + \sigma_v^2$ . Therefore,  $\rho(E)$  can be rewritten as

$$\rho(E) = \frac{1}{2\sqrt{2}b_1(h_0) E_{s1}(h_0, V_0)} \frac{1}{\sqrt{2\pi\sigma_x}} \times \int_{-\infty}^{\Delta\epsilon} \frac{dx}{\sqrt{\Delta\epsilon - x}} e^{-x^2/2\sigma_x^2}. \tag{26}$$

Changing variables to  $y = (\Delta\epsilon - x)/\sigma_x$ , we obtain  $\rho(E)$  as a function of  $\delta = \Delta\epsilon/\sigma_x$  in the form

$$\rho(E) = \frac{1}{2\sqrt{2}b_1(h_0) E_{s1}(h_0, V_0)} \frac{1}{\sqrt{2\pi\sigma_x}} \times \int_0^{+\infty} dy \frac{e^{-(y-\delta)^2/2}}{\sqrt{y}}. \tag{27}$$

The maximum of this expression can be found with the same procedure, as used in sections 3 and 4 and is located by  $\delta = 4\pi/\Gamma^2(\frac{1}{4}) \approx 0.96$ :

$$\rho_{max} = \frac{1}{4\sqrt{\pi\sigma_x} \sqrt{2}b_1(h_0) E_{s1}(h_0, V_0)} \frac{\Gamma(\frac{1}{4})}{2^{3/4}} \times \left(1 + \frac{4\pi^2}{\Gamma^4(\frac{1}{4})}\right),$$

where  $\Gamma$  denotes the gamma function [13]. There-

fore the desired expression for the magnetic viscosity is given by

$$S_{\max} \approx \frac{0.73}{\sqrt{11h_0 - 1} (1 - 2h_0)} \frac{1}{(\sigma_v^2 + \sigma_u^2)^{1/4}} \frac{T}{KV}. \quad (28)$$

Here  $\sigma_v^2$  is the reduced dispersion of the particle volume distribution and  $\sigma_u^2 = (1 - 2h_0)^2 \overline{h_z^2} / 9$ , where  $\overline{h_x^2}$  is given by eq. (22).

For one practically important case eq. (28) gives the exact quantitative results, despite the use of the incorrect distribution density for the interaction field. Namely, if the particle size distribution is relatively wide, so that  $\sigma_v^2 \gg \sigma_u^2$ , we can neglect the broadening resulting from the interparticle interaction (in other words, treat the distribution density  $f_h(h)$  as the delta function  $\delta(h - h_0)$ ). In this case the maximum of the magnetic viscosity is given by expression (28) with  $\sigma_u = 0$ .

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