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Influence of texture on the magnetic viscosity

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Abstract

The influence of texture on the time and field dependence of the magnetization, the magnetic viscosity, the irreversible susceptibility and the fluctuation field is calculated for two models of non-interacting uniaxial particles differing in the angular dependence of the switching field. The first model consists of single-domain particles reversing their magnetization by coherent rotation. In the second model the grains exhibit a $1/\cos\vartheta$ dependence of the switching field on the misalign angle. A detailed comparison of our results with the existing theory of the magnetic viscosity is given. For both cases it was shown, that texture alone cannot give a barrier distribution smooth enough to get the mostly measured ln t behaviour of the magnetization. The fluctuation field S_V is found to be independent on the texture, in good agreement with the experimental situation, but in contrast to the theoretical considerations reported in the literature.

1. Introduction

The magnetic viscosity is one of the characteristics of technically applied polycrystalline hard magnets, knowledge about which is necessary to give an expectation of the long term stability of these materials in technical devices [1–5]. Measurement and definition of the viscosity is based on the experimental fact, that nearly all known magnetic materials show a logarithmic decrease of the magnetization with time.

The theory of magnetic viscosity usually starts from a distribution of energy barriers, or, what is essentially the same, from a broad distribution of relaxation times, whereas the theory of the magnetization curve is based on a switching field distribution. Both the barrier distribution and the switching field distribution depend usually on the time and the history of the magnetization process. Deviations from the logarithmic time dependence are sometimes observed. They are not measurable if the change in the slope of the barrier distribution in the interval between $25k_BT$ and $30k_BT$ is small. From the theoretical point of view it would be desirable to start from time independent distributions of intrinsic parameters of the grains, e.g. the volume, the shape, the anisotropy, the saturation magnetization. This requires a detailed knowledge about the mode of magnetization reversal, since the activation energy necessary to overcome an energy barrier has to be calculated from micromagnetics.

In the following we will study, how a distribution of the particle easy axis (a texture) will influence the aftereffect. Therefore we consider first the Stoner–Wohlfarth model [6] with a fiber texture. This model

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is easy enough to get the complete time behaviour without the crude approximations inherent in the usual theory of the magnetic viscosity and it can serve as a reference system for other calculations. The same model was used to study the aftereffect in films of recording media [7,8], and it was used for ideal aligned grains [9]. Ideal aligned Stoner–Wohlfarth ensembles with a log-normal distribution of particle volumes were considered in Ref. [8] and it was shown that deviations from the ln t behaviour can be explained via a power series expansion of the barrier distribution. Berkov [10] calculated the barrier distribution and the field dependence of the viscosity for an ensemble with a random distribution of easy axes. His treatment is based on an approximate form for the misalignment angle and field dependence of the energy barrier of a single grain, given in Ref. [11]. This produces some unphysical pecularities at fields of $H = \frac{1}{11}H_A$ and it yields wrong results regarding the field dependence of the barrier distribution and the field dependence of the viscosity. Since we use the exact dependence of the barrier on the field and on the misalignment angle, we are able to correct the wrong pictures given in Ref. [10]. The latter is not the main point of the paper, since our interest is focused on the dependence of the viscosity and the susceptibilities on the texture function. Therefore, in a second point, the influence of texture is considered starting from the $1/\cos\vartheta$ ansatz [12] for the angle dependence of the switching field of the individual grains, which is of interest, since Martinez and Missell [13] measured the viscosity constant S_V of sintered Nd–Fe–B based magnets to be nearly independent on the degree of alignment, in sharp contrast to the theory given in their paper. The experimental result was also confirmed by Street et al. [14]. It is actually this controversy between theory and experiment, which motivated to produce this paper.

2. Outlines of the 'standard theory' of the magnetic viscosity

2.1. Time dependence of magnetization and viscosity

The calculation of the thermal aftereffect in polycrystals should start from a mesoscopic level. Therefore we assume the heterogeneous magnet to consist of a lot of small volume elements, which can exist in two magnetization states only, one of them stable and the other metastable. For example, the volume elements may be small single domain grains, or the volume between two neighbouring pinning sites of a domain wall, etcetera, in dependence of which micromagnetic model one has in mind. The thermodynamic potential of the volume element exhibits a two minima shape for internal fields below the switching field and a one minimum shape above. Usually a double well potential is sufficient for a theoretical description of the thermal aftereffect. The magnetization corresponds either to the global minimum or to the local minimum, depending on the prehistory of the magnetization process. In both situations the magnetization will change with time due to thermal fluctuations (other processes are left out of consideration). This means that the magnetization of the volume element at a future time can only be determined with a probability, which has to be calculated from

$$\frac{\mathrm{d}p}{\mathrm{d}t} = -w_{12}\,p + w_{21}(1-p). \tag{1}$$

Here p describes the probability to find the volume element in the metastable state (state 1), w_{12} and w_{21} gives the transition rates for jumping from state 1 to the stable state (state 2) and w_{21} vice versa. If an opposite field is applied, which is lower than the switching field H_S, the volume element remains metastable, but with a reduced barrier height. With increasing opposite field the energy barrier will be lowered and it vanishes at the switching field. If the function $E_B(H)$ describing the dependence of the barrier height on the applied field decreases monotonously, we can write down the solution of Eq. (1):

$$p(t) = \Theta(E_B(H)) \times \left\{ \frac{w_{21}}{w_{12}+w_{21}} + \frac{w_{12}}{w_{12}+w_{21}}\, \mathrm{e}^{-(w_{12}+w_{21})t} \right\}, \tag{2}$$

with Θ being the step function. The energy of the metastable state is called E_1, the energy of the stable

configuration E_2, and the energy of the maximum between them E_M. For thermal fluctuation we have

$$w_{12} = \Gamma_0 \, e^{-(E_M - E_1)/k_B T}, \tag{3}$$

$$w_{21} = \Gamma_0 \, e^{-(E_M - E_2)/k_B T}, \tag{4}$$

where Γ_0 is to be determined from microscopic theories, which is quite hard and till now not very satisfactorily done [1–3,15,16], or one can try to get it from experiment. It ranges typically from 10^9 to 10^{12} Hz [2,3]. Each of the volume elements of the heterogeneous magnet may be characterized by its barrier energy $E_B = E_M - E_1$ alone, since the barrier for backjumps, $E_M - E_2$, is related to E_B in a unique manner. The probability to find a volume element with a barrier energy between E_B and $E_B + dE_B$, i.e. $f(E_B)\,dE_B$ is the sum over all volume elements having a barrier in this interval divided by the total volume of the specimen. The averaged normalized magnetization $\bar{m} = \bar{M}/M_S = \bar{I}/I_S$ is calculated accordingly by

$$\bar{m}(H, t) = \int_0^\infty dE_B \, f(E_B) m(H, t), \tag{5}$$

$$\bar{m}(H, t) = \int_0^\infty dE_B \, (2p(t) - 1) f(E_B). \tag{6}$$

The derivation of the viscosity starting from Eq. (5) was given in an elucidated manner by Berkov [17], proving the universal character of the ln t law. Since Berkov has not given the well known relation between the viscosity and the irreversible susceptibility, we will do that by formulating the theory given by Gaunt [18] in the mathematical language of Berkov. For compatibility reasons we repeat the main steps given in Ref. [17] first. The magnetic viscosity measured in units of I_S we get from

$$S = -\frac{\partial \bar{m}(H, t)}{\partial \ln t}$$

$$= 2\int_0^\infty dE_B \, \frac{w_{12}}{w_{12} + w_{21}} f(E_B) \Gamma t \, e^{-\Gamma t}. \tag{7}$$

Here was abbreviated

$$\Gamma = w_{12} + w_{21}. \tag{8}$$

Usually backjumps are neglected, also in Ref. [17]. We emphasize, that neglection of backjumps from state 2 to state 1 replaces the long term behaviour of Eq. (2), resulting in the anhysteretic magnetization curve for $t \to \infty$, by the quite unphysical value $m(\infty) = \text{sign } H$. Nevertheless, for times relevant in experiments this assumption is justified. Using this approximation Eq. (8) simplifies to

$$\Gamma = w_{12} = \Gamma_0 \, e^{-E_B/k_B T} \tag{9}$$

and Eq. (7) becomes

$$S(H, t) = 2\int_0^\infty dE_B \, f(E_B) \Gamma t \, e^{-\Gamma t}. \tag{10}$$

Following Refs. [18,17] the peaked structure of $\Gamma t \exp(-\Gamma t)$ is used to evaluate the integral. In the vicinity of the peak position $E_{B,max}$, which is given by

$$E_{B,max} = k_B T \ln(\Gamma_0 t), \tag{11}$$

the distribution function $f(E_B)$ is expanded into a Taylor series:

$$S(H, t) = 2 \sum_{n=0}^{\infty} \frac{1}{n!} \frac{d^n}{dE_B^n} f(E_B)\Big|_{E_{B,max}} \times \int_0^\infty dE_B \, (E_B - E_{B,max})^n \Gamma t \, e^{-\Gamma t}. \tag{12}$$

The derivatives are to be evaluated at the extremal point $E_{B,max}$. Substituting in a first step

$$E_B = -k_B T \ln(\Gamma/\Gamma_0) \tag{13}$$

and afterwards $x = \Gamma t$ yields finally

$$S(H, t) = 2k_B T \sum_{n=0}^{\infty} \frac{(-k_B T)^n}{n!} \frac{d^n}{dE_B^n} f(E_B)\Big|_{E_{B,max}} \times \int_0^{\Gamma_0 t} dx \, \ln^n(x) \, e^{-x}. \tag{14}$$

If the measurement is started at a time t_s for which the relation

$$\Gamma_0 t_s \gg 1 \tag{15}$$

holds, the upper boundary of the integrals can be set to infinity. Thus, the final expression is [17]

$$S(H, t) = 2k_B T \sum_{n=0}^{\infty} C_n \frac{(-k_B T)^n}{n!} \frac{d^n}{dE_B^n} f(E_B)\Big|_{E_{B,max}} \tag{16}$$

with

$$C_n = \int_0^\infty \mathrm{d}x\ \ln^n x\ \mathrm{e}^{-x}. \tag{17}$$

Practically, due to the large value of Γ_0, Eq. (15) holds for all the experiments reported up to now. Therefore the time dependence of S in Eq. (16) is due to the time dependence of $E_{\mathrm{B,max}}$, the value at which the derivatives of $f(E_\mathrm{B})$ have to be evaluated. Usually this time dependence is neglected, then one finds for the magnetization, by integration from the starting time t_S

$$\overline{m}(H, t) = \overline{m}(H, t_\mathrm{S}) - S \ln(t/t_\mathrm{S}). \tag{18}$$

If one takes this time dependence into account, a suitable manner to handle it is the expansion of the derivatives at the starting time t_S, resulting in deviations from the simple ln t behaviour. This was shown by El-Hilo et al. [8] for an ensemble of ideally aligned Stoner–Wohlfarth fine particles; since they did not use the energy barrier distribution directly but instead a distribution of single particle volumes, the mathematical terms are different.

2.2. Irreversible susceptibility and fluctuation field

From the mechanism outlined above follows a tight relation between the isothermal time dependence ($\Delta H = 0$) and the irreversible field dependence in a very short time ($\Delta t = 0$) of the magnetization. Regarding the field dependence of $f(E_\mathrm{B})$ the susceptibility follows by differentiating Eq. (6) with respect to H at fixed time:

$$\chi_\mathrm{irr} = \frac{\partial \overline{m}(H, t)}{\partial H} = 2\int_0^\infty \mathrm{d}E_\mathrm{B}\ \frac{\partial f(E_\mathrm{B})}{\partial H}\ \mathrm{e}^{-\Gamma t}. \tag{19}$$

This is the irreversible part of the susceptibility only, since reversible mechanisms are left out of consideration. Due to the implicit field dependence of $f(E_\mathrm{B})$,

$$\chi_\mathrm{irr} = 2\int_0^\infty \mathrm{d}E_\mathrm{B}\ \frac{\partial f(E_\mathrm{B})}{\partial E_\mathrm{B}}\ \frac{\mathrm{d}E_\mathrm{B}}{\mathrm{d}H}\ \mathrm{e}^{-\Gamma t}. \tag{20}$$

Assuming $\mathrm{d}E_\mathrm{B}/\mathrm{d}H$ to be constant, i.e. taking it at the point $E_\mathrm{B} = E_{\mathrm{B,max}}$, and integrating by parts yields

$$\chi_\mathrm{irr} = \left.\frac{\mathrm{d}E_\mathrm{B}}{\mathrm{d}H}\right|_{E_{\mathrm{B,max}}} 2\int_0^\infty \mathrm{d}E_\mathrm{B}\ \frac{\partial}{\partial E_\mathrm{B}}\left(f(E_\mathrm{B})\ \mathrm{e}^{-\Gamma t}\right)$$

$$= -\left.\frac{\mathrm{d}E_\mathrm{B}}{\mathrm{d}H}\right|_{E_{\mathrm{B,max}}} 2\int_0^\infty \mathrm{d}E_\mathrm{B}\ f(E_\mathrm{B})\ \mathrm{e}^{-\Gamma t}(-t) \times \frac{\partial \Gamma}{\partial E_\mathrm{B}}. \tag{21}$$

Taking into account that

$$\Gamma(0) = \Gamma_0, \qquad \Gamma(\infty) = 0,$$
$$\frac{\partial \Gamma}{\partial E_\mathrm{B}} = -\frac{\Gamma}{k_\mathrm{B}T}, \qquad f(\infty) = 0 \tag{22}$$

holds, the susceptibility is found to be

$$\chi_\mathrm{irr} = \left.\frac{\mathrm{d}E_\mathrm{B}}{\mathrm{d}H}\right|_{E_{\mathrm{B,max}}} \left(\frac{-2}{k_\mathrm{B}T}\int_0^\infty \mathrm{d}E_\mathrm{B}\ f(E_\mathrm{B})\,\Gamma t\ \mathrm{e}^{-\Gamma t}\right). \tag{23}$$

Here a term was neglected, which is small due to $\Gamma_0 t \gg 1$. Finally, by comparison with Eq. (10),

$$\chi_\mathrm{irr} = \frac{S}{k_\mathrm{B}T}\left.\frac{\mathrm{d}E_\mathrm{B}}{\mathrm{d}H}\right|_{E_{\mathrm{B,max}}}. \tag{24}$$

Usually the quotient S_V of S and χ_irr,

$$S_\mathrm{V} = \frac{S}{\chi_\mathrm{irr}} = k_\mathrm{B}T \Big/ \left.\frac{\mathrm{d}E_\mathrm{B}}{\mathrm{d}H}\right|_{E_{\mathrm{B,max}}}, \tag{25}$$

is called the fluctuation field [1,2,5] or viscosity coefficient [13,19], and discussed to be a characteristic of the materials. We want to emphasize that this is only true if the field derivative of the energy barrier is constant in the energy range between $25k_\mathrm{B}T$ and $33k_\mathrm{B}T$, otherwise the fluctuation field remains field dependent. With $\mathrm{d}E_\mathrm{B}/\mathrm{d}H =$ constant, as was proved e.g. for domain wall pinning [18], the field dependence of E_B is determined to be

$$E_\mathrm{B} = E_\mathrm{B}^0\left(1 - \frac{H}{H_\mathrm{S}}\right). \tag{26}$$

The switching field H_S of a single volume element with square hysteresis or the switching field if thermal activation is absent. If the field dependence of E_B^0 and H_S is weak, then S_V should be linearly dependent on temperature. The switching field H_S is given by the work to overcome the energy barrier at zero temperature:

$$E_\mathrm{B}^0 = \mu_0 \int_{V_\mathrm{c}} \mathrm{d}V \int_0^{H_\mathrm{S}} \delta H\, M(H) \approx \mu_0 V_\mathrm{c} M_\mathrm{S} H_\mathrm{S}. \tag{27}$$

Here V_c is the activation volume times a numerical factor between one and two, dependent of the micromagnetic model considered [18]. A comparison of Eqs. (25) and (27) leads to

$$V_c = \frac{k_B T}{\mu_0 S_V M_S}. \tag{28}$$

The latter are the basic assumptions for the so called activation volume approach. We remark that the logarithmic time derivative of the irreversible susceptibility equals the derivative of the viscosity with respect to the magnetic field, due to the theorem of Schwarz, i.e.

$$\frac{\partial \chi_{irr}}{\partial \ln t} = -\frac{\partial S}{\partial H}. \tag{29}$$

As Berkov [17] emphasized, the logarithmic behaviour does obviously not depend on the special form of the barrier distribution function, as long as the higher derivatives of the barrier distribution are small or, in other words, if the switching field distribution is not too narrow. But moreover, from Eqs. (10) and (23) we find that even in those cases, where the magnetization would not follow the ln t law, the relation between the viscosity and the irreversible susceptibility is valid if the condition $dE_B/dH \approx$ constant holds. For the given derivation all the volume elements are assumed as non-interacting. It is not yet clear, how an interaction between the grains will modify the form of Eqs. (18) and (25). One may argue that the interaction shifts the switching field of a volume element by the interaction field due to the neighbouring volume elements thereby transforming the original switching field distribution to an effective one. This effect is included in the derivation if one identifies H with the internal field, as was supposed here. Nevertheless, every volume element will experience a local field, which deviates from the mean field due to fluctuations, caused by the heterogeneity of its neighbourhood. If this distribution is known, one can generalize Eq. (5) by averaging with respect to the local field fluctuations. Attempts into this direction are contained in Ref. [10] as well. A complication arises, since even in the case where the original switching field of a grain is time independent, the effective switching field becomes time dependent, as it contains the local interaction field which is not constant in time. Therefore one should expect additional terms in the time dependence of the magnetization for interacting systems.

3. The texturized Stoner–Wohlfarth ensemble

3.1. The angular dependence of the energy barrier

In this section we will deal with the Stoner–Wohlfarth model [6], which allows to calculate the aftereffect without making assumptions about the field dependence on the barrier height, since we can calculate it exactly. For this we choose an ensemble of uniaxial particles each reversing magnetization by coherent rotation. The interaction between grains is neglected. This model was frequently employed to study fine particle systems [10,20,21]. We assume further all particles to have the same volume (in contrast to Refs. [8,9,20,21]) and the same anisotropy constant K_1. To the magnetic part of the energy of an oblique oriented particle,

$$E = 2VK_1 f(h, \vartheta, \varphi) \tag{30}$$

with

$$f(h, \vartheta, \varphi) = \tfrac{1}{2}\sin^2\varphi - h\cos(\vartheta - \varphi), \tag{31}$$

both the anisotropy and the field energy contribute. Here V is the volume of the grain or the volume element in which the magnetization coherently rotates and h is measured in units of the anisotropy field $H_A = 2K_1/M_S$. The angle between the field direction and the c-axis of the uniaxial crystallite is ϑ, the angle between the magnetization direction and field direction is φ. The minima and maxima of the energy in dependence on the field h and the angle ϑ are obtained from the first and second derivative of Eq. (31) with respect to φ. Below a critical field h_c, which depends on ϑ according to

$$h_c = (\cos^{2/3}\vartheta + \sin^{2/3}\vartheta)^{-3/2}, \tag{32}$$

two minima and two maxima exist, whereas above h_c only one. This means that for large enough negative fields a barrier does not exist, whereas below the critical field we have a barrier to overcome by thermal activation. If the easy axis of a grain lies in

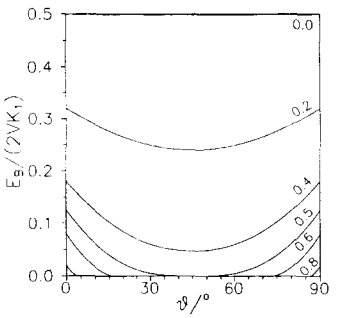

Fig. 1. Energy barrier height in dependence of angle ϑ for different values of the opposite field for the isotropic Stoner–Wohlfarth ensemble.

the direction of the applied field, one gets for the two energy barrier heights

$$E_{12} = E_M - E_1 = VK_1(1-h)^2, \tag{33a}$$

$$E_{21} = E_M - E_2 = VK_1(1+h)^2. \tag{33b}$$

For an arbitrary angle ϑ the minima and maxima position and the related barrier heights have to be calculated numerically. Fig. 1 shows the barrier height E_{12} in dependence on the angle ϑ for different applied fields. There is a significant difference between the results shown in Fig. 2 of Ref. [10] and our results. We do not find five different types for the dependencies of $E_B(\vartheta)$ for H between 0 and H_A but only two, one for $H < H_A/2$ and the other for $H > H_A/2$. We believe that the wrong results in Ref. [10] are due to the approximative form of the barrier height in dependence of the misalignment angle. We believe that a grain magnetized opposite to the field direction has the highest barrier as shown in our calculation, and not a zero barrier as given in Ref. [10].

3.2. The energy barrier distribution

From Eq. (33) and Fig. 1 it is evident that the barrier heights are not linearly dependent on the applied field, in contrast to the assumption necessary to get the commonly used Eq. (25). If the grains are ideally aligned, the barrier distribution in Eq. (5) is δ-like, resulting in a time behaviour proportional to $p(t)$ from Eq. (2). Otherwise, if the easy axes are distributed according to a texture function, one can calculate the barrier distribution from the results shown in Fig. 1 using the texture function. Since the details of the fiber texture function are of less importance, we use in the following the simple rotational-symmetric function [22]

$$f(\vartheta) = (2n+1)\cos^{2n}\vartheta, \tag{34}$$

where the angle ϑ is between the texture axis and the c-axis of the individual grains. Both the applied field and the texture axis should be parallel to the z-axis. Using the angle dependence of the barrier height together with the texture function, we are able to calculate the barrier distribution function for different fields and texture parameters. As an example we show the distribution of the barrier E_{12} for different magnetic fields for the isotropic sample ($n = 0$) in Fig. 2.

3.3. Magnetization and viscosity

If all grains of the ensemble are in the metastable state at $t = 0$, we get the averaged normalized magnetization parallel to the field from

$$\overline{m} = \langle M(t)/M_S \rangle = \langle \cos(\vartheta - \varphi_1)\, p(t) \rangle + \langle \cos(\vartheta - \varphi_2)(1 - p(t)) \rangle. \tag{35}$$

Here φ_1 and φ_2 denote the angles between the magnetization and the c-axis of the metastable and stable state respectively, and $p(t)$ is the time dependent probability to find a grain in a metastable state, according to Eq. (2). The brackets symbolize an average, which has to be calculated by integration with respect to the texture function:

$$\langle \cdots \rangle = \int_0^{\pi/2} \mathrm{d}\vartheta \, \sin\vartheta \, f(\vartheta) \cdots. \tag{36}$$

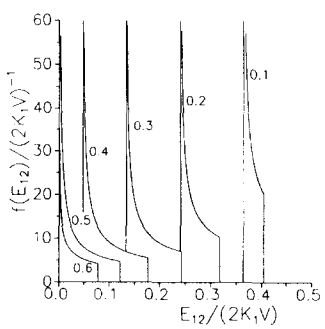

Fig. 2. Energy barrier distribution E_{12} of the isotropic ensemble for different opposite magnetic fields (parameter on the curves: $h = H/H_A$).

To get the full time dependence of the magnetization it is necessary to make an assumption about the parameter E_{12}/k_BT. To have a remarkable effect during experiments lasting typically from 1 to 10^4 s this parameter should lie between 25 and 33 [18]. In Figs. 1 and 2 the barrier height is measured in units of $2VK_1$. Therefore the value $25k_BT/2VK_1$ should be between 0 and 0.5. We choose $k_BT/2VK_1$ to be 0.001. This is essentially a choice of an effective particle volume V. For example for Ba-ferrite one gets 22 nm diameter, for $SmCo_5$ 8 nm, and for the other hard magnetic materials a value in between, which is of the order of the Bloch-wall thickness, instead of the order of the grain volumes of sintered magnets.

3.4. Susceptibilities and fluctuation field

Due to the differences of the presumptions of the model under consideration and the theory of section 2 yielding Eq. (25) we cannot expect S_V to be constant a priori. Therefore we computed the total and irreversible susceptibility, and the aftereffect constant in their dependence on field and time. The total susceptibility χ_{tot} we get by simply differentiating Eq. (35) with respect to H thereby taking the waiting time as constant. Concerning the irreversible susceptibility, we find in the literature two different methods to measure it. The first way is to differentiate the remanence curve [23]. For the model under consideration one has to evaluate

$$\frac{\partial m_R}{\partial h} = \left\langle \cos(\vartheta - \pi)\, \frac{\partial}{\partial h} p(t, h) \right\rangle + \left\langle \cos(\vartheta)\, \frac{\partial}{\partial h} (1 - p(t, h)) \right\rangle. \quad (37)$$

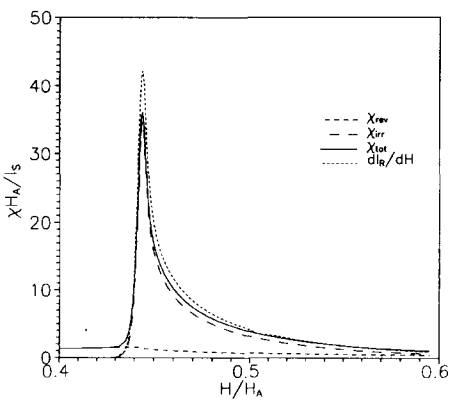

Fig. 3. Reversible, irreversible and total susceptibility for the isotropic sample for a waiting time of 1 s. The differentiated remanence for the same system is also shown.

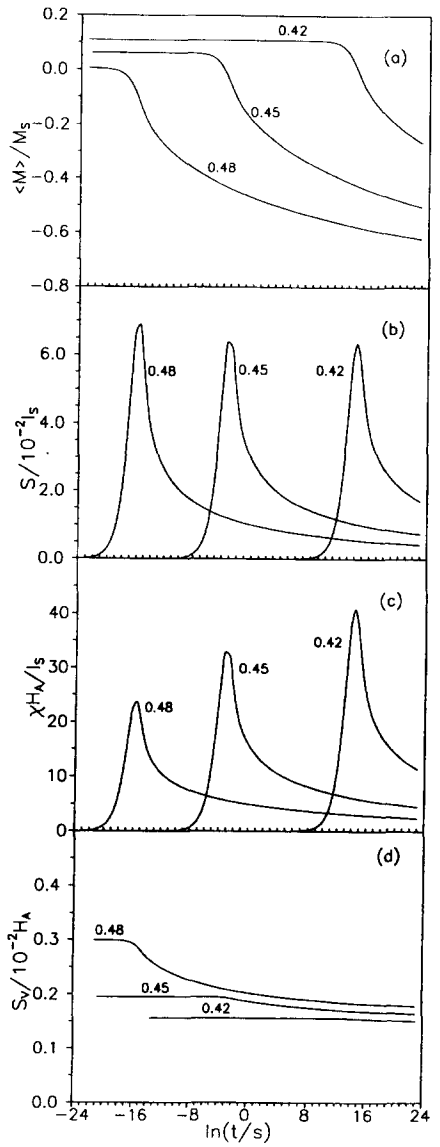

Fig. 4. Time dependence of the magnetization, viscosity, irreversible susceptibility and fluctuation field for the isotropic ensemble. Parameters on the curves: applied opposite field.

For $t = 0$ this can be calculated analytically [24], yielding

$$\frac{\partial m_R}{\partial h} = \Theta(h - 0.5)\ \Theta(1 - h) \times \sum_{i=1,2} \cos(\vartheta_i)\ \sin(\vartheta_i)\ f(\vartheta_i) \times \frac{\left(\cos^{2/3}\vartheta_i + \sin^{2/3}\vartheta_i\right)^{5/2}}{\left|\cos^{-1/3}\sin\vartheta_i - \sin^{-1/3}\cos\vartheta_i\right|}. \tag{38}$$

Here the two angles ϑ_i are determined by the intersection of the curve $h_c = h$ and the curve given by Eq. (32), i.e.

$$\vartheta_{1,2} = \arccos\sqrt{\frac{1}{2}\left(1 \pm \sqrt{1 - \frac{4}{27}\frac{(1-h^2)^3}{h^4}}\right)}. \tag{39}$$

For finite times the calculation has to be done numerically.

The second way to get the irreversible susceptibility is to measure the difference between the total and the reversible change of magnetization [25]. For the considered model one gets the reversible susceptibility by freezing the state of each grain, which it has, if a field was applied for a time t. Then one has to look how the magnetization changes due to the reversible rotation process, i.e.

$$\chi_{rev} = \left\langle p(h, t)\frac{\partial}{\partial h}\cos(\vartheta - \varphi_1)\right\rangle + \left\langle (1 - p(h, t))\frac{\partial}{\partial h}\cos(\vartheta - \varphi_2)\right\rangle. \tag{40}$$

The so defined irreversible susceptibility, i.e. $\chi_{irr} = \chi_{tot} - \chi_{rev}$, is obviously different from the differentiated remanence curve. In Fig. 3 we show the numerically calculated field dependence of the two differently defined irreversible susceptibilities together with the reversible and total ones. Fig. 4 shows the time dependence of the magnetization, the viscosity, irreversible susceptibility and fluctuating field for an isotropic ensemble for different magnetic fields in the vicinity of the coercivity field of about $h = 0.48$. The magnetization does not follow the ln t law. The peaks of the viscosity and the irreversible susceptibility cancel each other giving a nearly time independent value of the fluctuation field S_V. The same quantities are depicted for different waiting times in Fig. 5, and in Fig. 6 the dependence on the texture parameter n is plotted for the same quantities. The fluctation field is independent on the degree of alignment showing that the peak of $\Gamma t \exp(-\Gamma t)$ is sharp enough for the higher derivatives of dE_B/dH not to contribute.

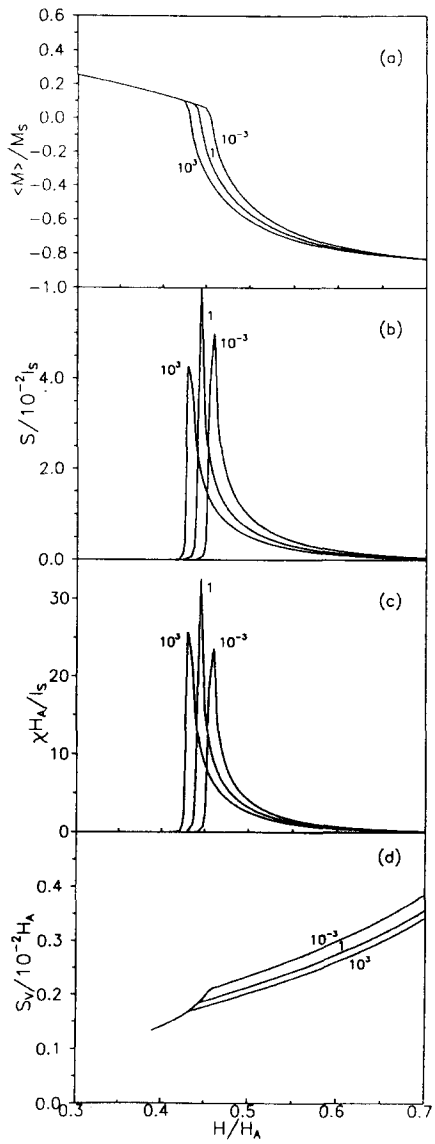

Fig. 5. Field dependence of the magnetization, viscosity, irreversible susceptibility and fluctuation field for waiting times of 10^{-3}, 1 and 10^3 s.

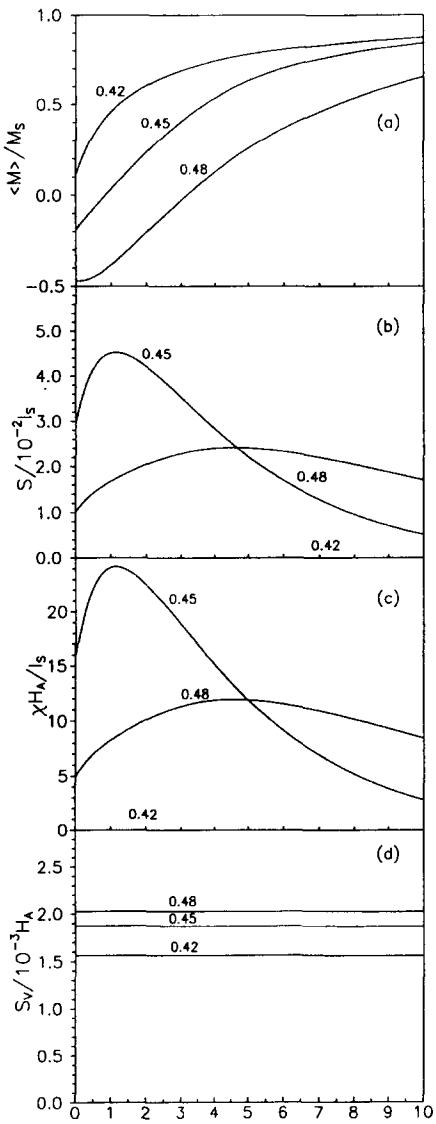

Fig. 6. Dependence of magnetization, viscosity, irreversible susceptibility and fluctuation field on the texture parameter n for a waiting time of 1 s. Parameters on the curves: applied opposite field.

4. The texturized Kondorsky ensemble

4.1. Magnetization and viscosity

In contrast to the Stoner–Wohlfarth model, where the angular dependence of the switching field is given by Eq. (32), in the following we will discuss the case where the angular dependence of the switching field exhibits a $1/\cos\vartheta$ law [12]. For the models of both weak and strong domain wall pinning as well as for the nucleation models [18,26] one has a linear dependence of the barrier height E_B on the applied field, according to Eq. (26). Assuming E_B^0 and H_S to be determined by intrinsic material parameters, the main contribution to the angular dependence is due to the fact that only the component parallel to the easy axis causes irreversible changes. We neglect the lowering of the domain wall energy due to rotation processes. For the barrier height follows

$$E_B(\cos\vartheta) = E_B^0\left(1 - \frac{H\cos\vartheta}{H_S}\right). \tag{41}$$

We get the viscosity from

$$S = -\frac{\partial\overline{m}(H,t)}{\partial \ln t} = 2\int_0^{\pi/2} \mathrm{d}\vartheta \sin\vartheta\cos\vartheta\, f(\cos\vartheta)\,\Gamma t\, \mathrm{e}^{-\Gamma t}, \tag{42}$$

where $\overline{m}$ is calculated analogous to Eq. (35), but with $\varphi_1 = \pi$ and $\varphi_2 = 0$, and $f(\cos\vartheta)$ is the texture function Eq. (34). Solving Eq. (41) for $\cos\vartheta$ and changing the integration variable to E_B yields

$$S = \frac{2H_S^2}{H^2E_B^0}\int_{E_B^0(1-H/H_S)}^{E_B^0} \mathrm{d}E_B\, f(E_B) \times\left(1 - E_B/E_B^0\right)\Gamma t\, \mathrm{e}^{-\Gamma t}. \tag{43}$$

If the maximum of $\Gamma t\exp(-\Gamma t)$ is outside the limits of the integral, S will be small. If it is inside, from a Taylor series expansion results

$$S \approx \frac{2H_S^2}{H^2E_B^0} f(E_{B,\max})\left(1 - E_{B,\max}/E_B^0\right) \times\int_{E_B^0(1-H/H_S)}^{E_B^0} \mathrm{d}E_B\, \Gamma t\, \mathrm{e}^{-\Gamma t}, \tag{44}$$

with $E_{B,\max}$ according to Eq. (11).

4.2. Irreversible susceptibility and fluctuation field

For the irreversible susceptibility we get

$$\chi_{\mathrm{irr}} = \frac{\partial\overline{m}}{\partial H} = -2\frac{E_B^0}{H_S k_B T}\int_0^{\pi/2} \mathrm{d}\vartheta \sin\vartheta\cos^2\vartheta \times f(\cos\vartheta)\,\Gamma t\, \mathrm{e}^{-\Gamma t}, \tag{45}$$

where we used

$$\frac{\partial\Gamma}{\partial H} = \frac{\Gamma E_B^0}{k_B T H_S}\cos\vartheta. \tag{46}$$

Changing to an energy integration yields

$$\chi_{\mathrm{irr}} = \frac{-2H_{\mathrm{S}}^2}{H^3 k_{\mathrm{B}} T} \int_{E_{\mathrm{B}}^0(1-H/H_{\mathrm{S}})}^{E_{\mathrm{B}}^0} \mathrm{d}E_{\mathrm{B}}\, f(E_{\mathrm{B}}) \times \left(1 - E_{\mathrm{B}}/E_{\mathrm{B}}^0\right)^2 \Gamma t\, \mathrm{e}^{-\Gamma t}. \tag{47}$$

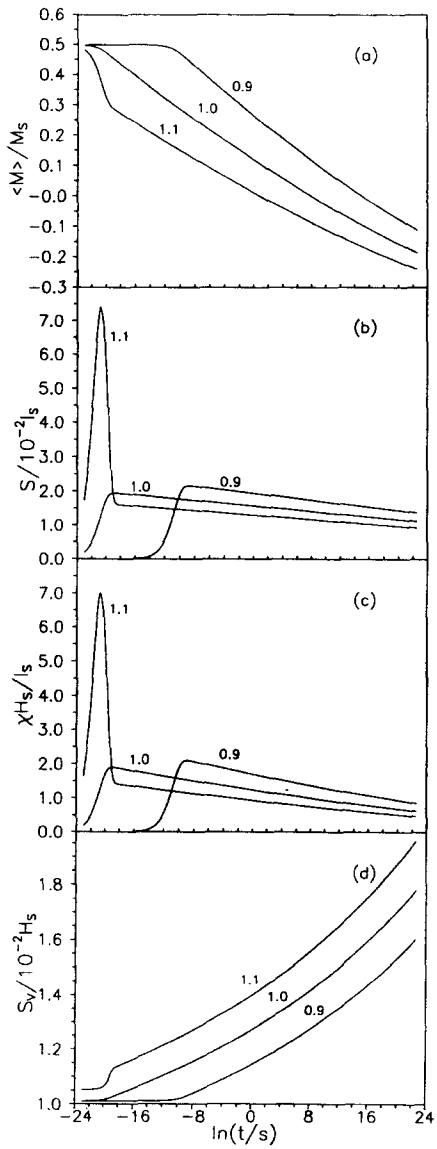

Fig. 7. Time dependence of the magnetization, viscosity, irreversible susceptibility and fluctuation field for an isotropic ensemble of grains with $1/\cos\vartheta$ angular dependence of the switching field H_{S} with $E_0/k_{\mathrm{B}}T = 100$, $\Gamma_0 = 10^{-9}$ s^{-1}. Parameter on the curves: applied opposite field $h = H/H_{\mathrm{S}}$.

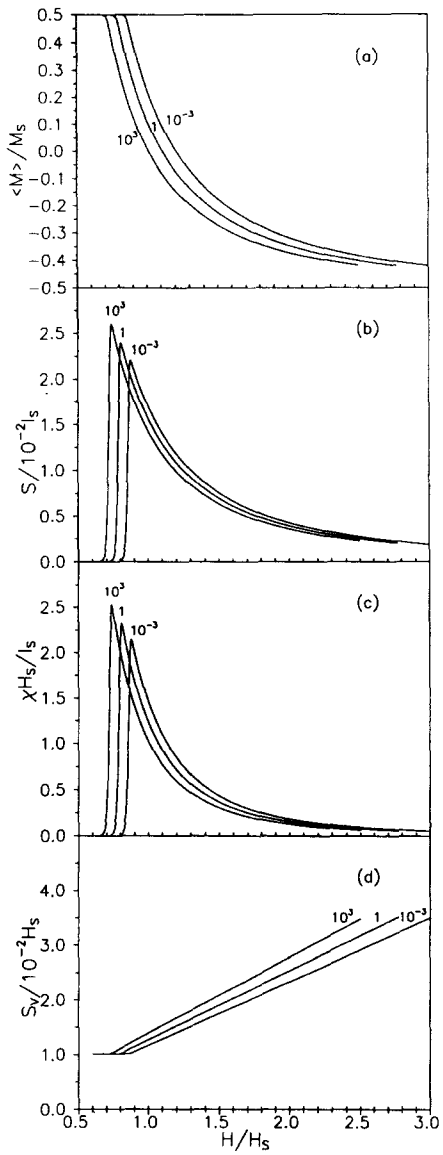

Fig. 8. Field dependence of the magnetization, viscosity, irreversible susceptibility and fluctuation field for the same ensemble as Fig. 7. Parameter on the curve: waiting time (in s).

With the same condition as cited above we get, as the leading term of a Taylor expansion,

$$\chi_{\mathrm{irr}} \approx \frac{-2H_{\mathrm{S}}^2}{H^3 k_{\mathrm{B}} T} f(E_{\mathrm{B,max}}) \left(1 - E_{\mathrm{B,max}}/E_{\mathrm{B}}^0\right)^2 \times \int_{E_{\mathrm{B}}^0(1-H/H_{\mathrm{S}})}^{E_{\mathrm{B}}^0} \mathrm{d}E\, \Gamma t\, \mathrm{e}^{\Gamma t}. \tag{48}$$

Whereas the viscosity and the irreversible susceptibility depend on the texture function, the quotient of Eq. (44) by Eq. (48),

$$S_V = \frac{-Hk_BT}{E_B^0 - E_{B,max}}, \tag{49}$$

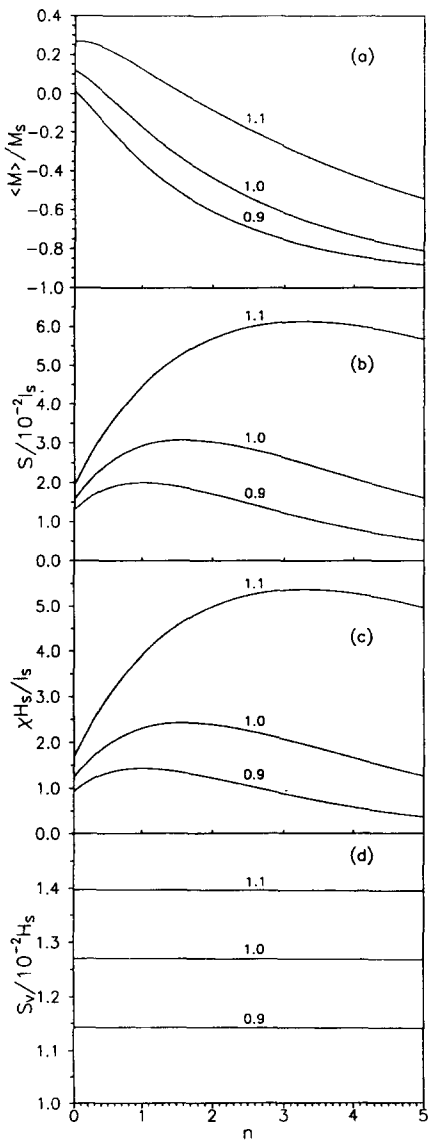

Fig. 9. The dependence of the magnetization, viscosity, irreversible susceptibility and fluctuation field on the alignment parameter n ($I_R/I_S = (2n+1)/(2n+2)$) for an ensemble of grains with $1/\cos\vartheta$ angular dependence of the switching field H_S with $E_0/k_BT = 100$, $\Gamma_0 = 10^{-9}$ s^{-1}. Parameters on the curves: applied opposite field $h = H/H_S$. The waiting time is 1 s.

does not depend on the texture function. Eq. (49) shows that S_V should be linearly dependent on the applied field if E_B^0 and Γ_0 are field independent. The temperature dependence is more complicated, since $E_{B,max}$ is linearly dependent on the temperature and also E_B^0 is expected to decrease with increasing temperature. The typical measuring times are between one and some thousands seconds which means that the energy $E_{B,max}$ is between $25k_BT$ and $33k_BT$. Therefore the barrier E_B^0 has to be larger than $\sim 35k_BT$, while here $k_BT/E_B^0 = 0.01$ was chosen, whereas the magnetic field should be larger than half of the switching field. If we do not restrict the calculation to that interval, the calculation of S, χ_{irr} and S_V has to be done numerically, and indeed, we calculated the following pictures from Eqs. (42) and (45) directly. Fig. 7 shows the overall time dependence of the magnetization, viscosity irreversible susceptibility, and fluctuation field. The field dependence of the same quantities for waiting times of 10^{-3}, 1 and 10^{+3} s is plotted in Fig. 8, and finally in Fig. 9 the dependence on the texture parameter n is given.

5. Discussion

The main topic of this paper is to calculate the influence of a distribution of easy axes on the time dependence of the magnetization, the magnetic viscosity, the susceptibilities and the fluctuation field using well defined assumptions on the intrinsic parameters of the models. Since our results were calculated without using an energy barrier distribution being the main entity to the 'standard theory', it is very useful to compare the given theory with the usual way to get the magnetic viscosity. This comparison is possible, since both investigated models, i.e. the texturized Stoner–Wohlfarth and Kondorsky ensemble respectively, allow to calculate the barrier distribution from the texture function.

In Section 3 we dealt with Stoner–Wohlfarth ensembles with different degrees of alignment. For a random oriented ensemble the angle dependence of the energy barrier and the barrier distribution was first given by Berkov [17]. We found his results to be incorrect. Our numerically calculated barrier distribution has only one singularity according to the energy

barrier minimum at 45°. Fig. 2 shows that even for a random ensemble the assumption of a nearly constant barrier distribution in the vicinity of $E_{B,max}$, necessary to evaluate the integrals in the 'standard theory', is violated. We found that the barrier distributions, which result from different degrees of misalignment, have always sharp edges and the slope in between is not small. Accordingly the computed time dependence of the magnetization is not linearly decreasing with ln t. In contrast to our model, where we used a δ-like distribution for the critical volumes and where the barrier distribution is due to a texture, in Ref. [7] an ideally aligned ensemble, where the barrier distribution is due to a distribution of particle volumes, was considered. There was shown that a small distribution width results in deviations from the ln t behaviour. The reason is again that the assumption of a constant barrier distribution in the vicinity of $E_{B,max}$ is violated. Taking into account higher order derivatives of the barrier distributions, the approximative evaluation of the integrals can be improved, giving leading corrections to the ln t law [7]. We did not follow this line, instead we calculated the integrals over the misalign angle numerically. In the Stoner–Wohlfarth model it is also possible to calculate the barrier height for backjumps. Taking them into account, it can be shown straightforwardly from Eqs. (2) and (6) that the magnetization for long waiting times approaches the anhysteretic curve, according to what one should expect. Furthermore, since that model contains both reversible and irreversible processes, it was possible to demonstrate the difference between the irreversible susceptibility, which is dependent on the applied field and the waiting time, and the differentiated remanence curve. From Eq. (23) follows that the susceptibility, which is used to determine the fluctuation field, has to be measured at the same field and the same waiting time at which the viscosity was determined. For this purpose the differentiated remanence curve seems less suited, since the remanence is dependent on the time the opposite field was applied before switching it off. Nevertheless our results show that the difference is not too big, so that it may be neglected in experiments.

For the models with Kondorsky-type angular dependence treated in section 4 the time behaviour depends mainly on the parameter E_B^0/k_BT. We used for our computation the value 100. In that case the texture functions under consideration are much wider than the peak of the function $\Gamma t \exp(\Gamma t)$. The numerical results shown in Figs. 7, 8 and 9 are therefore conform with Eq. (49). We find the ln t law nearly fulfilled if the applied field is in the region of the switching field H_S or larger. The shift of the magnetization curve to lower fields with increasing waiting time is in good qualitative agreement with experiments which measured the sweep rate dependence of the hysteresis curves in $SmCo_{3.5}Cu_{1.5}$ [27]. It is surprising that the texture dependence of the viscosity and the irreversible susceptibility cancel out, giving a texture independent fluctuation field S_V. This is in good agreement with the experimental results given in Refs. [13,14]. Our results are not related to the special form of the texture function, e.g. a Gaussian gives the same results [24]. This can be seen immediately from the derivation of Eq. (49), where it is not necessary to specify the form of the texture function at all. We regard that the discrepancy between the experiment and the theoretical considerations in Ref. [13] is due to the fact that in Ref. [13] S_V was averaged. Instead one has to average S and χ_{irr} separately.

Since we used models of identical particles, i.e. without any distribution of switching fields or activation volumes etc., the texture influence on the fluctuation field S_V should be stronger than in every more realistic model, i.e. when distributions of other intrinsic parameters are taken into account. Even for that case we found no dependence of S_V on the texture parameter n. Nevertheless, for applications not S_V but S itself is of more interest. This quantity shows a texture dependence in both models if the applied opposite field is in the vicinity of the coercivity field. The flat maxima in Figs. 6 and 9 will be smeared out if one takes into account distributions of other parameters. Therefore our main conclusion is that there is no chance to change the magnetic aftereffect significantly by manipulation of the texture.

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